Environmental Science
Processes & Impacts

Accepted Manuscript

Volume 1 | Number 1 | Jan 2013 | Pages 1–100

Environmental Science
Processes & Impacts

Formerly Journal of Environmental Monitoring
www.rsc.org/jem

ROYAL SOCIETY OF CHEMISTRY

This is an *Accepted Manuscript*, which has been through the Royal Society of Chemistry peer review process and has been accepted for publication.

Accepted Manuscripts are published online shortly after acceptance, before technical editing, formatting and proof reading. Using this free service, authors can make their results available to the community, in citable form, before we publish the edited article. We will replace this *Accepted Manuscript* with the edited and formatted *Advance Article* as soon as it is available.

You can find more information about *Accepted Manuscripts* in the Information for Authors.

Please note that technical editing may introduce minor changes to the text and/or graphics, which may alter content. The journal's standard Terms & Conditions and the Ethical guidelines still apply. In no event shall the Royal Society of Chemistry be held responsible for any errors or omissions in this *Accepted Manuscript* or any consequences arising from the use of any information it contains.

ROYAL SOCIETY OF CHEMISTRY

rsc.li/process-impacts

Environmental Impact Statement

The Fukushima Daiichi nuclear power plant accident on 11th of March 2011 had released directly into the Pacific Ocean enormous liquid waste of ^{129}I and other fission isotopes that were subsequently dispersed eastwards. The paper reports on the determination of the nuclear plume impact on the West Cost of the USA that happened during April-July 2013. It was determined by use of the sensitive Accelerator Mass Spectrometry (AMS) method that measured the 129 –iodine long lived fission isotope concentration in the ocean water, close to the cove La Jolla - San Diego. The determined maximum 129I concentration increase was in an amount of more than 2 times greater than the concentration of the isotope measured offshore of Fukushima at 40km distance immediately after the accident.

Environmental Science: Processes & Impacts Accepted Manuscript

Journal Name RSCPublishing

ARTICLE

Environmental Science: Processes & Impacts Accepted Manuscript

Cite this: DOI: 10.1039/x0xx00000x

Received 00th January 2012,
Accepted 00th January 2012

DOI: 10.1039/x0xx00000x

www.rsc.org/

AMS analyses of I-129 from the Fukushima Daiichi nuclear accident in the Pacific Ocean water of the Coast La Jolla – San Diego, USA

C. Stan-Sion[a], M. Enachescu[a] and A. R. Petre[a],

The paper presents the results of an experimental study we performed by using the Accelerator Mass Spectrometry (AMS) method with Iodine 129 ($T_{1/2}$=15,7 My), to determine the increase of the radionuclide content in the USA West Pacific Coast waters, two years after the March 2011 Fukushima Daiichi nuclear power plant accident. The collection of the water samples took place between April-July 2013 at regular intervals of time, from the Pacific Ocean, at the Cove of La Jolla San Diego, USA. The results of the experiments showed an important increase of the radionuclide concentration during the late spring 2013. Compared to the isotopic ratio $^{129}I/^{127}I$, measured at 40 km distance, offshore of Fukushima and immediately after the accident, our results show an increase on the USA West Coast that was more than a 2.5 factor higher. Also, compared with the pre-Fukushima background values, our results show an isotopic ratio of about two orders of magnitude higher. A distinct feature of the reconstructed radioactive plume was that it traveled with a speed we estimated of 12 cm/s, which is consistent with the zonal speed in the Pacific. We coupled our ^{129}I results with the measurements from the June 2011 KOK cruise and we derived the levels of activity for ^{3}H and ^{137}Cs. On the USA West Coast, they were not exceeding the international regulatory limits.

1. Introduction

The nuclear pollution of the Planetary Ocean was produced by controlled or uncontrolled releases and by the contaminated fall out from the atmosphere produced by the nuclear weapons open tests. For many years now, the ^{129}I fission product has been used as a very good tracer of the environmental nuclear pollution [1]. The sensitivity level of this kind of nuclear monitoring using the radioiodine is dependent on the existing nuclear background level in the corresponding environments. The Atlantic Ocean has the highest contamination with 129-iodine because of the releases from the La Hague and Sellafield nuclear reprocessing plants that spread from the North Sea down to the Antarctica [2,3]. On the other side, the Pacific Ocean has an overall lower contamination with 129-iodine because of the lack of reprocessing plants with direct releases and because the Kuroshio Current acts as a southern barrier for the transport of radionuclides between the two oceans.

The present concern is whether this situation changed in the Pacific Ocean after the 11th of March 2011 Fukushima Daiichi nuclear power plant accident, caused by the great Tohoku earthquake and the followed up tsunami [4-7]. As a result of this event, large amounts of radionuclides, including the 129 radioiodine, were emitted into the atmosphere or were released by direct discharges from the nuclear power plant (NPP) into the Pacific Ocean [8-11]. From the lower atmosphere (troposphere), the fall out of nuclear pollution has contaminated not only the area near the NPP, but extended to a large area of eastern Japan. Experimental efforts have been done [12,13] to investigate soil samples and establish maps of radionuclide contamination on soil [14]. The time evolution of radioiodine isotopes in the atmosphere was investigated by Xu et.al. [15] who measured precipitation long-term samples, collected for about 2 years after the accident. Same scientists [15] have determined after the NPP accident, an increase of the ^{129}I concentration of nearly 4 orders of magnitude. It has been suggested [16] that Fukushima NPP caused the largest-ever direct release of anthropogenic radionuclides into the Pacific Ocean.

Although several attempts have been made to quantify the total amount of these radionuclide releases, there is still considerable uncertainty related to their duration and intensity. As the nuclear debris passed over the Izu-Ogasawara islands, due to the vigorous Kuroshio Current and to the Kuroshio extension, a long-term advection dispersal started eastward into the NW Pacific Ocean.

For about one year after the accident, observations over the radionuclides from the surface water were carried out by 17 cruises made by cargo ships and also by several research vessel cruises. Important results regarding the increase of the ^{129}I concentration in the ocean after Fukushima accident were obtained by measurements of samples collected by the Ka’imikai-O-Kanaloa (KOK) international expedition carried out in June 2011 [17]. Other hundreds of water samples were systematically measured from the Pacific Ocean until 2012 by [18,19] showing that the radioactivity level remains in the background level. Since then, all efforts to locate the radioactive plume traveling ahead in the ocean remained unsuccessful.

In the past, the ^{129}I ($T_{1/2}$=15.7 My) long life fission product provided valuable information about radionuclide transport in the oceans [20-23]. It was used as a long time evidence for the presence of other radionuclides like ^{131}I, ^{137}Cs, ^{134}Cs and ^{3}H. It is the best proxy for ^{131}I dosimetry reconstruction [24] and it was assigned by IAEA [1,25] as a sensitive fingerprint of nuclear pollution in the environment.

During the 11th of March 2011 accident, the Fukushima nuclear reactors discharged directly into the ocean 2.4-7 GBq of 129-iodine as liquid wastes [17], offering a good tracer of the nuclear plume propagation. ^{129}I, with its long residence time and relatively low bioavailability, in very small quantities, is a good conservative tracer of the ocean waters. However, the impact of a certain nuclear accident can be assessed via the atomic ratio ^{129}I/^{127}I only if the natural background level is known. Fortunately, the pre-Fukushima background in Japan was well investigated [26,27]. As a result, it has been discovered that the ^{129}I concentrations collected in June 2011 from the Western Pacific Ocean water samples by the, Tumey et. al [28] showed an increase of nearly three orders of magnitude as compared with the pre-Fukushima background levels.

The aim of our study that took place between April-July 2013 was to measure a possible impact of the radioactive releases from the Fukushima accident on the radionuclide level of the Eastern Pacific Ocean waters. Particularly, the Accelerator Mass Spectrometry (AMS) was the experimental method used to measure this possible elevation of the long-lived radionuclide level. The selection of the AMS method was done because it has the world’s highest sensitivity to measure such isotopes, by selecting and counting the nuclei one by one. ^{129}I concentrations were measured from the water samples collected at regular time intervals from the ocean water of the West Coast of the USA. The chosen location for sample collection was the Cove of La Jolla, San Diego. Equilibrium values of radionuclides in the ocean and possible increases were followed. The work reports two sudden increases of the ^{129}I/^{127}I isotopic concentration in the ocean water, which were observed at the end of spring 2013.

2. Sampling and measurements

The results presented here were obtained by analyzing the ocean water samples collected during a campaign lasting from April to July 2013, with a collecting frequency of 2 up to 4 days, at the location La Jolla, San Diego, USA. Geographically speaking, La Jolla is a cove wide open to the Pacific Ocean. The distance on the earth’s surface between Fukushima (37^0 14’ N; 141^0 01’E [29]) and La Jolla (32^0 51’N; 117^0 16’ W [30]) is 8770 km (see Fig.1).

Samples were collected in 3L tubes by volunteer students from the University of California, San Diego. Wearing the appropriate gear, in order to withstand the strong currents, equipped in diving suits, the students were swimming into the sea at distances of 15-20 m from the coast. In order to make it easy for filling when opened under water, before collecting, tubes were evacuated. After transporting the tubes at the shore, water was transferred into bottles.

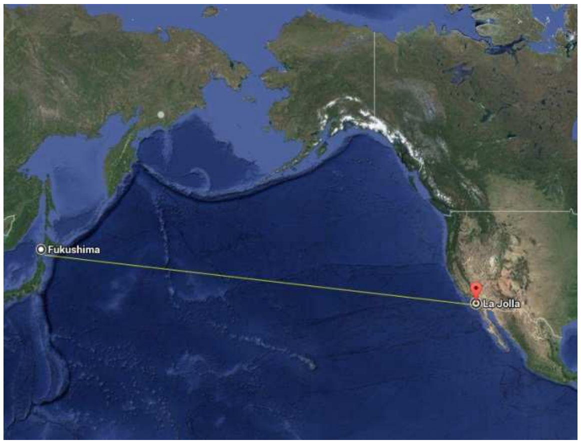

Fig.1: Partial map of the Pacific Ocean [30]. The distance over the earth’s Pacific Ocean of 8770 km, between Fukushima (37^{0}14’ N; 141^0 01’E) and La Jolla San Diego (32^0 51’N; 117^0 16’ W) is shown.

Prior of being transported to Europe for the ^{129}I AMS analysis, a number of 18 samples of unfiltered and not acidified Pacific Ocean water were collected and stored in 2L volume HDPE bottles with cap. Bottles were leached with hot distilled water and completely dried out before use. After that, they were completely filed with water and the cap was sealed with parafilm to exclude any exchange with the surrounding air. Finally, the samples covered in aluminium foil, packed into boxes and shipped to Romania. Samples were processed in the AMS chemistry laboratory of IFIN-HH Bucharest.

The amount of the stable isotope ^{127}I contained in the water samples was determined from aliquots extracted from the initial samples before any chemistry processing. Measurements of ^{127}I were done by the use of an ICP-MS system (NexION 300X, Perkin Elmer, Inc, USA). For the calibration, reference samples of ^{127}I were prepared in the chemistry laboratory and afterwards were used for the ICP-MS analysis. Results are shown in Fig.2 below. Each sample was measured three times and the relative standard errors were below 3%. The determined average value of iodine concentration was 57±1 µg L^{-1}, corresponding to 0.45±1 µM. The typical value for iodine in the open Pacific Ocean is also within few percent around 0.45 µM [31, 32].

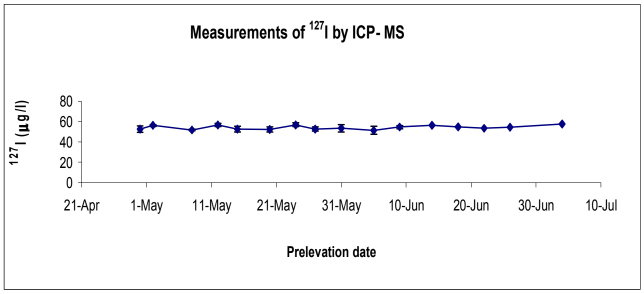

Fig. 2: ICP-MS results of the analysis of ^{127}I content in the sea water of the Pacific Ocean collected 2013.

For the AMS analysis, the ocean water samples were filtered through paper filters. Then, for the preparation of targets we applied the method of carrier free extraction of ^{129}I introduced by Yiou et al. [33]. Therefore, no carrier material was added to the samples.

Environmental Science: Processes & Impacts Accepted Manuscript

This journal is © The Royal Society of Chemistry 2012

Water samples were transferred to a separation funnel. The dissolved inorganic iodine was reduced to iodide (I^-) by addition of sodium sulfite and of hydroxylamine hydrochloride and then oxidized to molecular iodine by addition of nitric acid and sodium nitrate. The molecular iodine was extracted into carbon tetrachloride (now replaced by chloroform) and then, back-extracted into an aqueous phase by adding NaOH and water and agitating strongly. As a final step prior to obtaining the final AMS samples, I^- is precipitated in the form of AgI, then completely dried out at 80^0 C and mixed with the conductive material Nb powder (1:3). By this procedure, the 2 litres of collected water for each sample provided enough iodine material to produce 2 or 3 cathode samples of ca. 3 mg of AgI, each.

For the ^{129}I analysis, AMS measures the isotopic ratio $^{129}I/^{127}I$. In this respect, the 1 MV Pelletron HVEE machine of IFIN-HH Bucharest (see Fig.3) was used at terminal voltage of 1MV, selecting the 3+ charge state for the final acceleration and for the final analysis that is done on the high energy side of the machine [34]. ^{127}I iodine currents were measured in both, the Faraday cup after the low energy analyzing magnet and in the movable Faraday cup after the acceleration tank in "bouncing mode". The currents of $^{127}I^-$ selected by the first analyzing magnet were in the range of 30-40 nA.

The AMS measurement was done using the "bouncing mode" when, successively, the rare isotope is measured in the detector and then, the stable isotope is measured shortly in a Faraday cup placed of axis after the high energy analyzing magnet (see Fig.3, point 7). After each run, to correct for variations of the ion beam transmission through the tandem the $^{127}I^-$ current was measured in a Faraday cup at the entrance of the tandem accelerator (Fig.3, point 4).

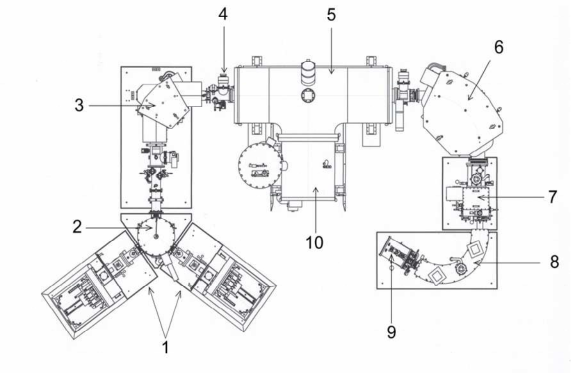

Fig.3 The general layout of 1 MV HVEE AMS facility of IFIN-HH, Bucharest: 1) Two ion beam injectors equipped with SNICS ion sources of type SO110–50 sample carousel HVEE and focusing ion lenses; 2) Electrostatic switching system; 3) Injection Magnet with multi beam switcher (bouncer); 4) Faraday cup and Q-Snout device; 5) 1MV tandetron accelerator, with gas stripper channel; 6) Analyzing Magnet; 7) Three offset Faraday cups 8) Electrostatic Analyzer; 9) Particle detector; 10) Cockcroft-Walton type HV power supply.

Each sample was measured several times (4-6 runs) and each measurement-run was divided in a counting process of *n* blocks of 30 second each, until 4000 counts were reached. In this way, the mean value had a fixed statistical error of 1.6% and the relative standard deviation between the 4-6 similar samples varied between 4% and 6%. The background sample was measured after each reference sample. Both were provided via HVEE from the Iso Trace Laboratory [35]. The standard sample had a value of 1.3×10^{-11} for the $^{129}I/^{127}I$ ratio. The blanks were prepared following the entire sample preparation procedure and in these experiments all $^{129}I/^{127}I$ ratio values were below 4×10^{-13} (process blank).

Fig.4 presents the results of the AMS analyses of the ^{129}I content in two of the collected water samples, one from the La Jolla San Diego location before the plume impact (background level) and the other during the impact on the West Coast from the 18th of June 2013. Each count in the spectra represents the response of the detector system to a final analyzed ion. Counts in the frames correspond, solely, to 129-iodine ions. The spectra were chosen to show the difference in counting statistics between a contaminated and a rather clean water sample from the Pacific Ocean.

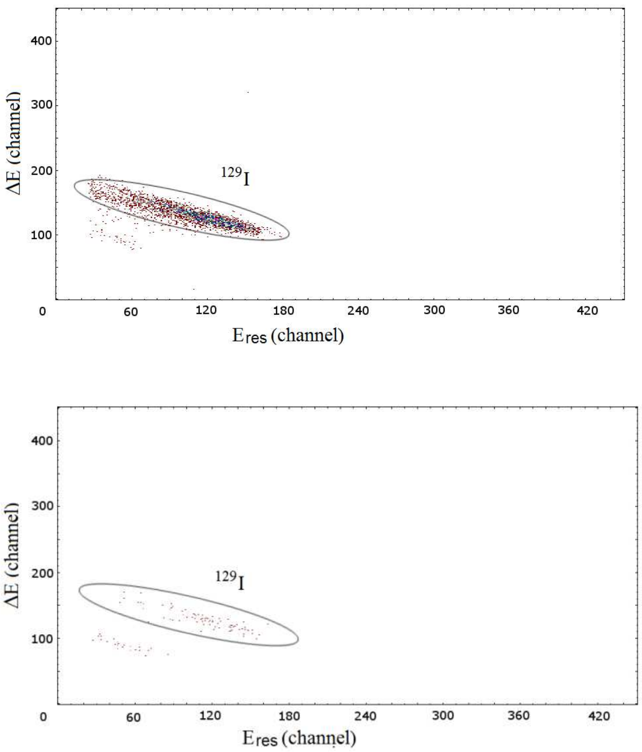

Fig.4: AMS bi-parametric spectrum E_{res}–ΔE of ^{129}I ions measured for the most intense impact on 18th June 2013 (upper picture) and of the background values (lower picture) before the impact at the La Jolla, San Diego location on the USA West Coast.

The bi-parametric spectrum E_{res}–ΔE, as shown in this figure, is typical for presenting graphically the responses of the AMS detector system. The rare isotope ions are identified in a large gas-filled ionization chamber with two anodes for ΔE-E_{res} measurement. A 75-nm-thick Si_3N_4 entrance window separates the detector gas from the beam line vacuum. The separation of ions species is based on the difference between their stopping powers. The optimum separation is achieved when the crossing of stopping power curves of ions happens exactly at the splitting boundary between the electrodes. The E_{res}–ΔE spectrum enhances visually/graphically the separation between ion species.

3. Results and Discussion

Each nuclear event, which produces even a very small increase of the radioiodine level, can be detected and assessed by AMS if the pre-event background level is known. Our exploratory measurements on the USA West Coast started on samples collected at the

Environmental Science: Processes & Impacts Accepted Manuscript

beginning of 2013. The lowest ^{129}I concentrations that we measured had values between (0.6–2) x 10^7 atoms L^{-1}. Such values correspond to the equilibrium concentration of iodine in the Pacific Ocean water, offshore from San Diego, La Jolla and represent good background values. These results were in accordance with the IAEA 97 measurements in the NW Pacific Ocean, that have been obtained for the iodine surface sea water concentration of 1,5x10^7 atoms L^{-1} [36]. The lower values we obtained are probably due to the collection of samples from the shore of La Jolla (ca. 15-20 m away from the coast), where strong waves are mixing surface and subsurface concentrations, dilution is rapid and where on shore deposition is also possible.

The collection time for samples was chosen to start in the early spring 2013, according to assumptions of different transport models. However, none of these models made a precise prognosis of the impact on the West Coast of the USA. Therefore, initially, the collection time was programmed to last for long time, even until spring of next year, since for ^{137}Cs the impact had been foreseen to happen in April 2014 [37].

Our results, the Pacific Ocean ^{129}I/^{127}I isotopic ratio-time profile measured offshore of the Cove La Jolla, San Diego, USA, during spring of 2013, is presented in Fig. 5. Two high and distinct spike maxima are visible. They reveal the maximum concentration values of 1.2±0.1x10^9 atoms L^{-1} measured on May 24th 2013 and 1.7±0.1x10^9 atoms L^{-1} measured on June18th 2013, with 24 days in between.

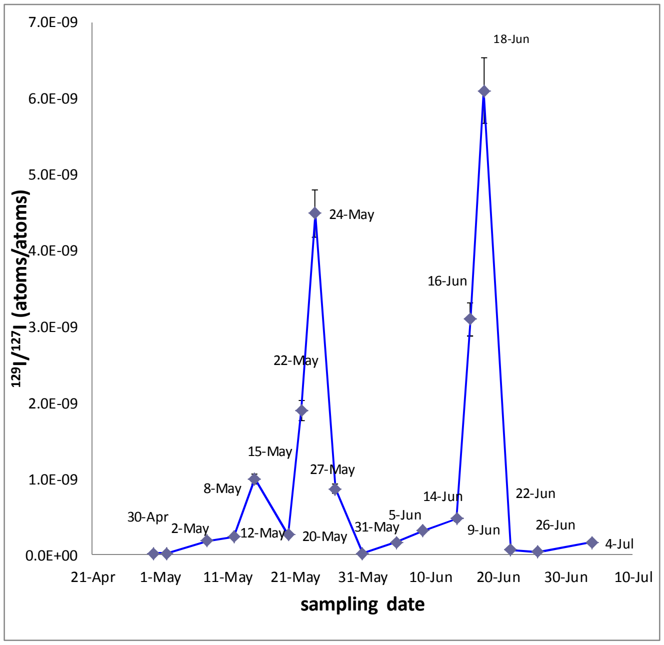

Fig.5: Results of the AMS measurements on ^{129}I/^{127}I concentrations in water samples collected from the USA West Coast, La Jolla, San Diego, 2013.

Both peaks occurred in the measurement spectrum after a slow increase in concentration that started about 15-20 days before the main increase. The two observed maxima are slightly different in height but, the full width at half maximum (FWHM) is for both of about 4-5 days.

As already mentioned in section 1, the Ka'imikai-O-Kanaloa (KOK) international expedition carried out in June 2011 collected soon after the accident water samples from different locations in the vicinity of the Fukushima NPP. Those samples were used by different laboratories to determine the nuclear pollution introduced in the Pacific Ocean [17, 28, 38]. Samples collected at a distance of about 40 km away from the coast were measured by P. Povinec et al. [17] and have determined for ^{129}I a maximum concentration value of 6.2x10^8 atoms L^{-1} in the surface water of the ocean.

Taking into account this value as a reference value, then the maximum 129-iodine concentration reaching the USA West Coast was by 2.5 times stronger than in the contaminated ocean water offshore of Fukushima after the accident. If we compare it to the equilibrium value of ^{129}I concentration in the ocean water then, during the impact its concentration was about 100 times higher. For more clarity, Table 1 presents data of past and actual measurements characterizing the Fukushima pollution of the Pacific Ocean.

Table 1. Important values of 129-iodine concentrations measured in the Pacific Ocean at different time moments.

Water sample in the Pacific Ocean	^{129}I/^{127}I (atoms/atoms)	^{129}I (atoms/L)	^{129}I (Bq/L)
PO equilibrium value (IAEA 97)*	5,5 x 10^{-11}	1,5 x 10^7	2,1 x 10^{-8}
Lowest values -this work	2,2-3 x 10^{-11}	0,6-0,8 x 10^7	8,3 – 11,3 x 10^{-9}
40 km from Fukushima -after the accident**	2,3 x 10^{-9}	6,2 x 10^8	8,7 x 10^{-7}
Highest impact value -this work (2013)	6,1 10^{-9}	1,7 x 10^9	2,3 x 10^{-6}
Second impact value -this work (2013)	4,5 10^{-9}	1,2 x 10^9	1,7 x 10^{-6}
Values offshore La Jolla after impact -this work (2013)	1,7 x 10^{-10}	4,6 x 10^7	6,4 x 10^{-8}

*[36]
**[17]

Between 21st of March (Day 10) and mid-July 2011 around 2.7x10^{16} Bq of ^{137}Cs entered the ocean, 82% having flowed into the sea before 8 April [39]. Presumably, during this period of time, important quantities of ^{129}I were also released into the ocean.

In time, according to [40] on 4th of April 2011 (Day 24) TEPCO had announced the decision to discharge approximately 11.5 tons of water with low levels of radioactivity into the ocean. This discharge was claimed necessary to allow the treatment of more highly contaminated water that was collected in other locations at the plant. So, it might be that the two 129-iodine concentration maxima observed in Fig.5 correspond to the releases that were done in the first days after the disaster and in the discharge ordered by TEPCO. First release was on 11-12 March 2011 and the second major release on 4th or 5th of April 2011. That is after an interval of 24 days in accordance with the result presented in Fig.5.

Based on these observation we calculated an average zonal speed of the radioactive plume that travelled 8770 km (distance over the Earth's surface) between Fukushima and La Jolla, San Diego (from $37^0$14' N; 141^0 01'E to 32^0 51'N; 117^0 16'W). Considering the time between first radioisotope emission on 11-12 March 2011 and the moment when it reached the West Coast of the USA on 18th June 2011, we got a traveling time of 806 days and a zonal speed of about 12 cm/s. This speed value is consistent with the zonal speed of the surface current in the north of the Pacific Ocean of 4-16 cm/s [41]. An estimate for the dimension of the radioactive plume is that it should be about 800 km long, having two spikes inside at distance of

Environmental Science: Processes & Impacts Accepted Manuscript

This journal is © The Royal Society of Chemistry 2012

280 km from each other, moving in forward direction. A distinct feature of the radioactive plume propagation found by naval missions is that this stayed confined along the longitude 37^0 - 40^0 N in traveling towards the West Coast of the USA [18].

As presented in section 1, the ^{129}I radioisotope is well-known as a very good tracer for nuclear pollution and it can be used for retrospective monitoring and dosimetry reconstruction for other fission radioisotopes. This is possible because in a nuclear reactor the radionuclides are formed by nuclear fission and sub-sequent by neutron activation. The fission yields in a nuclear reactor core are well known. If the concentration of a fission radionuclide is measured, then the amount of other fission products can be calculated. The nuclear releases at the Fukushima NPP were steaming from the burnt fuel storing reservoirs on the reactor roof and from deliberate discharges into the ocean. Therefore, measurements of fission products can be performed on water samples extracted from the ocean. However, to avoid dilution of isotopes the sample collection should be done from locations close to the NPP and in short time after the accident.

Such an opportunity is offered by the measurements of P.P. Povinec et al. [17] performed on samples collected by the scientific KOK cruise carried out in June 2011. Samples have been collected at a distance of 40 km offshore of Fukushima and experiments measured the specific radioactive levels for ^{137}Cs, ^{129}I and ^{3}H situated between 0.002 - 3.5 Bq L^{-1}, 0.01- 0.8 μBq L^{-1} and 0.05 - 0.15 Bq L^{-1}, respectively. From this set of data we considered the maximum value of the 129-iodine radioactivity and compared it to the increased measured 129-iodine concentration values when the USA West Coast was reached on 18th June 2013. From the ratio of these two values and using it for the other isotopes we got the estimates of specific radio-activities for ^{137}Cs and ^{3}H during the impact. Results are 10.6 Bq L^{-1} for ^{137}Cs and 0.45 Bq L^{-1} for ^{3}H. A similar specific radioactivity of ^{137}Cs, of about 10 Bq L^{-1}, was measured from August 2011 to July 2012 in the vicinity of the FNPP1 site [18]. Unfortunately, there are no other reported measurements of these radioisotopes or of an increased ^{137}Cs specific radioactivity.

The ^{137}Cs impact on the USA West Coast was predicted by the transport model calculations of Rossi et al. [37] to happen in spring 2014. The same paper, mentions low radioactivity values, in the mBq L^{-1} range, for all other radioisotopes which were measured, corresponding to the equilibrium values in the Pacific Ocean [Error! Bookmark not defined.]. Such low values were also measured in our experiment for the pre- and post-impact moments and could be reached due to dilution and deposition on the coast after the initial impact, or due to measurements of collected water samples that were not intercepting the main body of the radioactive surface plume.

In this respect, are also the low levels of radioactivity, of about 2 mBq L^{-1} measured for ^{137}Cs by [17] from the water samples collected by the KOK cruise at 40 km from Fukushima. As was shown before, the radioactivity levels of these measurements were oscillating for all three radio-isotopes between a maximum and minimum, far away from each other. Similar results were obtained in the elaborated work of Aoyama et al. [19]. In this work hundreds of water samples were systematically measured from the Pacific Ocean until 2012. For all measured values of the water samples, the contamination with ^{137}Cs, including those collected after the Fukushima disaster, were not exceeding 10 Bq m^{-3} (0.01 Bq L^{-1}).

However, the ocean biota (Zooplankton) close to Japan and the fish caught in California waters in 2011 were containing in average specific radio-activities of 20 Bq/kg and 6 Bq/kg, respectively [38,42]. Such values are in agreement with the measurements from the water samples collected after the accident at 40 km away from Fukushima and with our estimates based on the impact values of ^{129}I. Such values could be reached after a primary dissipation of the released nuclear plume in the ocean water and are about 10 times higher than the average values of ^{137}Cs in the Pacific Ocean before the nuclear accident.

Conclusions

The radioactive release from the Fukushima accident that had happened on 11th March 2011 was detected by use of the 129-iodine tracer in the Pacific Ocean water on the USA West Coast, more than two years after the event. It was manifested by two spikes of high elevation of the 129-iodine content in the water samples measured by AMS. Once again, ^{129}I has proven to be very sensitive and well-established tracer for detecting nuclear pollution in the environment. The first increase of the ^{129}I concentration was measured on May 24th, 2013 and the second on June 18th, 2013. AMS measurements of ^{129}I were performed on ocean water samples collected regularly form off shore of the Cove La Jolla, San Diego, USA, and definitely have shown an increase of the radioactivity more than two orders of magnitude over the natural level of the Pacific Ocean before the accident.

Comparing with radioactivity levels measured few days after the accident in samples collected from the Pacific water, at 40 km away from Fukushima, the 129-iodine tracer values were 2.3 times higher in concentration. All these values are in accordance to the measurements of samples collected by the KOK cruise in June 2011 and measured by [17]. According to the present AMS measurements on ^{129}I/^{127}I concentrations the radioactive plume travelled with an average speed of about 12 cm/s, speed in accordance with the zonal current speed in the Pacific Ocean.

The maximum concentrations of ^{137}Cs and ^{3}H in the Pacific water were estimated using the measurements of ^{129}I on the West Coast of La Jolla. We provided an initial estimate on maximum of the activity for the two radioisotopes when the Fukushima effluent reaches the USA coast. Our results have showed that none of the two calculated radioactivity values exceed the regulatory limits. For ^{137}Cs, which is the most dangerous isotope on long term due to its long half-life ($T_{1/2}$=30.17y), the estimated value is 10.6 Bq L^{-1} and should be compared with the Japan regulatory limit of 100 Bq L^{-1}. However, this estimated value slightly exceeds the more restrictive US drinking water limit of 8-9 Bq L^{-1}, but lasted for only the few days during the impact on the West Coast of the USA.

Acknowledgments.
The authors would like to thank to the volunteer students of the University of California San Diego and specially to Johanna Micuda for their devoted activity in water sample collection in very difficult conditions.

Notes and references

[a] Horia Hulubei National Institute for Physics and Engineering (IFIN-HH) , Bucharest, Magurele, Romania.

† Footnotes should appear here. These might include comments relevant to but not central to the matter under discussion, limited experimental and spectral data, and crystallographic data.

Electronic Supplementary Information (ESI) available: [details of any supplementary information available should be included here]. See DOI: 10.1039/b000000x/

Environmental Science: Processes & Impacts Accepted Manuscript

[1] G. M. Raisbeck, F. Yiou, *IAEA-SM-354/40*, 1997.

[2] S. Orre, J. N. Smith, V. Alfimov, M. Bensten, *Env. Fluid. Mechanics*, 2010, **10**, 213-233.

[3] P. He, X. Hou, A. Aldahan, G. Possnert and P. Yi, *Scientific Reports* 2013, **3**, Article no.2685.

[4] Y. Morino, T. Ohara, M. Nishizawa, *Geophys. Res. Lett*., 2011, **38,** doi: 10.1029/2011GL048689**.**

[5] A. Stohl, P. Seibert, G. Wotawa, D. Arnold, J.F. Burkhart., S. Eckhardt C .Tapia, A. Vargas., T.J. Yasunari, *Atoms. Chem. Phys.*, 2012, **12**, 2313-2343.

[6] H. Kawamura, T. Kobayashi, A. Furuno, T. In, Y. Ishikawa., T. Nakayama, S. Shima, T. Awaji, *J. Nucl. Sci. Technol.,* 2011, **48**, 1349-1356.

[7] C. Estournel, E. Bosc, M. Bocquet, C. Ulses, P. Marsaleix, V. Winiarek, I. Osvath, C. Nguyen, T. Duhaut, F. Lyard, H. Michaud, F. Auclair, *J. Geophys. Res*. 2012, **117** doi:10.1029/2012JC007933.

[8] M. Chino, H. Nakayama, H. Nagai, H. Terada, G. Katata, and H. Yamazawa, *J. Nucl. Sci. Technol*., 2012, **48**, 1129-1134.

[9] D. Butler, *Nature*, 2011, **471,** 555-556.

[10] *Nuclear Safety Commission of Japan,* 2011.

[11] K. Buesseler, M. Aoyama, M. Fukasawa, *Environ. Sci. Technol.,* 2011, **45**, 9931–9935.

[12] T. Ohno, Y. Muramatsu, C. Toyama, K. Nakano, S. Kakuta, H. Matsuzaki, *Anal. Sci.*, 2013, **29,** 271-274.

[13] Y. Miyake, H. Matsuzaki, T. Fujiwara, T. Saito, T. Yamagata, M. Honda, Y. Muramatsu, *Geochem. J.*, 2012, **46,** 327-333.

[14] T. J. Yasunari, A. Stohl, R.S Hayano, J.F. Burkhart, S. Eckhardt, T. Yasunari, *Proc. Natl. Acad. Sci.*, U.S.A, 2011, **108**, 19530-19534.

[15] S. Xu, S. P. H. T. Freeman, X. Hou, A. Watanabe, K. Yamaguchi, L. Zhang, *Environ. Sci. Technol.* 2013, **47,** 10851-10859.

[16] P. Bailly duBois, P. Laguionie, D. Boust, I. Korsakissok, D. Didier, B. Fiévet, *J. Environ. Radioact.,* 2012, **114**, 2–9.

[17] P. P. Povinec, M. Aoyama. D. Biddulph. R. Breier, K. Buesseler, C. C. Chang, R. Golser, X.L. Hou, M. Jeskovsky. A.J.T. Jull, J. Kaizer, M. Nakano, H. Nies, L. Palcsu, L.Papp, M.K. Pham, P. Steier, L.Y. Zhang., *Biogeosciences*, 2013, **10**, 5481-5496.

[18] M. Aoyama, M. Fukasawa, K. Hirose, Y. Hamajima, , T. Kawano, P.P. Povinec, J.A. Sanchez-Cabeza, *Prog. in Oceanography*, 2011, **89**, 7-16.

[19] M. Aoyama, M. Uematsu,, D. Tsumune, Y. Hamajima, *Biogeosciences*, 2013, **10,** 3067-3078.

[20] F. Yiou , G.M. Raisbeck, Z. Q. Zhou, I.R. Kilius*, Instr. Meth. Phys. Res. Sect. B*, 1994, **92,** 436-439.

[21] G.M. Raisbeck, F. Yiou , Z. Q. Zhou, I.R. Kilius, P.J. Kershaw, *Radioprotection*, 1997, **32**, 91-96.

[22] H. N. Edmonds, Z. Q. Zhou, G. M. Raisbeck, F. Yiou, L. Kilius, J.M.Edmond, *J. Geophys. Res.*, 2001, **106**, 6881-6894.

[23] X. Hou, Y. Hou, *J. Anal. Sci. Technol*., 2012. **3**, 135-153.

[24] S. Xu, L. Zhang, S.P.H.T. Freeman, X. Hou, Y.Shibata, D. Sanderson, A. Cresswell, T. Doi and A. Tanaka, *Environ. Sci. Technol.*, 2015, **49**, 1017-1024.

[25] M. L. Roberts, M. W. Caffee, I. D. Proctor, UCLR-ID-128218, LLNL, 1997.

[26] G. Steinhauser, *Environ. Sci. Technol*. 2014, **48,** 4649-4663.

[27] C. Toyama, Y. Muramatsu, Y. Igarashi, M. Aoyama, H. Matsuzaki, *Environ. Sci. Technol.,* 2013, **47**, 8383-8390.

[28] S. J. Tumey, T. P. Guilderson, T. A. Brown, T. Broek, K. O. Buesseler, *J. Radioanal. Nucl. Chem.* 2013, **296,** 957-962.

[29] www.tageo.com/index-e-ja-v-08-d-m33711602.htm

[30] www.google.ro/maps/place/32%C2%B051%2702.0%22N+117%C2%B016%2718.0%22W/@32.8505551,-117.2716667,10754a,13.1y/data=!3m5!1e2!3m3!1s-jY8xDDWHKBE%2FSPtPvGJYrRI%2FAAAAAAAAOAY%2F6_AehX6RKVA!2e4!3e15!4m2!3m1!1s0x0:0x0?hl=ro

[31] G.T.F. Wong, *Rev. Aquatic Sci.,* 1991, **4**, 45-73.

[32] G.T.F. Wong, A.U. Piumsomboon, W. M. Dunstan, *Marine Ecology Progress Series,* 2002, **237**, 27–39.

[33] F. Yiou, K. Diel, F.X. Martin , G.M. Raisbeck, *Nucl. Instr. Meth. Phys. Res. B,* 2000, **172**, 395-398.

[34] C. Stan-Sion, M. Enachescu, D.G. Ghita, C.I. Calinescu, A. Petre, D.V. Mosu, M. Klein, *Nucl. Instr. Meth. Phys. Res. B,* 2014, **319**, 117-122.

[35] ISO TRACE LABORATORY, Toronto, ON M5A4K8 , 60 St. George, CANADA.

[36] P. P. Povinec, S.H. Lee, L. L . W. Kwong, B. Oregioni, A.J.T. Jull, W.E. Kieser, U. Morgenstern, Z. Top , *Nucl. Instr. Meth. Phys. Res. B*, 2010, **268**, 1214-1218.

[37] V. Rossi, E. Van Sebille, A. S.Gupta, V. Garcon, M. H. England, *Deep-Sea Res*. 2013, **1-80,** 37-46.

[38] K. O. Buesseler, S. R. Jayne, N. S. Fisher, I. I. Rypina, H. Baumann, Z. Baumann, C. F. Breier, E. M. Douglass, J. George, A. M. Macdonald, H. Miyamoto, J. Nishikawa, S. M. Pike, and S. Yoshida, *Proc. Natl. Acad. Sci.*, U.S.A, 2012, **109**, 5984-5988.

[39] http://www.irsn.fr/FR/Actualites_presse/Actualites/Documents/IRSN-NI-Impact_accident_Fukushima_sur_milieu_marin_11072011.pdf

[40] OECD-NEA , Rep.7.03.2012 Timeline for Fukushima Daiichi nuclear power plant accident, day 24, 2012.

[41] N. Maximenko, P. Niiler, L. Centurioni, M.-H. Rio, O. Melnichenko, D. Chambers, V. Zlotnicki, B. Galperin, *J. Atmos. Ocean. Technol*., 2009, **26**, 1910-1919.

[42] D.J. Madigan, Z. Baumann, N.S. Fisher, *Proc. Natl. Acad. Sci.*, U.S.A, 2012, **109**, 9483–9486.

Environmental Science: Processes & Impacts Accepted Manuscript