ZAKLJUCNO POROCILO
O REZULTATIH OPRAVLJENEGA RAZISKOVALNEGA DELA NA PROJEKTU V OKVIRU CILJNEGA RAZISKOVALNEGA PROGRAMA (CRP) »KONKURENCNOST SLOVENIJE 2006 – 2013«

I. Predstavitev osnovnih podatkov raziskovalnega projekta

1. Naziv težišca v okviru CRP:

5.8.3. Nadzor radioaktivnosti v življenjskem okolju

2. Šifra projekta:

V2-0555

3. Naslov projekta:

Ugotavljanje razmerja med 129-I in 127-I v morskem in kopenskem okolju na obmocju Slovenije

3. Naslov projekta

3.1. Naslov projekta v slovenskem jeziku:

Ugotavljanje razmerja med 129-I in 127-I v morskem in kopenskem okolju na obmocju Slovenije

3.2. Naslov projekta v angleškem jeziku:

Establishment of the ratio between 129-I and 127-I in marine and terrestial enviroment in Slovenia

4. Kljucne besede projekta

4.1. Kljucne besede projekta v slovenskem jeziku:

I-129, I-127, alge, školjke, padavine, zemlja, smrekove iglice

4.2. Kljucne besede projekta v angleškem jeziku:

iodine-129, iodine-127, seawater, alga, mussels, precipitation, soil, pine needles

5. Naziv nosilne raziskovalne organizacije:

Institut "Jožef Stefan"

5.1. Seznam sodelujocih raziskovalnih organizacij (RO):

6. Sofinancer/sofinancerji:

Ministrstvo za okolje in prostor, Uprava za jedrsko varnost Republike Slovenije, Železna 16, 1000 Ljubljana

7. Šifra ter ime in priimek vodje projekta:

1873	Vekoslava Stibilj

Datum: 15.9.2010

Podpis vodje projekta:	Podpis in žig izvajalca:
izr.prof. dr. Vekoslava Stibilj	prof.dr. Jadran Lenarcic

II. Vsebinska struktura zakljucnega porocila o rezultatih raziskovalnega projekta v okviru CRP

1. Cilji projekta:

1.1. Ali so bili cilji projekta doseženi?

- [x] a) v celoti
- [ ] b) delno
- [ ] c) ne

Ce b) in c), je potrebna utemeljitev.

1.2. Ali so se cilji projekta med raziskavo spremenili?

- [ ] a) da
- [x] b) ne

Ce so se, je potrebna utemeljitev:

2. Vsebinsko porocilo o realizaciji predloženega programa dela[1]:

1. Vzorcenje

Casovni nacrt vzorcenja ter potek analiz so natancneje predstavljeni v preglednici 1 (Priloga I).

1.1. Padavine, tla in smrekove iglice

Opravili smo vsa štiri kontinuirana vzorcenja padavin (Bovec, Bilje, Ljubljana in Iskrba - Kocevsak Reka) v obdobju od 1.2 do 30.4.2009, od 1.5 do 31.07.2009, od 1.8 do 31.10.2009 ter od 1.11.2009 do 31.1.2010.

Opravili smo vsa tri vzorcenje tal in smrekovih iglic (Bovec, Bilje, Ljubljana, Iskrba - Kocevska Reka). Tla smo na vsaki lokaciji vzorcili na odprtem obmocju ter v gozdu oz. pod smreko.

1.2 Morska voda, sediment, alge in školjke

Opravili smo vse tri serije vzorcenj morske vode (Portorož, Žusterna in Debeli Rtic).

Opravili smo vse tri serije vzorcenj sedimenta na izbranih lokacijah v Piranskem in Koprskem zalivu.

Opravili smo vsa tri vzorcenja alg (Portorož, Žusterna in Debili Rtic). Pri tretjem vzorcenju alg na lokaciji Debeli Rtic nismo uspeli najti alg zaradi zmanjšane vegetacije v zimskem obdobju.

Opravili smo vsa tri vzorcenja školjk (Koper, Piran-Seca in Strunjan). Ker mase školjk vzorcenih 10.7.09 ni bilo dovolj za ugotavljanje joda-129, smo vzorcili školjke 10.3.2010 na vseh treh mestih.

Zbrane vzorce smo sproti pripravili (sušenje oz. liofilizacija ter homogenizacija - mletje in sejanje) za dolocitev vsebnosti I-129 in I-127.

2 Vsebnost I-129 in I-127 v vzorcenih okoljskih vzorcih

2.1 Kopensko okolje

2.1.1 Padavine

Ugotavljali smo jod v padavinah v Bovcu, Biljah, Iskrbi in Ljubljani v letu 2009. Koncentracije I-129 so v obmocju manj kot 0,5 do 18,2 • 10E-10 µg/g (manj kot 2,2–120,8 • 10-07 Bq kg-

[1] Potrebno je napisati vsebinsko raziskovalno porocilo, kjer mora biti na kratko predstavljen program dela z raziskovalno hipotezo in metodološko-teoreticen opis raziskovanja pri njenem preverjanju ali zavracanju vkljucno s pridobljenimi rezultati projekta.

1) ter I-127 v obmocju od 1,7 do 6,5 ng/g (Priloga II, preglednica 1).

Kolicina padavin zbranih v Biljah (< 9 L) v prvem trimesecju ni zadostovala za dolocitev I-129 z razvito metodo. Prav tako je bila kolicina padavin zbranih v Bovcu in Biljah v drugem, tretjem in cetrtem trimesecju manjša od 10 L, zato prekoncentracije in radiokemijske separacije joda-129 nismo naredili. Poskusali bomo jod 129 ugotavljati z AMS v okviru strokovnega izpopolnjevanja na Japonskem (glej tocko 4.1) .

Tudi literatura (Hou in sod., 2009) navaja podatke za I-127 v padavinah v podobnem intervalu od 0,78 do 2,7 ng/g ter izotopsko razmerje I-129 /I-127 med 5,04 in 76,5 • 10E-08 s srednjo vrednostjo (30,1 ± 16,8) • 10E-08 v obdobju od 2001 do 2006. Padavine so vzorcili v razlicnih krajih na Danskem. Podobne vrednosti navajajo tudi Buraglio in sod. (2001) za padavine vzorcene na razlicnih lokacijah na Švedskem v obdobju od 1998 do 1999 (Priloga III, preglednica 1).

2.1.2 Tla

Analizirali smo vzorce tal vzorcenih v Bovcu, Biljah, Iskrbi (Kocevska Reka) in Dolu (Reaktor). Zemlja, vzorcena v Bovcu vsebuje najvec joda-129. Iz preglednice 2 (Priloga II) je razvidno, da je koncentracija I-129 v vzorcu tal vzorcenih v Bovcu 29.1.2009 izpod smreke (114 ± 18) • 10E-08 µg/g za en krat višja od koncentracije I-129 v vzorcu tal vzorcenem na prostem (56 ± 10) • 10E-08 µg/g. Enak trend smo opazili tudi pri tretjem vzorcenju. Tudi koncentracija I-127 na prostem (22,2 ± 0,8) µg/g je manjša kot pod smreko (26,6 ± 0,6) µg/g. Izjema je drugo vzorcenje v Bovcu, kjer je vsebnost joda 129 enaka na prostem kot pod smreko. Tla vzorcena v Biljah in Iskrbi in Dolu so vsebovala nekoliko vec joda-129 pod smreko kot na prostem.

Podobne vrednosti za tla so navedene v literaturi (Priloga III, Preglednica 2) za obomcja na Švedskem in Danskem, medtem ko je vsebnost I-129 v tleh na obmocju Japonske tudi do en velikostni razred nižja.

2.1.3 Smrekove iglice

Analizirali smo vzorce smrekovih iglic, vzorcenih v Bovcu, Biljah, Iskrbi in Dolu. Dolocili smo koncentracije I-129 v obmocju manj kot 5,0 do 15 • 10E-08 µg/g (3,9–10 • 10-04 Bq/kg) ter I-127 v obmocju od 81 do 413 ng/g (Priloga II, preglednica 3).

Smrekove iglice so delni pokazatelj atmosferskih depozicij I-129. Quintana in Thyssen (2000) sta dolocila vsebnost I-129 v iglicah pinij in ceder v okolici jedrskih elektrarn Atucha in Embalse ter blizu atomskega centra Ezeiza v Argentini. Dolocila sta dokaj visoke aktivnosti I-129, in sicer za elektrarno Atucha 4000 in 4700 • 10E-04 Bq/kg, za Embalese 2000 in 35500 • 10E-04 Bq/kg ter za Ezeiza 730 • 10E-04 Bq/kg. Vsebnosti stabilnega I-127 ne navajata (Priloga III,Preglednica 3.

2.2 Morsko okolje

2.2.1 Morska voda

Ugotovili smo, da vsebnost I-129 v morski vodi zelo niha (Priloga II, preglednica 4). V morski vodi vzorceni v Žusterni novembra 2008 smo v povprecju dolocili (1,9 ± 0,24) • 10E-04 Bq I-129/kg morske vode. Vsebnost I-129 v morski vodi vzorceni 1.4.2009 na isti lokaciji (Žusterna) ter v Portorožu je bila pod mejo zaznavnosti metode. Za lokacijo Debeli Rtic smo dolocili aktivnost (0,1 ± 0,01) • 10E-04 Bq I-129/kg morske vode, kar je za en velikostni razred manj kot v morski vodi vzorceni novembra 2008. Koncentracije I-127 so primerljive z vrednostmi dolocenimi za morsko vodo vzorceno novembra 2008. V vseh primerih smo odmerili 4 L vzorca. Pri izracunu vsebnosti I-129 smo upoštevali gostoto.

Mejo zaznavnosti metode smo izboljšali s povecanjem volumna vzorca na 8 L. S postopkom izhlapevanje ni možno izolirati joda iz vecjega volumna (vec kot 8 L) vzorca, saj je ionska moc dobljenega koncentrata morske vode previsoka za uspešno solventno ekstrakcijo joda. Tako smo uspešno dolocili vsebnost I-129 v morski vodi vzorceni 15.7 in 26.11.2009. Dolocene koncentracije I-129 (5,5–13,4 • 21E-10) so v enakem velikostnem razredu kot pri morski vodi vzorceni v Debelem Rticu 1.4.2009 (Priloga II, preglednica 4).

Dolocene koncentracije I-129 v morski vodi vzorceni v Sloveniji so za en velikostni razred višje kot na obmocju Grenlandije (vzorceno 1999, Priloga III, preglednica 4), za tri velikostne razrede višje kot na obmocju Japonske ter za dva velikostna razreda nižje kot na obmocju Rokavskega preliva in Severnega morja, ki je pod vplivom direktnih izpustov I-129 iz La Haguea in Sellafielda. Koncentracije I-127 so v enakem obmocju.

2.2.2 Morski sediment

Analizirali smo vzorce morskega sedimenta vzorcene v Kopru, Piranu in Strunjanu . 5). Dolocene koncentracije I-129 so v obmocju manj kot 5,0 do 17 • E-08 µg/g ter I-127 od 79,1 do 110,7 µg/g (Priloga II, preglednica 5).

2.2.3 Alge

Analizirali smo vzorce morskih alg (Jadranski bracic, Fucus virsoides) vzorcenih v Portorožu, Žusterni Debelem rticu in sicer spomladi, poleti in pozni jeseni. Jadranskega bracica v mesecu novembru na Debelem rticu nismo našli zadostno kolicino. Koncentracija I-129 in I-127 v analiziranih morskih algah (Portorož, Žusterna in Debeli Rtic, Priloga II, preglednica 6) je primerljiva z vrednostmi za morske alge vzorcene na istih lokacijah septembra 2005 (Priloga II, preglednica 6).

Preglednica 5 (Priloga III) prikazuje primerjavo vsebnosti I-129 v morskih algah z razlicnih lokacij po svetu. Izotopsko razmerje za Jadransko morje dosega vrednosti vse do 10E-09, kar je za en velikostni razred vec kot vzdolž kitajske obale (Hou in sod., 2000), do dva velikostna razreda manj kot v Baltskem morju (Hou in sod., 1999), ki je pod vplivom neposrednih tekocih izpustov iz La Haguea in Sellafielda, in do štirih velikostnih razredov manj kot na obmocju predelovalnega obrata La Haguea (Fréchou in sod., 2003). Iz preglednice 5 je razvidno, da se je izotopsko razmerje I-129/I-127 v Fucus vesicolosus na obmocju Baltskega morja od leta 1989 do 1998 povecalo s ~400 na ~3400 • 10-E10, kar je skoraj za faktor deset (Hou in sod., 1999 in 2000a).

2.2.4 Školjke

Dolocili smo koncentracijo I-129 in I-127 v školjkah (sredozemska klapavica, Mytilus gallprovincialis,Priloga II, preglednica 7). Dolocene koncentracije I-129 so v obmocju od 4,2 do 93 • 10E-08 µg/g (7–62 • 10E-04 Bq/kg) ter I-127 v obmocju od 6,9 do 18,3 µg/g, vse izraženo na suho snov. V literaturi nismo našli podatkov za vsebnost I-129 in I-127 v školjkah.

Zakljucek

Podatki za vsebnost joda -129 v kopenskem okolju (padavinah, zemlji, smrekovih iglicah) ter v morskem okolju (morska voda, školjke in alge) prispevajo k vedenju o kroženju joda-129 v naravi in so hkrati prvi za južni del Evrope.

3. Izkoriščanje dobljenih rezultatov:

3.1. Kakšen je potencialni pomen[2] rezultatov vašega raziskovalnega projekta za:

- ☒ a) odkritje novih znanstvenih spoznanj;
- ☒ b) izpopolnitev oziroma razširitev metodološkega instrumentarija;
- ☒ c) razvoj svojega temeljnega raziskovanja;
- ☐ d) razvoj drugih temeljnih znanosti;
- ☐ e) razvoj novih tehnologij in drugih razvojnih raziskav.

3.2. Oznacite s katerimi družbeno-ekonomskimi cilji (po metodologiji OECD-ja) sovpadajo rezultati vašega raziskovalnega projekta:

☐ a) razvoj kmetijstva, gozdarstva in ribolova - Vkljucuje RR, ki je v osnovi namenjen razvoju in podpori teh dejavnosti;

☐ b) pospeševanje industrijskega razvoja - vkljucuje RR, ki v osnovi podpira razvoj industrije, vkljucno s proizvodnjo, gradbeništvom, prodajo na debelo in drobno, restavracijami in hoteli, bancništvom, zavarovalnicami in drugimi gospodarskimi dejavnostmi;

☐ c) proizvodnja in racionalna izraba energije - vkljucuje RR-dejavnosti, ki so v funkciji dobave, proizvodnje, hranjenja in distribucije vseh oblik energije. V to skupino je treba vkljuciti tudi RR vodnih virov in nuklearne energije;

☐ d) razvoj infrastrukture - Ta skupina vkljucuje dve podskupini:

- transport in telekomunikacije - Vkljucen je RR, ki je usmerjen v izboljšavo in povecanje varnosti prometnih sistemov, vkljucno z varnostjo v prometu;
- prostorsko planiranje mest in podeželja - Vkljucen je RR, ki se nanaša na skupno nacrtovanje mest in podeželja, boljše pogoje bivanja in izboljšave v okolju;

☒ e) nadzor in skrb za okolje - Vkljucuje RR, ki je usmerjen v ohranjevanje fizicnega okolja. Zajema onesnaževanje zraka, voda, zemlje in spodnjih slojev, onesnaženje zaradi hrupa, odlaganja trdnih odpadkov in sevanja. Razdeljen je v dve skupini:

☒ f) zdravstveno varstvo (z izjemo onesnaževanja) - Vkljucuje RR - programe, ki so usmerjeni v varstvo in izboljšanje clovekovega zdravja;

☐ g) družbeni razvoj in storitve - Vkljucuje RR, ki se nanaša na družbene in kulturne probleme;

☒ h) splošni napredek znanja - Ta skupina zajema RR, ki prispeva k splošnemu napredku znanja in ga ne moremo pripisati dolocenim ciljem;

☐ i) obramba - Vkljucuje RR, ki se v osnovi izvaja v vojaške namene, ne glede na njegovo vsebino, ali na možnost posredne civilne uporabe. Vkljucuje tudi varstvo (obrambo) pred naravnimi nesrecami.

[2] Oznacite lahko vec odgovorov.

3.3. Kateri so **neposredni rezultati** vašega raziskovalnega projekta glede na zgoraj oznacen potencialni pomen in razvojne cilje?

Rezultat projekta prispeva k vedenju o prisotnosti joda-129 v okolju v Sloveniji ter hkrati omogoca primerjavo s podatki raziskav, ki so jih naredili raziskovalci na Švedskem, Franciji in Nemciji.

3.4. Kakšni so lahko **dolgorocni rezultati** vašega raziskovalnega projekta glede na zgoraj oznacen potencialni pomen in razvojne cilje?

Pri oceni izpostavljenosti radioaktivnosti v okolju je prispevek aktivnosti joda-129 v okolju zanemarljiv, hkrati pa je globalni onesnaževalec.

3.5. Kje obstaja verjetnost, da bodo vaša znanstvena spoznanja deležna zaznavnega odziva?

- ☐ a) v domacih znanstvenih krogih;
- ☒ b) v mednarodnih znanstvenih krogih;
- ☒ c) pri domacih uporabnikih;
- ☒ d) pri mednarodnih uporabnikih.

3.6. Kdo (poleg sofinancerjev) že izraža interes po vaših spoznanjih oziroma rezultatih?

Univerza iz Upsale, Uppsala universitet, Teknisk-naturvetenskapliga vetenskapsområdet, Tekniska sektionen, Institutionen för teknikvetenskaper, Jonfysik. Markus Meili in njegova skupina spremljajo porazdelitev joda 129 v padavinah v Severni Evropi.

3.7. Število diplomantov, magistrov in doktorjev, ki so zakljucili študij z vkljucenostjo v raziskovalni projekt?

4. Sodelovanje z tujimi partnerji:

4.1. Navedite število in obliko formalnega raziskovalnega sodelovanja s tujimi raziskovalnimi inštitucijami.

Sodelavec dr. Andrej Osterc je dobil štipendijo (za sedem tednov), ki jo podeljuje Japan Society for Promotion of Science, za strokovno izpopolnjevanje na podrocju ugotavljanja joda-129 s pospeševalnim masnim spektrometrom (AMS) pri dr. Takashi Suzuki, Japan Atomic Energy Agency, 4-24 Minatomachi Mutsu Aomori, Japanska.

4.2. Kakšni so rezultati tovrstnega sodelovanja?

Dr Andrej Osterc je na izpopolnjevanju od 13.9. 2010 do 30.10.2010. Skušal bo pripraviti vzorce padavin, ki smo jih vzorcili v letu 2009 za ugotavljanje z AMS. Kolicina padavin v Bovcu in Biljah je bila premajhna, da bi lahko jod-129 v njih ugotavljali z RNAA. Za AMS potrebujemo manj kot 2 litra vzorca vode.

5. Bibliografski rezultati[3] :

Za vodjo projekta in ostale raziskovalce v projektni skupini priložite bibliografske izpise za obdobje zadnjih treh let iz COBISS-a) oz. za medicinske vede iz Inštituta za biomedicinsko informatiko. Na bibliografskih izpisih oznacite tista dela, ki so nastala v okviru pricujocega projekta.

6. Druge reference[4] vodje projekta in ostalih raziskovalcev, ki izhajajo iz raziskovalnega projekta:

Ker je projekt dvoleten na podrocju radioaktivnosti v okolju, clanek s podrocja projekta še ni objavljen. Delo projekta je bilo predstavljeno na dveh konferencah in sicer Commission internationale pour l'exploration scientifique de la Mer Méditeranée, CIESM - 39th Congress, Venezia (Italia), May 2010 in na 16th Radiochemical Conference, 18-23 April, 2010, Mariánské Lázne, Ceška.

Pripravili smo clanek z naslovom: Fisiongenic I-129 levels in soil from Ukraine and Slovenia in last decade in je trenutno pri lektorju.V letošnjem letu še zbiramo padavine, tako da bomo tudi pripravili clanek o jodu-129 v padavinah v obdobju dveh let.

Do aprila meseca 2011 bomo pripravili clanek o prisotnosti joda v Severnem delu Jadranskega morja (sedimenti, morska voda, alge in školjke).

[3] Bibliografijo raziskovalcev si lahko natisnete sami iz spletne strani:http:/www.izum.si/

[4] Navedite tudi druge raziskovalne rezultate iz obdobja financiranja vašega projekta, ki niso zajeti v bibliografske izpise, zlasti pa tiste, ki se nanašajo na prenos znanja in tehnologije.
Navedite tudi podatke o vseh javnih in drugih predstavitvah projekta in njegovih rezultatov vkljucno s predstavitvami, ki so bile organizirane izkljucno za narocnika/narocnike projekta.

Priloga I Potek vzorčenja in določitve I-129 ter I-127

Preglednica 1 Časovni načrt vzorčenja in opravljene analize (posodobljeno 10.9.2010)

Kopensko okolje

Vzorec	Lokacija	Čas vzorčenja	A	B	C	Čas vzorčenja	A	B	C	Čas vzorčenja	A	B	C	Čas vzorčenja	A	B	C
padavine	Bovec, Bilje, Ljubljana, Iskrba	1.2 – 30.4.09	☺	☺	☺	1.5 – 31.7.09	☺	☺	☺	1.8 – 31.10.09	☺	☺	☺	1.11.09 – 31.1.10	☺	☺	☺
tla	Bovec, Bilje, Ljubljana, Iskrba	29-30.1.09	☺	☺	☺	29-30.4.09	☺	☺	☺	4-5.08.09	☺	☺	☺				
smrekove iglice	Bovec, Bilje, Ljubljana, Iskrba	29-30.1.09	☺	☺	☺	29-30.4.09	☺	☺	☺	4-5.08.09	☺	☺	☺				

Morsko okolje

Vzorec	Lokacija	Čas vzorčenja	A	B	C	Čas vzorčenja	A	B	C	Čas vzorčenja	A	B	C
morska voda	Žusterna, Portorož, Debeli rtič	1.4.09	☺	☺	☺	15.7.09	☺	☺	☺	26.11.09	☺	☺	☺
sediment	Piranski in Koprski zaliv	19.4.09	☺	☺	☺	30.7.09	☺	☺	☺	2.10.09	☺	☺	☺
alge	Žusterna, Portorož, Debeli rtič	1.4.09	☺	☺	☺	15.7.09	☺	☺	☺	26.11.09	☺	☺	☺
školjke	Koper, Piran-Seča, Strunjan	10.3.09	☺	☺	☺	15.9.10*	☺	☺	☺	10.3.10	☺	☺	☺

A…vzorčenje
B…predpriprava vzorca (zračno sušenje, liofilizacija ter homogenizacija – mletje in sejanje oz. izparevanje pri padavinah in morski vodi)
C…določitev (določitev I-129 in I-127)
☺--opravljeno delo
*--v prvotni verziji so bile navedene školjke vzorčene 10.7.09, vendar ni bilo dovolj vzorca; analizirali smo školjke vzorčene 10.3.2010 v okviru rednega monitoringa

Priloga II Rezultati za določitev vsebnosti ^{129}I in ^{127}I v analiziranih vzorcih

1. Kopensko okolje

Preglednica 1 Določitev koncentracije ^{129}I in ^{127}I v padavinah

Lokacija	^{129}I (10^{-10} µg g^{-1})	^{129}I (10^{-07} Bq kg^{-1})	^{127}I (ng g^{-1})	$^{129}I/^{127}I$ (10^{-08})
Obdobje vzorčenja: 1.2 – 30.4.09				
Bilje	< 0,5	< 0,33	2,2 ± 0,1	< 2,3
Bovec	5,4 ± 0,6	36,0 ± 4	6,5 ± 0,9	8,3
Iskrba – Kočevska Reka	18,2 ± 2,1	120,8 ± 13,9	2,0 ± 0,2	91,0
Ljubljana	1,1 ± 0,3	7,2 ± 2,0	1,7 ± 0,1	6,5
Obdobje vzorčenja: 1.5 – 31.7.09				
*Bilje				
*Bovec				
Iskrba – Kočevska Reka	8,7 ± 1,0	57,8 ± 6,6	2,1 ± 0,1	40,4
Ljubljana	15,6 ± 1,9	103,8 ± 12,6	1,7 ± 0,2	89,6
Obdobje vzorčenja: 1.8 – 31.10.09				
*Bilje				
*Bovec				
Iskrba – Kočevska Reka	< 0,5	< 0,33	2,8 ± 0,2	< 1,8
Ljubljana	7,1 ± 1,0	46,9 ± 6,6	2,9 ± 0,2	24,5
Obdobje vzorčenja: 1.11.09 – 31.1.10				
*Bilje				
*Bovec				
Iskrba – Kočevska Reka	3,9 ± 0,6	26,0 ± 4,0	2,1 ± 0,2	18,6
Ljubljana	< 0,5	< 0,33	2,3 ± 0,2	< 2,2

Rezultati so podani z merilno negotovostjo (95 % interval zaupanja, k = 2).
*Volumen vode je premajhen za določitev I-129 z razvito metodo

Preglednica 2 Določitev koncentracije ^{129}I in ^{127}I v tleh

Lokacija		^{129}I (10^{-08} μg g^{-1})	^{129}I (10^{-04} Bq kg^{-1})	^{127}I (μg g^{-1})	$^{129}I/^{127}I$ (10^{-08})
Datum vzorčenja: 29.1-5.2.2009					
Bovec	na prostem	56 ± 10	37 ± 7	22,2 ± 0,8	2,5
	pod smreko	114 ± 18	75 ± 12	26,6 ± 0,6	4,4
Bilje	na prostem	19 ± 3	13 ± 2	14,3 ± 0,4	1,4
	pod borom	91 ± 13	61 ± 9	27,6 ± 1,3	3,3
Iskrba	na prostem	8,2 ± 1,3	5,5 ± 0,9	12,3 ± 0,5	0,7
	pod smreko	24 ± 3	16 ± 2	15,2 ± 0,7	1,6
Dol	na prostem	15 ± 2	10 ± 1,3	4,5 ± 0,3	3,3
	pod smreko	37 ± 5	24 ± 3	12,6 ± 0,6	2,9
Datum vzorčenja: 29.4-4.5.2009					
Bovec	na prostem	73 ± 10	48 ± 6,6	14,1 ± 0,3	5,2
	pod smreko	64 ± 10	43 ± 6,7	21,8 ± 1,3	2,9
Bilje	na prostem	22 ± 4	14 ± 2,5	11,6 ± 0,4	1,9
	pod borom	30 ± 5	20 ± 3	35,2 ± 2,2	0,9
Iskrba	na prostem	20 ± 4	13 ± 2,6	15,5 ± 0,4	1,3
	pod smreko	22 ± 4	15 ± 2,7	16,9 ± 0,8	1,3
Dol	na prostem	17 ± 3	11 ± 1,9	2,4 ± 0,1	7,1
	pod smreko	29 ± 5	19 ± 3,3	3,9 ± 0,2	7,4
Datum vzorčenja: 4-5.08.2009					
Bovec	na prostem	68 ± 10	45 ± 6,6	13,6 ± 0,6	5,0
	pod smreko	129 ± 19	85 ± 12,5	28,1 ± 1,3	4,6
Bilje	na prostem	17 ± 3	12 ± 2,1	15,2 ± 0,7	1,1
	pod borom	25 ± 4	17 ± 2,7	34,9 ± 1,6	0,7
Iskrba	na prostem	9,9 ± 1,4	6,6 ± 0,9	8,8 ± 0,4	1,1
	pod smreko	29,6 ± 5	19,6 ± 3	14,8 ± 0,7	2,0
Dol	na prostem	12,5 ± 2	8,3 ± 1,3	4,1 ± 0,2	3,0
	pod smreko	30,2 ± 5	20,1 ± 3	7,5 ± 0,3	4,0

Rezultati so podani na zračno suho snov z merilno negotovostjo (95 % interval zaupanja, k = 2).

Preglednica 3 Določitev koncentracije ^{129}I in ^{127}I v smrekovih iglicah

Lokacija	**^{129}I (10^{-08} μg g^{-1})**	**^{129}I (10^{-04} Bq kg^{-1})**	**^{127}I (ng g^{-1})**	**$^{129}I/^{127}I$ (10^{-08})**
Datum vzorčenja: 29.1-5.2.2009				
Bovec	5,9 ± 1,0	3,9 ± 0,7	81 ± 2	73
Bilje	6,2 ± 1,2	4,1 ± 0,8	413 ± 12	15
Iskrba*	15 ± 2,7	10 ± 1,8	145 ± 3	105
Dol	< 5	< 3,3	74 ± 4	< 68
Datum vzorčenja: 29.4-4.5.2009				
Bovec	9,0 ± 1,6	6,0 ± 1,1	58 ± 2	156
Bilje	9,7 ± 1,8	6,5 ± 1,2	303 ± 10	32
Iskrba	8,1 ± 1,3	5,4 ± 0,9	101 ± 3	80
Dol	6,8 ± 1,0	4,6 ± 0,7	74 ± 3	92
Datum vzorčenja: 4-5.08.2009				
Bovec	12 ± 2,1	8,0 ± 1,4	64 ± 3	189
Bilje	13 ± 2,4	8,6 ± 1,6	263 ± 9	49
Iskrba	10 ± 1,9	6,8 ± 1,3	80 ± 4	129
Dol	7,2 ± 1,1	4,8 ± 0,7	89 ± 5	81

Rezultati so podani na zračno suho snov z merilno negotovostjo (95 % interval zaupanja, k = 2).
* v Vmesnem poročilu ARRS –V2-0555-VP4-2010 je bila navedena napačna lokacija Bilje.

2 Morsko okolje

Preglednica 4 Določitev koncentracije ^{129}I in ^{127}I v morski vodi

Lokacija	^{129}I (10^{-10} µg g^{-1})	^{129}I (10^{-07} Bq kg^{-1})	^{127}I (ng g^{-1})	$^{129}I/^{127}I$ (10^{-08})
Datum vzorčenja: 1.4.2009				
Portorož	< 0,5*	< 3,3	56 ± 1	< 1,0
Žusterna	< 0,5	< 3,3	63 ± 5	< 1,0
Debeli Rtič	17,2 ± 2,0	115,0 ± 13,4	59 ± 5	2,9
Datum vzorčenja: 15.7.2009				
Portorož	13,4 ± 1,6	89,2 ± 10,7	70 ± 1	1,9
Žusterna	10,7 ± 1,3	71,3 ± 8,7	53 ± 2	2,0
Debeli Rtič	5,5 ± 0,7	36,8 ± 4,7	61 ± 2	0,9
Datum vzorčenja: 26.11.2009				
Portorož	21 ± 2	139 ± 13	69 ± 5	3,0
Žusterna	5,1 ± 0,6	34,0 ± 4,0	61 ± 3	0,8
Debeli Rtič	< 0,5	< 3,3	69 ± 6	< 1,0

Rezultati so podani z merilno negotovostjo (95 % interval zaupanja, k = 2).
*Mejo zaznavnosti metode za ^{129}I je 0,5 *E-10ug/g pri odtehti 8 kg morske vode.

Preglednica 5 Določitev koncentracije ^{129}I in ^{127}I v morskem sedimentu

Lokacija	**^{129}I (10^{-08} µg g^{-1})**	**^{129}I (10^{-04} Bq kg^{-1})**	**^{127}I (µg g^{-1})**	**$^{129}I/^{127}I$ (10^{-08})**
Datum vzorčenja: 19.4.2009				
Koper	17 ± 3	12 ± 2	79,1 ± 0,6	0,22
Piran	16 ± 3	11 ± 2	110,7 ± 1,3	0,15
Strunjan	8,7 ± 1,6	5,8 ± 1,1	98,9 ± 1,8	0,09
Datum vzorčenja: 30.7.2009				
Koper	5,7 ± 1,0	3,8 ± 0,7	87,7 ± 1,7	0,07
Piran	8,7 ± 1,5	5,7 ± 1,0	100,2 ± 1,9	0,09
Strunjan	5,7 ± 1,1	3,8 ± 0,8	82,0 ± 1,4	0,07
Datum vzorčenja: 2.10.09				
Koper	11,7 ± 1,7	7,7 ± 1,1	81,3 ± 2,4	0,14
Piran	< 5,0	< 3,3	99,9 ± 2,1	< 0,07
Strunjan	4,6 ± 1,2	3,0 ± 0,8	74,9 ± 1,5	0,06

Rezultati so podani na suho snov z merilno negotovostjo (95 % interval zaupanja, k = 2).

Preglednica 6 Določitev koncentracije ^{129}I in ^{127}I v morskih algah Jadranski bračič (*Fucus virsoides*)

Lokacija	**^{129}I (10^{-08} µg g^{-1})**	**^{129}I (10^{-04} Bq kg^{-1})**	**^{127}I (µg g^{-1})**	**$^{129}I/^{127}I$ (10^{-10})**
Datum vzorčenja: 1.4.2009				
Portorož	34 ± 7	22 ± 5	464 ± 10	7
Žusterna	26 ± 6	17 ± 4	274 ± 9	10
Debeli Rtič	32 ± 8	22 ± 6	328 ± 6	10
Datum vzorčenja: 15.7.09				
Portorož	43 ± 9	28 ± 6	451 ± 10	9
Žusterna	32 ± 7	21 ± 5	324 ± 7	10
Debeli Rtič	44 ± 7	29 ± 5	417 ± 9	10
Datum vzorčenja: 26.11.09				
Portorož	30 ± 6	20 ± 4	438 ± 12	7
Žusterna	20 ± 4	13 ± 3	387 ± 8	5

Rezultati so podani na liofilizirano snov z merilno negotovostjo (95 % interval zaupanja, k = 2).

Preglednica 7 Določitev koncentracije ^{129}I in ^{127}I v školjkah klapavicah

Lokacija	**^{129}I (10^{-08} µg g^{-1})**	**^{129}I (10^{-04} Bq kg^{-1})**	**^{127}I (µg g^{-1})**	**$^{129}I/^{127}I$ (10^{-08})**
Datum vzorčenja: 10.3.2009				
Koper	10 ± 2	7,0 ± 1,4	13,2 ± 0,4	0,8
Strunjan	13 ± 3	8,0 ± 1,8	14,6 ± 0,2	0,9
Piran-Seča	93 ± 15	62 ± 10	9,9 ± 0,2	9,4
Datum vzorčenja: 15.9.2009				
Koper	< 5,0	< 3,3	6,9 ± 0,3	< 0,7
Strunjan	9,0 ± 1,7	6,0 ± 1,1	9,0 ± 0,4	1,0
Piran-Seča	5,3 ± 1,0	3,6 ± 0,7	10,3 ± 0,5	0,5
Datum vzorčenja: 10.3.2010				
Koper	4,9 ± 0,9	3,3 ± 0,6	18,3 ± 0,9	0,3
Strunjan	10,9 ± 1,8	7,2 ± 1,2	14,2 ± 0,4	0,8
Piran-Seča	4,2 ± 0,8	2,8 ± 0,5	13,8 ± 0,5	0,3

Rezultati so podani na liofilizirano snov z merilno negotovostjo (95 % interval zaupanja, k = 2).

Priloga III Primerjava rezultatov s podatki iz literature

1. Kopensko okolje

Preglednica 1 Koncentracije ^{129}I in ^{127}I v padavinah (Buraglio in sod., 2001)

Lokacija	**Datum vzorčenja**	**^{129}I (10^{-10} µg g^{-1})**	**^{129}I (10^{-07} Bq kg^{-1})**	**^{127}I (ng g^{-1})**	**$^{129}I/^{127}I$ (10^{-08})**
Švedska	25-30.10.98	8,7 ± 0,5	57,7 ± 0,4	5,2	17
Švedska	06.1.99	0,10 ± 0,01	0,70 ± 0,06	0,7	1,6
Švedska	25.1.99	1,0 ± 0,1	6,4 ± 0,4	1,9	5,1
Švedska	27.8.99	1,7 ± 0,1	11,2 ± 0,4	2,9	5,8

Preglednica 2 Koncentracije ^{129}I in ^{127}I v tleh

Lokacija	**Mesto vzorčenja**	**Datum vzorčenja**	**^{129}I (10^{-08} µg g^{-1})**	**^{129}I (10^{-04} Bq kg^{-1})**	**^{127}I (µg g^{-1})**	**$^{129}I/^{127}I$ (10^{-10})**	**Referenca**
Japonska*							Matsuzaki in sod., 2007
Tokyo (Tachikawa)	polje za posevke	ni podan	-	4,3 ± 0,2	17,0 ± 0,8	39	
Iwate	polje za posevke	ni podan	-	2,3 ± 0,1	21,1 ± 0,9	16	
Ishikawa	polje za posevke	ni podan	-	2,4 ± 0,1	9,9 ± 0,4	38	
Hokkaido	gozd	ni podan	-	1,3 ± 0,1	18,0 ± 0,8	11	
Kagoshima	gozd	ni podan	-	0,80 ± 0,05	55,3 ± 2,5	2	
Osaka	riževo polje	ni podan	-	1,1 ± 0,1	0,80 ± 0,05	218	
Hokkaido	riževo polje	ni podan	-	0,50 ± 0,03	1,2 ±0,1	59	
Aomori	riževo polje	ni podan	-	0,70 ±0,03	2,3 ± 0,1	50	
Ibaraki (Tokai-mura A)	polje za posevke	ni podan	-	260 ± 20	33,0 ± 2,5	1200	
Ibaraki (Tokai-mura A)	polje za posevke	ni podan	-	40 ± 2	11,0 ± 0,4	570	
Švedska**							Hou in sod., 2003
Nora	gozd	July, 2000	38,6 ± 2,7	-	ni podan	ni podan	
Nora	gozd	July, 2000	6,8 ± 1,5	-	ni podan	ni podan	
Gävle	vrt	August, 1996	4,5 ± 1,3	-	ni podan	ni podan	
Gävle	trata	August, 1996	24,7 ± 2,7	-	ni podan	ni podan	
Gävle	trata	August, 1996	7,3 ± 1,5	-	ni podan	ni podan	
Danska***							Hou in sod., 1999
Roskilde	ni podan	January, 1999	43,2 ± 4,4	-	5.92 ± 0.10	730	

*zračno suhe vzorce tal so nato še sušili 3 h pri 80 °C
**vzorce tal so sušili dva dni pri 60 °C
*** vzorec tal so sušili dva dni pri 70 °C

Preglednica 3 Koncentracije ^{129}I in ^{127}I v iglicah pinij in ceder (Quintana in Thyssen, 2000)

Lokacija	Oddaljenost od vira (m)	Smer	^{129}I (10^{-04} Bq kg^{-1})	^{127}I (ng g^{-1})	$^{129}I/^{127}I$ (10^{-08})
Argentina					
CNA I	2000	JJZ	4680	ni podan	ni podan
CNA I	2500	JJV	4010	ni podan	ni podan
CNE	3000	V	35500	ni podan	ni podan
CNE	500	J	<210	ni podan	ni podan
CNE	1000	JZ	1880	ni podan	ni podan
CAE	200	S	<210	ni podan	ni podan
CAE	50	S	100	ni podan	ni podan
CAE	100	J	730	ni podan	ni podan
Buenos Aires	***		<80	ni podan	ni podan

Rezultati so podani na sveže vzorce
*datum vzorčenja ni podan
**CNA I - jedrska elektrarna Atucha, CNE - jedrska elektrarna Embalese, CAE - atomski center Ezeiza
***Buenos Aires je služil za določitev naravnega ozadja, zaradi oddaljenosti od virov (100 km, 800 km in 40 km od CNA I, CNE in CAE)

2. Morsko okolje

Preglednica 4 Koncentracije ^{129}I in ^{127}I v morski vodi

Lokacija	**Datum vzorčenja**	**^{129}I (10^{-10} μg g^{-1})**	**^{129}I (10^{-07} Bq kg^{-1})**	**^{127}I (ng g^{-1})**	**$^{129}I/^{127}I$ (10^{-08})**	**Vir**
Grenlandija (vzhod)	22.6.99	1,4 ± 0,1	9,0 ± 0,7	56	0,24 ± 0,03	Hou, 2004
Grenlandija (vzhod)	8.10.99	1,2 ± 0,1	8,2 ± 0,8	56	0,22 ± 0,03	
Grenlandija (zahod)	26.8.99	1,0 ± 0,1	6,5 ± 0,9	56	0,17 ± 0,03	
Grenlandija (zahod)	02.9.99	0,6 ± 0,1	3,7 ± 0,7	57	0,10 ± 0,03	
Grenlandija (jug)	15.9.99	0,4 ± 0,1	2,5 ± 0,4	57	0,07 ± 0,01	
Rokavski preliv	sep. 2005	804	5340	51	158,3	Hou in sod. 2007*
Severno morje	avg. 2005	488	3240	32	150,6	
Japonska (zaliv Toyama)	17.10.2006	0,04 ± 0,01	0,30 ± 0,04	49 ± 1	0,010 ± 0,001	Suzuki in sod. 2008

*merilne negotovosti so 3-7% za ^{127}I in 4-10% za ^{129}I

Preglednica 5 Koncentracije ^{129}I in ^{127}I v morskih algah

Vrsta alge	**Datum vzorčenja**	**Lokacije**	**^{129}I (10^{-4} Bq kg^{-1})**	**^{127}I (μg g^{-1})**	**$^{129}I/^{127}I$ (10^{-10})**	**Vir**
F. virsoides	sep. 2005	Portorož	31 ± 9	420 ± 31	11,2 ± 1,2	Osterc in Stibilj., 2008*
F. virsoides	sep. 2005	Žusterna	24 ± 6	371 ± 26	9,8 ± 0,9	
F. virsoides	sep. 2005	Debeli rtič	29 ± 8	506 ± 35	8,6 ± 0,8	
F. virsoides	sep. 2005	Piran	27 ± 7	411 ± 27	10,1 ± 1,1	
F. virsoides	sep. 2005	Ankaran	27 ± 7	448 ± 34	8,9 ± 0,8	
F. virsoides	jun. 2006	Ribiška vas	23 ± 6	234 ± 18	14,6 ± 1,5	
F. virsoides	jun. 2006	Nabrežina	17 ± 4	491 ± 28	5,3 ± 0,4	
F. virsoides	jun. 2006	Sesljan	19 ± 4	410 ± 35	6,8 ± 0,6	
F. virsoides	jun. 2006	grad Miramar	22 ± 5	441 ± 20	7,6 ± 0,8	
F. virsoides	jun. 2006	Trst	25 ± 7	484 ± 22	7,9 ± 0,9	
F. virsoides	okt. 2006	Poreč	37 ± 10	380 ± 21	14,7 ± 1,6	
F. virsoides	okt. 2006	Umag	34 ± 10	280 ± 18	18,2 ± 2,0	
F. virsoides	okt. 2006	Rovinj	73 ± 16	348 ± 26	31,4 ± 3,6	
F. vesicolosus	dec. 1988	Islandija(1)	9 ± 0,5	330,0 ± 0,7	3,95 ± 0,24	Hou in sod., 1999**
F. vesicolosus	dec. 1988	Islandija(2)	14 ± 0,6	364,0 ± 1,5	5,97 ± 0,26	
F. vesicolosus	nov. 1989	Klint, Danska	870 ± 65	328,0 ± 4,2	399,0 ± 5,9	

Vrsta alge	Datum vzorčenja	Lokacije	^{129}I (10^{-4} Bq kg^{-1})	^{127}I (μg g^{-1})	^{129}I/^{127}I (10^{-10})	Vir
F. vesicolosus	april 1992	Klint, Danska	1614 ± 145	375,0 ± 5,3	648,0 ±10,4	
F. vesicolosus	mar. 1994	Klint, Danska	3508 ± 13	382,0 ± 3,0	1380 ± 10	Hou in sod., 2000a**
F. vesicolosus	mar. 1996	Klint, Danska	5474 ± 27	504,0 ± 12,0	1630 ± 40	
F. vesicolosus	jan. 1998	Klint, Danska	5793 ± 33	256,0 ± 10,0	3410 ± 130	
F. vesicolosus	okt. 1998	Goury, Francija	~196000	~297,4	~101000	Fréchou in sod., 2003***
F. vesicolosus	mar. 1999	Goury, Francija	~448000	~354,0	~194000	
L. japonica	julij 1996	Xiamen, Kitajska	35 ± 2	2980 ± 10	1,73 ± 0,12	Hou in sod., 2000***
L. japonica	april 1997	Weihai, Kitajska	22 ± 1	2850 ± 10	1,09 ± 0,06	

*Rezultati so podani na liofilizirano snov
**Rezultati so podani na suho snov

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I-129 LEVELS IN MARINE ENVIRONMENT ALONG THE SLOVENIAN COAST

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Abstract

^{129}I is considered as a global pollutant and its role as a global tracer to follow the dissemination of radionuclides from a source point such as nuclear power reprocessing plants increases. A radiochemical neutron activation analysis method was developed to measure the concentration of ^{129}I in environmental and biological samples. The method was validated using the IAEA-375 Soil, FC98 Seaweed and NIST 4357 Ocean Sediment. The method was applied to analyze $^{129}I/^{127}I$ isotopic ratios in the marine environment of Slovenia. The results found were in the range from 1.3 to 55.4 · 10^{-9} μq g^{-1} for seawater, from 104 to 127 · 10^{-9} μq g^{-1} for blue mussel, from 334 to 471 · 10^{-9} μq g^{-1} for alga *Fucus virsoides* and from 72 to 256 · 10^{-9} μq g^{-1} for marine sediment.

Keywords: Algae, Coastal Waters, Fallout, Radionuclides, Sediments

1 Introduction

The only stable natural iodine isotope is ^{127}I and the total amount of this element in the Earth's crust was estimated to be 8.6 · 10^{15} kg of which nearly 70 % resides in marine sediments and 28 % in sedimentary rocks. The marine environment, i.e. the oceans, is the major source of iodine with average concentrations of around 60 μg L^{-1} iodine in seawater. The biogeochemical cycling of iodine is driven with its volatilization from oceans and soil to the atmosphere in the form of iodinated hydrocarbons of which methyl-iodide predominates. From the atmosphere the iodine is washed out to the marine and terrestrial environment by wet (precipitation) and dry (aerosol) depositions [1].

^{129}I ($T_{½}$ = 1.57 · 10^7 years) is the only natural radioactive isotope of iodine, which is formed in nature by two processes. The cosmogenic ^{129}I is produced in the atmosphere by the interaction of cosmic rays with xenon isotopes and the fissiogenic ^{129}I by spontaneous fission of uranium in the lithosphere. For the pre-nuclear era (no addition of anthropogenic ^{129}I to the environment) an $^{129}I/^{127}I$ isotopic ratio of about $1.5 \cdot 10^{-12}$ has been estimated. The quantity of ^{129}I in the pre-nuclear age ocean was ~100 kg. Since 1945 anthropogenic production of ^{129}I started which shifted the natural isotopic ratio for 3 to 6 orders of magnitude in favour of ^{129}I.
The main sources of ^{129}I are nuclear fuel reprocessing plants.

To our knowledge the ^{129}I level has not been measured in any biological or environmental sample from the Mediterranean area (Adriatic Sea). The aim of our work was to investigate the distribution of ^{129}I in the marine environment of Slovenia.

2 Methodology

2.1 Sampling and preparation
First sampling of alga (*Fucus virsoides*) and sediment was performed in 2005 and another sampling in 2009 including seawater, alga (*Fucus virsoides*) and blue mussel (*Mytilus galloprovincialis*). Alga and blue mussel were dried by freeze dryer to constant mass and homogenized. Seawater and sediment samples were analysed as collected.

2.2 Determination of ^{129}I and ^{127}I
Radiochemical neutron activation analysis method (RNAA) was used for the determination of ^{129}I [2] and ^{127}I [3] in environmental samples. Environmental samples contain very low amounts of ^{129}I therefore pre-concentration of iodine from up to 100 g of alga, blue mussel and sediment and up to 8 L of seawater are needed. Irradiation of sample, combustion in an oxygen atmosphere and extraction of iodine with $CHCl_3$ followed. Induced radioisotopes were measured on a HPGe detector. The chemical yield for the whole procedure was determined spectrophotometrically and by using the ^{126}I activity.

3 Results and Discussions

The method was applied to analyze $^{129}I/^{127}I$ isotopic ratios as well as ^{129}I and ^{127}I concentrations in the marine environment of Slovenia. The results found for analysed samples collected in 2005 and 2009 are summarised in Table 1. There are no literature dates for ^{129}I and ^{127}I concentrations in blue mussel. Values found in analysed seawater and sediment samples are in agreement with values found in literature for areas that are not under the influence of direct liquid discharges of ^{129}I from nuclear fuel reprocessing plants. ^{129}I and ^{127}I concentrations found in analysed alga collected in 2005 and 2009 (Table 1) are in the same range. The ratio of $^{129}I/^{127}I$ found for alga *Fucus virsoides* is up to 10^{-9}, which is one order of magnitude higher than along the coast of China, up to two orders of magnitude lower than in the Baltic Sea, which is influenced by direct liquid discharges from La Hague and Sellafield, and up to four orders of magnitude lower than in the vicinity of the La Hague reprocessing plant.

Tab. 1. Range of ^{129}I and ^{127}I in marine environment of North Adriatic Sea

Sample	Number of samples	Year of sampling	Range	
			^{127}I (μg g^{-1} dry weight)	^{129}I (10^{-9} μg g^{-1} dry weight)
Seawater	6	2009	0.052 – 0.070	1.3 – 55.4
Alga (*Fucus virsoides*)	3	2009	267 – 470	334 – 452
	5	2005[a]	371 – 448	362 – 471
Blue mussel (*Mytilus galloprovincialis*)	3	2009	9.9 – 14.6	104 – 127
Sediment	4	2005	59.3 – 76.8	72 – 256

[a] published in 2008 [4]

The data of this study represent a survey of ^{129}I in the marine environment of Slovenia. The most likely source of ^{129}I are nuclear fuel reprocessing plants in La Hague and Sellafield, which are known to be the major sources of ^{129}I in the environment of North Europe. ^{129}I is transferred to the atmosphere and washed out to the marine environment of Slovenia by precipitation, so it is of atmospheric-precipitation origin.

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I-129 levels in marine environment along the Slovenian coast

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Introduction

^{129}I is considered as a global pollutant and its role as a global tracer to follow the dissemination of radionuclides from a source point such as nuclear fuel reprocessing plants increases. The operating plants in Europe are located in England (Sellafield), France (La Hague) and Russia, and outside Europe in India and Japan (Tokaimura, Rakkasho). The natural $^{129}I/^{127}I$ isotopic ratio in the order of 10^{-12} was significantly influenced by releases of anthropogenic ^{129}I to the environment. The ratio of $^{129}I/^{127}I$ in the marine environment has increased to 10^{-11} – 10^{-10}. Anthropogenic ^{129}I predominates in the biosphere and in upper layers of the oceans, therefore it can be expected that the isotopic ratio $^{129}I/^{127}I$ is increasing in these compartments of the ecosystem.

Aim

The aim of our work was to determine the levels and distribution of ^{129}I in marine environment of Slovenia.

Methodology

Sampling and sample preparation

First sampling of alga (*Fucus virsoides*) and sediment was performed in 2005 and another sampling in 2009 including seawater, alga (*Fucus virsoides*) and blue mussel (*Mytilus galloprovincialis*). Alga and blue mussel were dried by freeze dryer to constant mass and homogenized. Seawater and sediment samples were analysed as collected.

1 Debeli rtič
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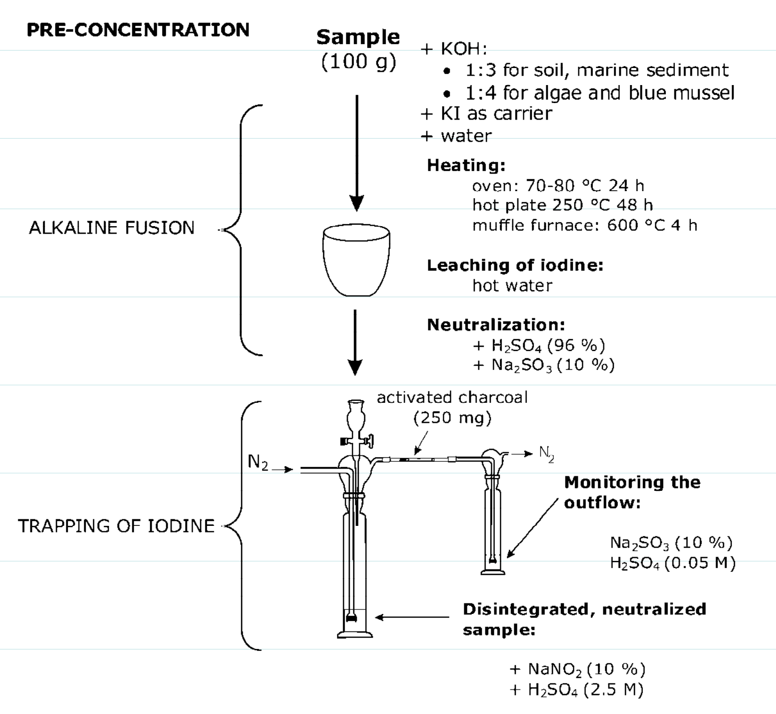
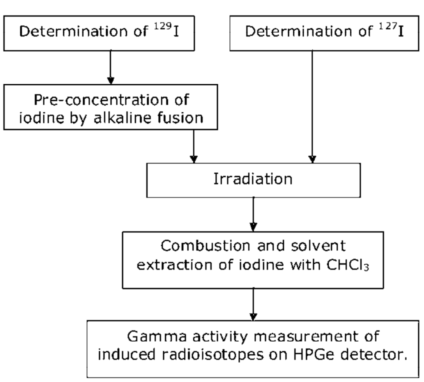


Determination of ^{129}I and ^{127}I by RNAA



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The accuracy of the method was checked by certified reference material IAEA 375, by reference material NIST 4357 and by literature data (Table 1).

Table 1 ^{129}I and ^{127}I in IAEA 375, NIST 4357 and FC-98 Seaweed

Reference	^{129}I (10^{-7} µg g^{-1})	^{129}I (10^{-3} Bq kg^{-1})	^{127}I (n = 4) (µg g^{-1})	$^{129}I/^{127}I$ (10^{-10})
IAEA 375 Soil				
Our value	2.63 ± 0.39 (n = 2)	1.75 ± 0.26 (n = 2)	1.74 ± 0.10 (n = 4)	~1510
Recommended value	-	1.7 (1.3 – 2.1)	-	-
NIST 4357 Sediment				
Our value	2.2 ± 0.5 (n = 3)	1.5 ± 0.4 (n = 3)	3.8 ± 0.1 (n = 4)	580 ± 60
Informative value	-	9 (6.0 – 12.0)	3.1	-
FC 98 Seaweed				
Our value(Osterc and Stibilj, 2008)	95400 ± 8600	63500 ± 5700	795.5 ± 10.7	120000 ± 14000
Fréchou et al., 2001 (n = 6)	-	61300 ± 10700 (53300 – 68300)	-	122000 ± 21000 (106000 – 136000)

Results and Discussions

^{129}I levels in marine environment of Slovenia

The results found for analysed samples are summarised in Table 2 and this are the first data on ^{129}I in the environment of North Adriatic Sea.

1 Values found in analysed seawater and sediment samples are in agreement with values found in literature for areas that are not under the influence of direct liquid discharges of ^{129}I from nuclear fuel reprocessing plants.

2 ^{129}I and ^{127}I concentrations found in analysed alga collected in 2005 and 2009 are in the same range. The ratio of $^{129}I/^{127}I$ found for alga *Fucus virsoides* is up to 10^{-9}.

Table 2 Range of ^{129}I and ^{127}I in marine environment of Slovenia and other countries

Sample	Location	Number of samples	Year of sampling	Range			Reference
				^{127}I (µg g^{-1} dry weight)	^{129}I (10^{-9} µg g^{-1} dry weight)	$^{129}I/^{127}I$ (10^{-08})	
Seawater	Slovenia	8	2009	0.053 – 0.070*	0.5 – 2.1*	0.8 – 2.9	This study
Sediment		6	2009	79.1 – 110.7	57 – 170	0.07 – 0.22	
		4	2005	59.3 – 76.8	72 – 256	0.12 – 0.33	
Alga (*Fucus virsoides*)		3	2009	274 – 464	320 – 340	0.07 – 0.10	
		5	2005**	371 – 448	362 – 471	0.09 – 0.15	
Blue mussel (*Mytilus galloprovincialis*)		3	2009	9.9 – 14.6	100 – 930	0.8 – 9.4	
Seawater	Greenland	5	1999	0.056 – 0.057*	0.06 – 0.14*	0.10 – 0.24	Hou, 2004
	Japan	1	2006	0.049*	0.004*	0.01	Suzuki et al., 2008
Sediment	Baltic Sea	11	1997	41.6 – 75.9	11 – 740	0.02 – 1	Aldahan et al., 2007
Alga (*Fucus vesicolosus*)	China	2	1996, 1997	2850 – 2980	331 – 527	0.01 – 0.02	Hou et al., 2000
	Iceland	2	1988	330 – 364	135 – 210	0.04 – 0.06	Hou et al., 1999
	Denmark	5	1989 – 1998	256 – 504	131000 – 872000	4 – 34	Hou et al., 1999
	France	2	1998, 1999	297 – 354	2950000 – 6740000	1010 – 1940	Frechou et al. 2003

*µg g^{-1} seawater
**already published (Osterc and Stibilj, 2008)

measurement uncertainty of ^{127}I determination was below 6 %
measurement uncertainty of ^{129}I determination was below 20 %

Conclusions

To the best of knowledge, this work presents the first data for ^{129}I levels in the environment of this region of Europe.

Slovenia and the Adriatic Sea are isolated from direct liquid discharges of ^{129}I to the environment. The only possible discharges of ^{129}I to this environment are of atmospheric-precipitation origin, either coming directly from the atmospheric discharges or indirectly from the liquid discharges of ^{129}I to the ocean from which the iodine is transferred to the atmosphere and washed out to the marine environment of Slovenia by precipitation.

Acknowledgement

Authors wish to thank the Slovenian Nuclear Safety Administration, Ministry of the Environment and spatial planning and Slovenian Research Agency for financing the research.

The blue mussel and marine sediment samples were kindly provided by dr. Andreja Ramšak and assist. prof. Oliver Bajt from the Marine Biology station in Piran, National Institute of Biology.

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Osterc A. and Stibilj V., 2005, Accredit. qual. assur., 10: 235-240.

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Origin of ^{129}I in the environment of Slovenia

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I-129 is considered as a global pollutant and its role as a global tracer to follow the dissemination of radionuclides from a source point such as nuclear fuel reprocessing plants increases. The natural I-129/I-127 isotopic ratio in the order of 10E-12 was significantly influenced by releases of anthropogenic I-129 to the environment. The ratio of I-129/I-127 in the marine environment has increased to 10E-11 – 10E-10, and in the terrestrial environment to 10E-9 – 10E-7, even 10E-6 in the vicinity of nuclear fuel reprocessing plants. Anthropogenic I-129 predominates in the biosphere, soil and in upper layers of the oceans, therefore it can be expected that the isotopic ratio I-129/I-127 is increasing in these compartments of the ecosystem.

At the Josef Stefan Institute we have developed a radiochemical neutron activation analysis method (RNAA) for the determination of I-129 in environmental samples and the detection limit of the method is 5·10E-14 g I-129 / g solid sample. The developed method was used to trace I-129 in the environment of Slovenia. From marine environment seawater, alga, thorny oyster and sediment samples were analyzed and from terrestrial environment precipitation, soil and pine needles. The results found were in the range from 7.2 to 120.8 • 10E-7 Bq/kg for precipitation, up to 75 • 10E-4 Bq/kg for soil, from 0.1 to 2.12 • 10E-4 Bq/kg for seawater, from 7 to 62 • 10E-4 Bq/kg for thorny oyster and from 17 to 73 • 10E-4 Bq/kg for brown alga Fucus virsoides (Donati) J. Agardh., which is endemic to the Adriatic Sea. The content of I-129 in marine sediment and pine needles samples analyzed till now was below the detection limit of the method.

The data of this study represent a survey of I-129 in the environment of Slovenia. The most likely source of I-129 are nuclear fuel reprocessing plants in La Hague and Sellafield, which are known to be the major sources of I-129 in the environment of North Europe. I-129 is transferred to the atmosphere and washed out to the marine and terrestrial environment of Slovenia by precipitation, so it is of atmospheric-precipitation origin.

Content :

I-129 is considered as a global pollutant and its role as a global tracer to follow the dissemination of radionuclides from a source point such as nuclear fuel reprocessing plants increases. The natural I-129/I-127 isotopic ratio in the order of 10E-12 was significantly influenced by releases of anthropogenic I-129 to the environment. The ratio of I-129/I-127 in the marine environment has increased to 10E-11 – 10E-10, and in the terrestrial environment to 10E-9 – 10E-7, even 10E-6 in the vicinity of nuclear fuel reprocessing plants. Anthropogenic I-129 predominates in the biosphere, soil and in upper layers of the oceans, therefore it can be expected that the isotopic ratio I-129/I-127 is increasing in these compartments of the ecosystem.

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Type : Poster

Origin of ^{129}I in the Environment of Slovenia

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1 Introduction

^{129}I is considered as a global pollutant and its role as a global tracer to follow the dissemination of radionuclides from a source point such as nuclear fuel reprocessing plants increases. The operating plants in Europe are located in England (Sellafield), France (La Hague) and Russia, and outside Europe in India and Japan (Tokaimura, Rakkasho). The natural $^{129}I/^{127}I$ isotopic ratio in the order of 10^{-12} was significantly influenced by releases of anthropogenic ^{129}I to the environment. The ratio of $^{129}I/^{127}I$ in the marine environment has increased to 10^{-11} – 10^{-10}, and in the terrestrial environment to 10^{-9} – 10^{-7}, even 10^{-6} in the vicinity of nuclear fuel reprocessing plants. Anthropogenic ^{129}I predominates in the biosphere, soil and in upper layers of the oceans, therefore it can be expected that the isotopic ratio $^{129}I/^{127}I$ is increasing in these compartments of the ecosystem.

2 Aim

The aim of our work was to determine the levels and distribution of ^{129}I in terrestrial and marine environment of Slovenia.

3 Methodology

3.1 Sampling

Sampling locations are shown in Figure 1.

- marine environment: seawater, alga (*Fucus virsoides*, Figure 2), blue mussel (*Mytilus galloprovincialis*, Figure 3) and sediment;
- terrestrial environment: precipitation, soil and pine needles.

Figure 2 *Fucus virsoides*

Figure 3 *Mytilus galloprovincialis*

3.2 Sample preparation

- Seawater and precipitation samples were analyzed as collected.
- Marine sediment was analyzed as collected – dry mass was determined on an aliquot.
- Alga and blue mussel were dried by freeze dryer to constant mass and then homogenized with a Pulverisette 14 rotor speed mill from Fritsch.
- Soil and pine needles were air dried (natural drying) in a dark room. Dried soil samples were homogenized by sieving, opening 0.2 mm. Pine needles were homogenized with the speed mill.

3.3 Determination of ^{129}I

At the Josef Stefan Institute we have developed a radiochemical neutron activation analysis method (RNAA) for the determination of ^{129}I in environmental samples and the detection limit of the method is $5 \cdot 10^{-14}$ g ^{129}I g^{-1} sample. After pre-concentration of iodine, irradiation and combustion of sample followed. Iodine was extracted with $CHCl_3$. The induced radionuclides ^{126}I [$^{127}I(n, 2n)^{126}I$ ($t_{½}$ = 13 d, $E\gamma$ = 389 keV)] and ^{130}I [$^{129}I(n, \gamma)^{130}I$ ($t½$ = 12.36 h, $E\gamma$ = 536 keV)] were measured on a coax type HPGe detector (Osterc and Stibilj, 2007).

3.4 Determination of ^{127}I

The method for stable iodine determination by RNAA was based on simultaneous irradiation of the sample and a standard solution of ^{127}I. Combustion of sample and extraction of iodine with $CHCl_3$ followed. The induced radionuclide [$^{127}I(n, \gamma)^{128}I$, $t_{½}$ = 25 min, E_{γ} = 422.9 keV] was measured on a well type HPGe detector (Osterc and Stibilj, 2005).

3.5 Validation of the method

The accuracy of the method was checked by certified reference material IAEA 375, by standard reference material NIST 4357 and by literature data (Table 1).

Table 1 ^{129}I and ^{127}I in IAEA 375, NIST 4357 and FC-98 Seaweed

Reference	^{129}I (10^{-7} µg g^{-1})	^{129}I (10^{-3} Bq kg^{-1})	^{127}I (n = 4) (µg g^{-1})	$^{129}I/^{127}I$ (10^{-10})
IAEA 375 Soil				
Our value	2.63 ± 0.39 (n = 2)	1.75 ± 0.26 (n = 2)	1.74 ± 0.10 (n = 4)	~1510
Recommended value	-	1.7 (1.3 – 2.1)	-	-
NIST 4357 Sediment				
Our value	2.2 ± 0.5 (n = 3)	1.5 ± 0.4 (n = 3)	3.8 ± 0.1 (n = 4)	580 ± 60
Informative value	-	9 (6.0 – 12.0)	3.1	-
FC 98 Seaweed				
Our value(Osterc and Stibilj, 2008)	95400 ± 8600	63500 ± 5700	795.5 ± 10.7	120000 ± 14000
Fréchou et al., 2001 (n = 6)	-	61300 ± 10700 (53300 – 68300)	-	122000 ± 21000 (106000 – 136000)

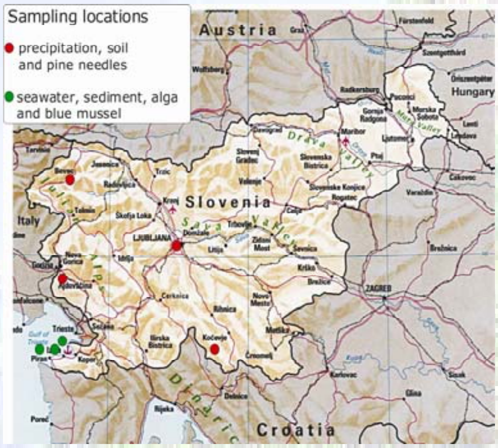

Figure 1 Sampling locations

4 Results and Discussions

4.1 ^{129}I levels in environment of Slovenia

The results found for analysed samples are summarised in Table 2 and 3.

1. Values found in analysed precipitation and soil samples are similar as values found in literature.
2. Concentrations of ^{129}I in soil samples from Japan are up to one order of magnitude lower than in analysed soil samples from Slovenia.
3. With exception of Quintana et al. (2000) there are no literature data on ^{129}I concentrations in pine needles. They determined ^{129}I levels in samples collected in the vicinity of nuclear power plants, so their values are elevated and higher in comparison to values we found for pine needles collected in Slovenia.
4. Values found in analysed seawater and sediment samples are in agreement with values found in literature for areas that are not under the influence of direct liquid discharges of ^{129}I from nuclear fuel reprocessing plants.
5. ^{129}I and ^{127}I concentrations found in analysed alga collected in 2005 and 2009 are in the same range. The ratio of $^{129}I/^{127}I$ found for alga *Fucus virsoides* is up to 10^{-9}.

Table 2 Range of ^{129}I and ^{127}I in terrestrial environment of Slovenia and other countries

Sample	Location	Number of samples	Year of sampling	Range			Reference
				^{127}I (µg g^{-1} dry weight)	^{129}I (10^{-9} µg g^{-1} dry weight)	$^{129}I/^{127}I$ (10^{-08})	
Precipitation	Slovenia	12	2009	0.0017 – 0.0065*	0.1 – 1.8*	7 – 91	This study
Soil		9	2009	11.6 – 27.6	82 – 1150	0.7 – 5.2	
Pine needles		2	2009	0.08 – 0.15	59 – 150	73 – 105	
Precipitation	Sweden	4	1998, 1999	0.0007 – 0.0052*	0.01 – 0.9*	1.6 – 17	Buraglio et al., 2001
Soil	Japan	10	not given	1.2 – 55.3	11 – 3900	0.02 – 12	Matsuzaki et al., 2007
	Sweden	5	1996, 2000	not given	45 – 386	not given	Hou et al., 2003
Pine needles	Argentina	9	not given	not given	1200 – 70400	not given	Quintana et al., 2000

*µg g^{-1} precipitation

Table 3 Range of ^{129}I and ^{127}I in marine environment of Slovenia and other countries

Sample	Location	Number of samples	Year of sampling	Range			Reference
				^{127}I (µg g^{-1} dry weight)	^{129}I (10^{-9} µg g^{-1} dry weight)	$^{129}I/^{127}I$ (10^{-08})	
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Sediment		6	2009	79.1 – 110.7	57 – 170	0.07 – 0.22	
		4	2005	59.3 – 76.8	72 – 256	0.12 – 0.33	
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	Japan	1	2006	0.049*	0.004*	0.01	Suzuki et al., 2008
Sediment	Baltic Sea	11	1997	41.6 – 75.9	11 – 740	0.02 – 1	Aldahan et al., 2007
Alga (*Fucus vesicolosus*)	China	2	1996, 1997	2850 – 2980	331 – 527	0.01 – 0.02	Hou et al., 2000
	Iceland	2	1988	330 – 364	135 – 210	0.04 – 0.06	Hou et al., 1999
	Denmark	5	1989 – 1998	256 – 504	131000 – 872000	4 – 34	Hou et al., 1999
	France	2	1998, 1999	297 – 354	2950000 – 6740000	1010 – 1940	Frechou et al. 2003

*µg g^{-1} seawater
**already published (Osterc and Stibilj, 2008)

5 Conclusions

To the best of knowledge, this work presents the first data for ^{129}I levels in the environment of this region of Europe.

Slovenia and the Adriatic Sea are isolated from direct liquid discharges of ^{129}I to the environment. The only possible discharges of ^{129}I to this environment are of atmospheric-precipitation origin, either coming directly from the atmospheric discharges or indirectly from the liquid discharges of ^{129}I to the ocean from which the iodine is transferred to the atmosphere and washed out to the marine and terrestrial environment of Slovenia by precipitation.

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