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Deciphering the measured ratios of Iodine-131 to Cesium-137 at the Fukushima reactors

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Abstract

We calculate the relative abundance of the radioactive isotopes Iodine-131 and Cesium-137 produced by nuclear fission in reactors and compare it with data taken at the troubled Fukushima Dai-ichi nuclear power plant. The ratio of radioactivities of these two isotopes can be used to obtain information about when the nuclear reactions terminated.

The current state of the troubled Japanese Fukushima Dai-ichi nuclear power plant is of considerable concern. The plant, on the east shore of Fukushima prefecture was first hit, at 14:46 pm local time on March 11, 2011, by the magnitude 9.0 earthquake on the Richter scale about 150 km east of Honshu, the main island of Japan, and then hit by a giant tsunami, exceeding 15m high, about one hour after the earthquake. Although three reactors (units 1, 2, 3), which had been on operation at the plant, were automatically shut down immediately after the earthquake, the electric power for cooling of the reactor system was knocked out by the tsunami. Uncontrolled decay heat from the fuel rods has caused serious trouble in containing the radioactive materials which had been accumulated in the reactors and in the spent-fuel rods in the cooling pools. Explosions of hydrogen gas, created by the reaction of the heated Zirconium fuel cladding with water vapor, took place at the buildings of the units 1, 3, and 4 reactors, severely damaging those buildings.

At the moment, only very limited information is available about the interior conditions of the reactor complex owing to the highly radioactive environment, with intense gamma rays from radioactive materials that apparently escaped from the reactors preventing direct human access for inspections. However, certain information has been released by the Tokyo Electric Power Company (TEPCO) [1] on measurements of the radioactivity at several monitoring posts near the troubled reactors and from a water sample taken from the spent-fuel cooling pool of the unit-4 reactor building.[2]

We call attention here to the significance of the relative strength of the reported radioactivities of different radioactive isotopes. In particular, we focus on the two radioactive isotopes abundantly produced by fission, the Iodine isotope I-131 and the Cesium isotope Cs-137; the former has the half-life of 8 days while the latter has 30 years. The ratio should decrease on the time scale of days after the termination of nuclear fission processes and hence may be used for measuring the age of the fission products, similar to the carbon dating method using the ratio of C-14 to C-12 in the remains of ancient life[3].

One crucial difference between the proposed iodine-cesium dating and the carbon dating is that while the initial condition of the latter is essentially fixed by the equilibrium C-14/C-12 ratio in atmosphere with respect to cosmic ray interactions, the initial condition of the former depend on the prehistory of controlled nuclear reactions in the reactor. Another potential problem, which does not exist in the carbon dating, is the effect of the different chemical properties of iodine and cesium[4], possibly reflected in the change of their relative

abundance depending on the environment they are found, for example in the water (fresh or sea), in the air, or in the aerosols[5].

With these caveats in mind, we first make a simple estimate of time dependent numbers of I-131 and Cs-137 nuclei produced from a uranium burning nuclear reactor. We assume that in an operating nuclear reactor, where criticality is sustained for chain nuclear fission reactions, these isotopes are produced with a constant production rate, while the produced isotopes decay exponentially with a constant decay rate. The number of I-131 isotopes created by a nuclear reactor which had been in operation from time t_i to t_f is then given at later time t by [6]

$$N_{\rm I}(t) = \int_{t_i}^{t_f} dt' f_{\rm I} \mathcal{N}_0 e^{-\lambda_{\rm I}(t-t')} \tag{1}$$

where $\mathcal{N}_0$ is the number of fissions per unit time, $f_{\rm I}$ is the fraction of the I-131 produced per fission, and the decay rate $\lambda_{\rm I}$ is given in terms of the half-life by $\tau_{\rm I} = \ln 2/\lambda_{\rm I} = 8$ days. The integration gives

$$\begin{aligned} N_{\rm I}(t) &= f_{\rm I}\mathcal{N}_0 e^{-\lambda_{\rm I}(t-t_f)} \frac{1-e^{-\lambda_I(t_f-t_i)}}{\lambda_{\rm I}} \\ &\simeq \frac{f_{\rm I}\mathcal{N}_0}{\lambda_{\rm I}} e^{-\lambda_{\rm I}(t-t_f)} \end{aligned} \tag{2}$$

where we have assumed that the working time of the reactor is much longer than the half-life of I-131, $\lambda_{\rm I}(t_f - t_i) \gg 1$. The abundance of I-131 saturates on a time scale τ_I and remains constant during the operation of the reactor.

A similar estimate can be made for Cs-137. In this case, however, the decay of Cs-137 is very slow with a long half-life $\tau_{\rm Cs} = \ln 2/\lambda_{\rm Cs} = 30$ years; hence $\lambda_{\rm Cs}(t_f - t_i) \ll 1$ and we obtain,

$$\begin{aligned} N_{\rm Cs}(t) &= f_{\rm Cs}\mathcal{N}_0 e^{-\lambda_{\rm Cs}(t-t_f)} \frac{1-e^{-\lambda_{\rm Cs}(t_f-t_i)}}{\lambda_{\rm Cs}} \\ &\simeq f_{\rm Cs}\mathcal{N}_0 \Delta t e^{-\lambda_{\rm Cs}(t-t_f)} \end{aligned} \tag{3}$$

where $\Delta t = t_f - t_i$ is the time interval that the reactor has been in operation. The Cs-137 accumulates during the operation of the reactor due to its long lifetime, and hence becomes proportional to Δt.

The radioactivity of each of these elements measured in becquerel (Bq) is given by the decay rates

$$\Gamma_{\rm I}(t) = -\frac{dN_{\rm I}}{dt} = f_{\rm I}\mathcal{N}_0 e^{-\lambda_{\rm I}(t-t_f)} \tag{4}$$

and

$$\Gamma_{\rm Cs}(t) = -\frac{dN_{\rm Cs}}{dt} = f_{\rm Cs}\mathcal{N}_0\lambda_{\rm Cs}\Delta t e^{-\lambda_{\rm Cs}(t-t_f)}, \tag{5}$$

so that the ratio becomes

$$R_{\rm I/Cs}(t) = \frac{f_{\rm I}}{f_{\rm Cs}}\frac{\tau_{\rm Cs}}{\Delta t \ln 2}\left(\frac{1}{2}\right)^{(t-t_f)/\tau_{\rm I}}, \tag{6}$$

where we have again used the condition $\lambda_{\rm Cs}(t-t_f) \ll 1$. This formula gives the average ratio of the radioactivities of I-131 and Cs-137 contained in the fuel rods after the termination of the nuclear reaction at t_f.

The fractions of each fission product are $f_{\rm I} = 2.88 \times 10^{-2}$, $f_{\rm Cs} = 6.22 \times 10^{-2}$ for the cumulative thermal fission yields of U-235, taken from the IAEA data base[7]. For the three reactors in operation at the time t_X of the earthquake, the ratio at $t = t_X$ was

$$R_{\rm I/Cs}(t_X) = \frac{f_{\rm I}}{f_{\rm Cs}}\frac{\tau_{\rm Cs}}{\Delta t \ln 2} = \frac{240}{\Delta t} \tag{7}$$

where Δt is measured in months. If we choose $\Delta t = 7(12)$ months then $R_{\rm I/Cs}(t_X) = 34(20)$. On April 12, this ratio would be reduced by factor $2^{32/8} = 16$ to 2.1 (1.3).

In fig. 1, we show a semi-log plot of the ratio (6) against the number of days past X-day (March 11, 2011) with two different values for Δt; the upper line corresponds to $\Delta t = 7$ months and the lower line to $\Delta t = 1$ year. Note that the slope of these lines are fixed by the half-life of I-131; only the normalization changes with a change of Δt. The larger the value of Δt, the lower the position of the line. The blue dots are the data of the sea water samples taken at the South Discharge Channel, a monitoring post located 330m south of the Discharge Channel of the unit 1 - 4 reactor sites. The data is better fit by the lower line with $\Delta t = 1$ year, but the slope is a little steeper. As mentioned, the slope of the decay line is essentially fixed by the half-life of I-131, so this may at first look puzzling. The change of the slope may, however, well have been caused by the mixture of 9000 tons of old low level contaminated water which had been discharged from the Central Radioactive Waste Disposal Facility from April 4 to 10[8]. In these wastes I-131 had been completely depleted and only long half-life isotopes like Cs-137 were present.

Although we have ignored the effects caused by the chemical or physical differences of iodine and cesium in compound formation, ionization, diffusion, etc., the overall fit of the sea water data with the present theoretical calculation is very encouraging.

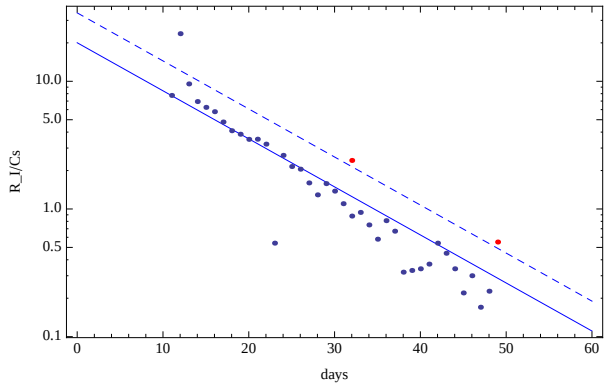

FIG. 1: Ratio of the radioactivities of I-131 and Cs-137 plotted against number of days since the earthquake (March 11, 2011). The blue solid line (Δt = 12 months) and the blue dashed line (Δt = 7 months) are the theoretical lines based on the formula (6); the blue dots are the measured ratios from the samples of sea water taken at the southern monitoring post. The red dots are the data of the water samples of the unit-4 reactor cooling pool.

We now turn our attention to the spent-fuel rods in the unit-4 cooling pool. Note that the unit 4 reactor was not in operation at the time of the earthquake. The same ratio for the fission products contained in the spent-fuel rods in the unit-4 cooling pool should be smaller by an extra attenuation factor

$$\left(\frac{1}{2}\right)^{(t_X - t_f)/\tau_{\rm I}} = 1/2^{90/8} \simeq 1/2400, \tag{8}$$

if all these fuel rods were intact since they had been removed from the unit-4 reactor three months earlier (December 2010). Since some of the fuel could be older, this ratio may be even smaller[9]. The calculated ratio is not, however, consistent with what has been reported. According to the TEPCO press release on April 14[2], the radioactivities of 220 Bq/cc of I-131 and 93 Bq/cc of Cs-137 have been detected from the water sample taken on April 12 from the unit-4 cooling pool, where more than 1500 spent-fuel rods were stored. The data was analyzed on April 13. The ratio 2.4 is closer to the values of the ratio of the radioactivities from the other reactors as shown by the red dot on the figure 1. [10]

If the data are correct, it would imply that at least some of the radioactive fission products found in the unit-4 cooling pool have been produced by nuclear reactions that took place at time t_X or later.[11] One possible explanation could be contamination by fallout of the fission products created in the neighboring reactors[12]. Another possible explanation could

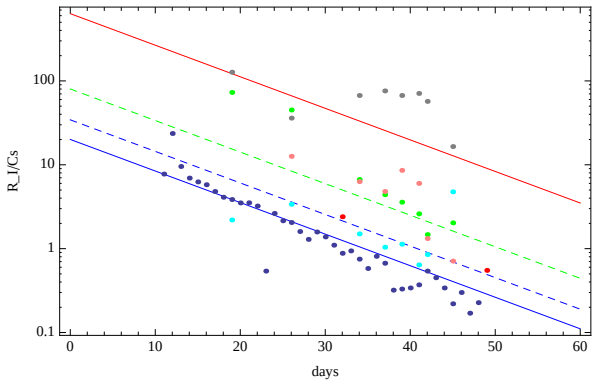

FIG. 2: Same plot as fig. 1, now including the data of the water samples taken from the sub-drains near the unit 1 (green), unit 2 (gray), unit 3 (cyan) and unit 4 (pink) reactor buildings. Two more theoretical lines are shown as a guide to the eye; they are computed from formula (6) with $\Delta t = 3$ months (dashed green) and formula (11) with $t'_f = t_X$ (solid red) which gives an upper bound if the fission ends on X-day.

be that a nuclear chain reaction was reignited in the melted used fuel in the unit-4 cooling pool for a certain period.

We now explore whether criticality had been re-established only briefly, in a period δt from time t'_i to $t'_f = t'_i + \delta t$. For simplicity in the following analysis, we assume $\delta t \ll \tau_{\rm I} = 8$ days. This would be reasonable since the heat produced by a nuclear reaction in a chunk of the melted fuel would cause rapid disintegration of the chunk, terminating criticality. In this case, the number of newly produced I-131 nuclei increases in proportion to δt:

$$\delta N_{\rm I}(t) \;\simeq\; f_{\rm I}\mathcal{N}_1 \delta t e^{-\lambda_{\rm I}(t-t'_f)}, \tag{9}$$

where $\mathcal{N}_1$ is the average number of nuclear fissions per unit time taking place during this brief time period. Similarly the number of newly produced Cs-137 nuclei is

$$\delta N_{\rm Cs}(t) \;\simeq\; f_{\rm Cs}\mathcal{N}_1 \delta t e^{-\lambda_{\rm Cs}(t-t'_f)}, \tag{10}$$

so that the ratio of the radioactivities of the newly produced I-131 and Cs-137 would be

$$R^{\rm new}_{\rm I/Cs}(t) = \frac{f_{\rm I}}{f_{\rm Cs}}\frac{\tau_{\rm Cs}}{\tau_{\rm I}} e^{-\lambda_{\rm I}(t-t'_f)} = 630 \times \left(\frac{1}{2}\right)^{(t-t'_f)/\tau_{\rm I}}. \tag{11}$$

This result with the replacement $t'_f \to t_X$ also gives an upper bound on the radioactivity ratio for the fission products created in the reactor under normal conditions before X-day for short Δt.

The radioactivity measured in the sample water taken from the unit-4 cooling pool could perhaps be from a mixture of the newly created radioactive fission products and the remnant of old fission products which is mostly composed of Cs-137, since I-131 has been depleted considerably, leading to the ratio 2.4 accidentally coincident with the ratio from the other reactors.

In any event, the amount of new fission products may be small since the absolute value of the measured radioactivity in the sample is not significant[13] and the overall data taken from the nearby sea water can be well fit by the older fission products, as shown in fig. 1. There is no indication so far that fresh fission products have been released into the sea water.

However, that the data from water samples taken from four sub-drains near the reactor buildings show even more puzzling features, as shown in fig. 2.[14] In particular, the water samples from the sub-drain near the unit-2 reactor building show an anomalously high radioactivity ratio.[15], even greater than the upper bound given by (11) if the nuclear fission ends on X-day as indicated by the red solid line in the figure. If there is no strong chemical filtering effect in draining contaminated water from the reactor buildings, it would be difficult to understand the observed anomaly near the unit-2 reactor without assuming that a significant amount of fission products were produced at least 10 - 15 days after X-day.

The data from the unit-3 sub-drain before April 23 sit close to the decay line which fits to the sea water data, hence they may be understood as due to radiation from fission products produced before the X-day. However, the data of the unit-1 sub-drain and unit-4 sub-drain give high radioactivity ratio, even larger than that of the samples from the unit-4 cooling pool and from the unit-3 sub-drain. The data therefore cannot be explained by the contamination of the old fission products which had existed in the spent-fuel rods in the unit-4 cooling pool. The new data of April 25, however, show very different characteristics which are difficult to be understood unless there was considerable mixing of waters belonging to different sub-drains due to in-flow and out-flow of water through underground water channels.

In conclusion, the ratio of the measured radioactivity of I-131 and Cs-137 may be used to extract useful information about when these fission products were produced in the nuclear reactor complex of the Fukushima Dai-ichi plant. The sea water data is consistent with the expected radioactivity by the fission products which had been produced before the earthquake. The data of the water samples from the unit-4 cooling pool and from the sub-

drain near the unit-2 reactor show anomaly which may indicate, if they are correct, that some of these fission products were produced by chain nuclear reactions reignited after the earthquake.

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[1] http://www.tepco.co.jp/en/index-e.html

[2] http://www.tepco.co.jp/en/press/corp-com/release/11041412-e.html

[3] W. F. Libby, E. C. Anderson, J. R. Arnold, Science **109** (1949) 227. See also E. Fermi, *Nuclear Physics* (Univ. Chicago Press, 1949) pp. 220.

[4] This problem can be avoided by using Cs-134 and Cs-137. However, Cs-134 has a half-life of 2 years which is too long to see observable effects in the currently available data.

[5] It is known that most of Iodine isotopes are released into the coolant water in the form of CsI salt which then dissolve into the water by forming Cs^+ and I^- ions. However the amount of cesium isotopes release into the water is an order of magnitude greater than that of the iodine isotopes. See E.C. Beahm, C. F. Weber, T. S. Kress,*Iodine Forms in LWR Severe Accidents*, NUREG/TM-11861, Oak Ridge National Laboratory (1991).

[6] This result may be obtained by integration of the equation, $dN_{\rm I}/dt = f_{\rm I}\mathcal{N}_0\theta(t;t_1,t_f) - \lambda_{\rm I}N_{\rm I}$, where $\theta(t;t_i,t_f) = 1$ for $t_i < t < t_f$ and $\theta(t;t_i,t_f) = 0$ otherwise, with the initial condition $N_{\rm I}(t_i) = 0$.

[7] INDC International Nuclear Data Committee, *Handbook of nuclear data for safeguards: Database extensions, August 2008* , INDC(NDS)-534.

[8] http://www.tepco.co.jp/en/press/corp-com/release/11041507-e.html

[9] The data taken from the unit-2 skimmer surge tank near the cooling pool shows a very small I/Cs ratio of 0.0256 which may reflect mostly the value in the used fuel rods with perhaps with contamination by fresher fission products from the unit-2 reactor. http://www.tepco.co.jp/en/press/corp-com/release/betu11_e/images/110418e4.pdf.

[10] In figure 1, new data of the unit4 cooling pool, released on April 29,

(http://www.tepco.co.jp/en/press/corp-com/ release/11042909-e.html) is also shown. It sits close to the same decay line confirming the previous data.

[11] We have tacitly assumed that the solubility of iodine or cesium in water is not changed considerably by boric acid, which has been added to the water to suppress chain nuclear reactions,.

[12] This explanation was first suggested to the author by M. Ichimura and K. Yazaki, independently. Actually, the hydrogen explosions of the unit-4 reactor building took place on March 15 after the explosion of the unit-3 reactor building on March 14. However, the explosion on March 15 may have sent fallout from the hydrogen explosion of the unit-3 building on the roof of the unit-4 reactor building into the pool.

[13] The total amount of the radioactivity by I-131 contained in the unit 4 cooling pool, which normally contains about 1500 tons of water, may be estimated as $220 \times 1.5 \times 10^9 = 3.3 \times 10^{11}$ Bq which is small compared to the radioactivity of fresh fission products contained in the nuclear reactor, which is the order of 10^{18} Bq.

[14] Some of the sub-drain data can be found in
http://www.tepco.co.jp/en/press/corp-com/release/betu11_e/images/110331e18.pdf,
http://www.tepco.co.jp/en/press/corp-com/release/betu11_e/images/110417e9.pdf,
http://www.tepco.co.jp/en/press/corp-com/release/betu11_e/images/110419e11.pdf,
http://www.tepco.co.jp/en/press/corp-com/release/betu11_e/images/110421e15.pdf.

[15] The author thanks Gordon Baym for calling his attention to this point.