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Land contamination activity data interpretation from Fukushima Daiichi accident

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HIGHLIGHTS

- Radiochemical analysis of activity sources from land contamination.
- Calculation of activity release from unit 3.
- Analysis of origin of land contamination from units 1–3.

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ABSTRACT

Radioactive aerosols were released during the accident from the units 1–3 of the Fukushima Daiichi nuclear power station. Release paths were due to leakage from the containment through the reactor building into the environment, due to containment venting and due to the explosions of the reactor buildings of the units 1, 3 and 4. The release due to containment venting can normally be estimated by dose measurements in the release stacks but for the other release paths there is no direct dose measurement available. Dose measurements away from the reactors only showed the release of fission products, but necessarily cannot identify the source of the radioactivity. Using the ratio of the activity measurements of the Cesium nuclides Cs-134g and Cs-137 allows the comparison with the ratios examined in the spent fuel pools and also the turbine halls of the three destroyed reactor cores. From the analysis of more than 1600 activity measurements in the area north-west from the Fukushima Daiichi power plant it could be proved, that mostly the units 1 and 3 are responsible for the land contamination in this area. The local contamination measured at the Fukushima Daiichi site is mainly caused by aerosol release from unit 2 and the sea water contamination origins from all three of the degraded reactor cores.

1. Introduction

The radioactive isotope distributions of the land contamination in the area north-west from the Fukushima Daiichi power plant are used to determine the possible source of the deposited activity. The international OECD/NEA program BSAF phase 1 to investigate with best estimate severe accident codes the status of the three degraded reactor cores has been finished (BSAF, 2012). Phase 2 of this program has been launched for the calculation of the accident progression and the aerosol release from the three partially molten nuclear reactor cores on the Fukushima Daiichi site, into the reactor buildings (RB) and into the environment and also the distribution of the aerosols on the country side and the Pacific Ocean in January 2015. The different timing of the reactor accidents of the units 1–3 regarding the core melting, the aerosol releases due to containment depressurization and leakage and the reactor building destructions have to be taken into account together with the weather data like wind direction and velocity and also rain fall in the first days after the accident has started. From the dose rate measurements in the air during the accident progression it is not possible to examine the source of the radioactivity except if the measurement place is very close to the unit. The present work was executed to support this program with the analysis of measured activity data.

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Table 1
Important events of Fukushima Daiichi nuclear accident.

	Event	Date
	Earthquake	11.03.2011 14:46
Unit 1	Start of core uncovery	11.03.2011 16:50
	Initiation of water injection	12.03.2011 05:46
	Deflagration in reactor building	12.03.2011 15:36
Unit 2	Start of core uncovery	14.03.2011 16:20
	Initiation of water injection	14.03.2011 19:54
	Sound at suppression pool	15.03.2011 06:00
Unit 3	Start of core uncovery	13.03.2011 03:29
	Initiation of water injection	13.03.2011 09:25
	Deflagration in reactor building	14.03.2011 11:01
Weather data	Wind direction to west	15.03.2011 11:00
	Start of rain fall	15.03.2011 17:00

The ratio of the activity between the two Cesium nuclides Cs-134g and Cs-137 can be used to classify the source of the measured activity. Both nuclides undergo the same chemistry during the release paths and they have a different production history. The different production history (Cs-134g is a neutron capture product and Cs-137 is a fission product) causes a different ratio for every nuclear reactor and the same chemistry is responsible for the fact that this ratio will not change during release and transport. The dependency of the ratio on the burnup in the reactor core (Jäckel, 2015) allows the identification of the contributing units to the land contamination.

In Table 1 some important events of the Fukushima Daiichi nuclear accidents are shown. The latest core melt process was that from unit 2, where the water injection for stopping the degradation process starts in the evening on 14th of March (Tanabe, 2011) about one day before the rainfall started north-west from the Fukushima Daiichi power station (RJSF, 2011). A large release of radioactive aerosols probably from unit 2 was observed in the early morning hours of 15th of March in parallel to the deflagration in the unit 4 reactor building (GRS, 2011). Until this time the degradation of the reactor cores 1, 2 and 3 should have been stopped due to the cooling water injection. The wind changed to the west and north-west direction at about noon on 15th of March and the rainfall started at the evening of this day. The aerosols transported with the wind come from different release paths, namely the explosions from the reactor buildings, the leakages from the containments and the depressurization of the containment through the wetwell.

During the time between start of fission product release into the reactor building and the explosion of the upper part of the building the aerosols undergoes different processes which reduces the airborne fission product density inside the building. The aerosols are transported into the reactor building together with steam and hydrogen. Due to agglomeration and sedimentation part of the aerosols will settle down on the floors of the building. Steam condensation on the relative cold walls will transfer part of the aerosols to the wall surfaces due to thermophoresis and diffusiophoresis.

At the explosion of the reactor buildings part of the deposited aerosols from the floor will be resuspended and released into the environment due to the large mechanical forces (pressure wave). The aerosols deposited on the walls will also be resuspended and the still airborne aerosols will be surely released into the atmosphere. The explosion transports the aerosols several hundred meters high into the atmosphere because of the high temperatures following the hydrogen deflagration and also the mechanical forces. These aerosols will undergo again sedimentation and agglomeration in the air, but the agglomeration process is much less important because of the much lower particle density in the upper atmosphere compared with the particle density in the closed reactor building. The sedimentation is a very slow process as the aerosols are small but rainfall will add the washout effect, this means aerosols will be collected from the water droplets falling through the aerosol clouds. This washout effect seems to be the major contributor for the land contamination in the north-west area of the Fukushima Daiichi nuclear power station (Renaud et al., 2004).

2. Land contamination analysis

The activity measurements far away in the contaminated area north-west from the Fukushima Daiichi power plant (IAEA, 2012) can deliver information of the source of the activity, if the ratio of the activity of the Cesium nuclides Cs-134g and Cs-137 is calculated (Fig. 1). The mean value of the ratio of more than 1600 data points was fitted by an EXCEL sheet using the AVERAGE and STD-DEV function for the decay corrected nuclide ratios. The half-life of Cs-137 is 30.17 years and of Cs-134g is 2.06 years, respectively. The average value of the nuclide ratio at the time of start of the Fukushima accident (time of earthquake) is calculated to be 0.95. The standard deviation was calculated to be 0.05. These values are shown in Fig. 1 as average value (black) and as upper (red, average + std-dev) and lower (green, average–std-dev) bound.

In a recent work (Jäckel, 2015) the activity ratios of the spent fuel pools (SFP) of Fukushima Daiichi units 1–4 were investigated and also the nuclide ratio from the turbine halls could be found (ENSI, 2012). From this data the nuclide ratio of the four units were calculated taking the mean value of the SFP and the turbine hall measurements (Table 2). The mean ratio given in Table 2 is calculated back to the time of the earthquake, while the measured activities are the originally measured values at the date of the measurement.

The perfect estimation of the mean ratios calculated for unit 3 and unit 4 confirms the assumption that the activity of unit 4 turbine hall and SFP is originating from the degradation of the unit 3 reactor core. The values of the units 1, 2 and 3 can now be compared with the measured values from the north-west land contamination. In Fig. 2 the measured ratios are corrected for the decay of the radio nuclides Cs-134g and Cs-137 to the time of the earthquake. The calculated ratios for the units 1, 2 and 3 are presented as straight lines. It can be seen clearly, that the origin of the land contamination agrees well with the values from unit 1 and unit 3 as given in Table 2. For the spreading and scattering of the data it has to be explained that in none of the activity publications the uncertainties of the measurements were given, neither in the IAEA data bank nor in the TEPCO handouts. Also information's about the measuring times are missing. In standard gamma measurements the uncertainty is between 1.5% and 5%. If data are measured close to the detection limit the uncertainties can easily reach up to 100% which is the definition of the detection limit.

A projection of the corrected ratios into ratio bins with a bin width of 0.02 is shown in Fig. 3. This figure shows clearly that reactor unit 2 did not significantly contribute to the land contamination in the north-west area of the Fukushima Daiichi nuclear power station. In a rough data fit it was calculated that about 2/3 of the contamination could originate from unit 1 and about 1/3 from unit 3. This is in agreement with the assumption that the explosions of the two belonging reactor buildings are driving the aerosols high into the atmosphere for a farther distribution.

3. Analysis of Fukushima Daiichi site contamination

For a qualitative mass balance now the question rises, where are the radioactive aerosols released from the reactor accident of unit 2? The reactor building of unit 2 stayed intact during the reactor accident sequence and therefore no explosion could transfer the aerosols very high into the atmosphere. The most logical