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Evolution of radioactive dose rates in fresh sediment deposits along coastal rivers draining Fukushima contamination plume

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Measurement of radioactive dose rates in fine sediment that has recently deposited on channel bed-sand provides a solution to address the lack of continuous river monitoring in Fukushima Prefecture after Fukushima Dai-ichi nuclear power plant (FDNPP) accident. We show that coastal rivers of Eastern Fukushima Prefecture were rapidly supplied with sediment contaminated by radionuclides originating from inland mountain ranges, and that this contaminated material was partly exported by typhoons to the coastal plains as soon as by November 2011. This export was amplified during snowmelt and typhoons in 2012. In 2013, contamination levels measured in sediment found in the upper parts of the catchments were almost systematically lower than the ones measured in nearby soils, whereas their contamination was higher in the coastal plains. We thereby suggest that storage of contaminated sediment in reservoirs and in coastal sections of the river channels now represents the most crucial issue.

The Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident led to the release of large radionuclide quantities into the atmosphere^{1,2} and to the formation of a soil contamination plume across Fukushima Prefecture, in northeastern Japan^{3,4}. As radionuclides are strongly sorbed by fine particles, they are likely to be redistributed within the landscape and supplied to the rivers, in association with the mobilization and transport of soil and sediment particles by erosion processes and runoff⁵. Depending on the levels reached locally, radiations resulting from radioactive contamination lead to an external exposure threat depending on the spatial distribution of radionuclides and the duration of exposition⁶. In addition, the transfer of contamination to plants and animals may affect human activities in the region⁷. However, there is little information on the dispersion of initial radioactive deposits and their redistribution along rivers draining the main radioactive pollution plume in Fukushima Prefecture. The Japanese Ministry of Environment supervises the frequent collection of river bank sediment along most rivers flowing across Fukushima Prefecture, but those samples may only reflect changes in river sediment contamination in case of river overflow⁸. Furthermore, the earthquake and the subsequent tsunami led to the destruction of river gauging stations in the coastal plains, and background data (discharge and suspended sediment concentrations) were unavailable during the study period. Monitoring stations have only become operational again from spring 2013 onwards. To overcome this information gap, radioactive dose rates were measured during four fieldwork campaigns in soils and fine sediment that appeared to have been recently deposited (i.e., mud drapes on channel-bed sand) along rivers draining the main radioactive pollution plume. Campaigns were organized in November 2011, April 2012, November 2012 and May 2013 after the main hydro-sedimentary events (summer typhoons, spring snowmelt) that occurred in this region during the two years that followed FDNPP accident. We used local ground dose rate measurements to estimate whether fresh sediment drape deposits were more or less contaminated compared to local soils. We supported the interpretation of the dose rate measurements by calculating a sedimentary connectivity index that evaluates the potential mobility of sediment between hillslopes and the rivers.



Results

Initial distribution of radionuclides in soils and potential connectivity between hillslopes and rivers. We focused our investigation on two radionuclides (i.e. ^{134}Cs and ^{137}Cs) that will represent the largest threat for the population over the medium to long term given their rather long half-lives (2 yrs for ^{134}Cs and 30 yrs for ^{137}Cs). Other gamma-emitting anthropogenic radionuclides ($^{110\text{m}}\text{Ag}$ and ^{125}Sb) were detected during our surveys but because of their low activities⁹, their contribution to the global dose rates was considered to be negligible.

Significant wet and dry radiocaesium deposits occurred on 15–16 March 2011, leading to the formation of a strong contamination plume over a distance of 70 km to the northwest of FDNPP. The area where soil contamination exceeds 100 kBq m^{-2} of ^{137}Cs was estimated to cover ca. 3000 km^2 (Fig. 1). We focused our study on the coastal catchments (i.e., from north to south, Mano R., Nitta R., Ota R., Uda R. and Ukedo R. catchments) covering a total area of ca. 1000 km^2 and comprising the most contaminated part of the radioactive plume. Those systems drain to the Pacific Ocean from an upstream altitude comprised between 700–1055 m a.s.l.

Forests largely dominate the land use, especially in mountainous areas, and cover 69 to 85% of the total surface, depending on the catchment (Figure S1a). Cropland and built-up areas, which are the most sensitive to soil erosion and leaching of contaminated particles (and also the most problematic in terms of human exposure to radiations), are concentrated in valley bottoms across the mountains and in the coastal plains.

Cropland areas are therefore very likely to provide the main source supplying contaminated particles to the rivers, as shown by the highest values of the hillslope-to-river connectivity index calculated in those areas (Figure S1b). Paddy fields located in upper parts of Nitta and Mano River catchments are well connected to the thalweg and they constituted therefore an important supply of contaminated material to the rivers. In contrast, in the flat coastal plains of those catchments, large cultivated surfaces are poorly connected to the rivers. A distinct situation was observed in the Ota River catchment. In the upper part of this catchment, land use is dominated by forests that are much less erodible than cropland, but where the storage and migration of radioactive contamination after its initial deposition is

much more complex depending on the dominant tree species¹⁰. Furthermore, the higher slope gradients observed in this area may have led to the more frequent occurrence of mass movements and river bank collapse events in this area.

Rapid supply of contaminated material to the rivers. The first fieldwork campaign conducted in November 2011 followed the occurrence of violent typhoons (i.e. Man-On on 20 July and Roke on 22 September) in the region (Figure S2). Cumulative rainfall during the four most intense events that occurred during summer 2011 varied between 313–458 mm across the study area. Those erosive events led to intense soil erosion in upper parts of the catchments, with a selective export of fine particles concentrating the contamination. This explains the higher dose rates measured in sediment drape deposits found in rivers compared to the ones measured in nearby soils (Fig. 2a).

In the coastal plains, typhoon-driven supply of contaminated material to the rivers is much less evident. It is also less problematic in terms of exposition to radiations and contaminant transfer, as this area was relatively spared by the initial radioactive fallout. This apparently low supply of contaminated material to the rivers may be explained by the much lower values of hillslope-to-river connectivity indices in those areas compared to the ones calculated for upper parts of the catchments (Figure S1). Those low values are controlled by the particularly gentle slope gradients observed in this area. Moreover, the well-connected areas of the coastal plains are systematically located close to the Pacific Ocean, and the material that they could have supplied to the rivers is likely to have been directly exported to the Ocean immediately after the typhoons.

Export of contaminated material from inland mountain ranges to the coastal plain. The differences in dose rates measured throughout the succession of fieldwork campaigns (Fig. 2a–d; Fig. 3a–d) indicate that the system is very reactive to rainfall and snowmelt events, and that export of contaminated material to the rivers, and possibly to the Pacific Ocean, has already been achieved during the year that followed the accident. The comparison of dose rate levels in soils and sediment drape deposits suggests that this dispersion of contaminated material towards downstream sections of the rivers was amplified by the spring snowmelt and the typhoons that

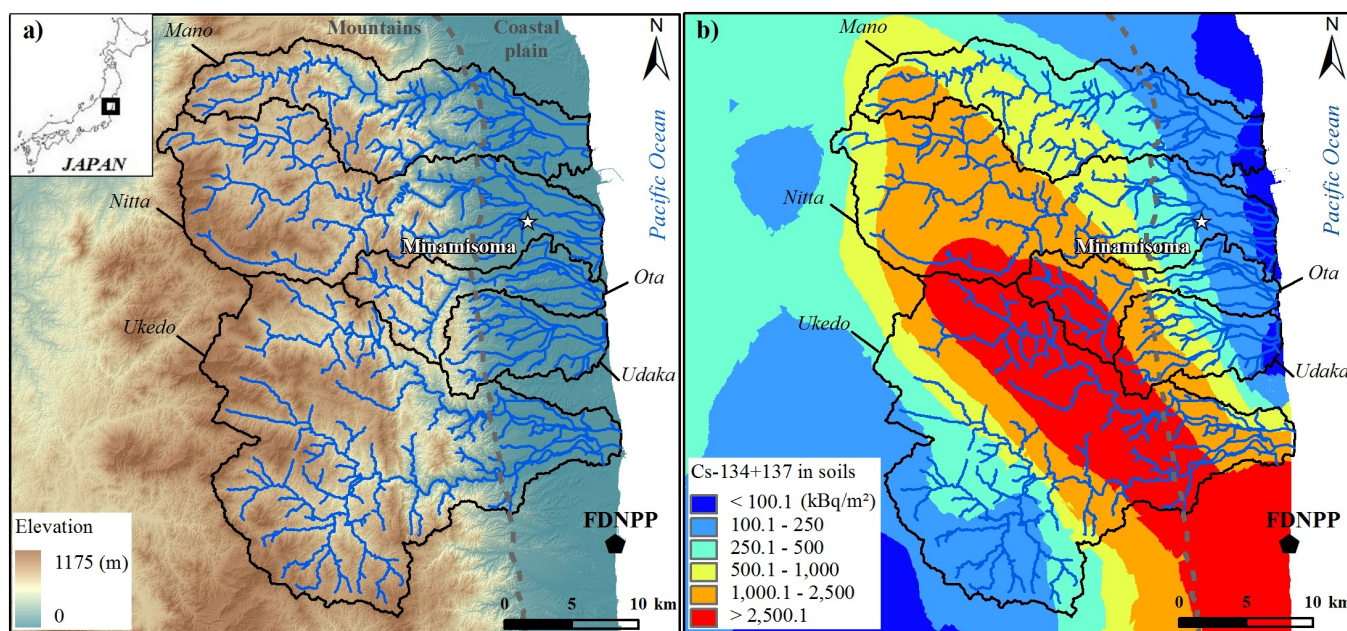


Figure 1 | Overview of the investigated contaminated catchments: (a) elevation map based on LIDAR monitoring (provided by GSI), and (b) contamination map for $^{134+137}\text{Cs}$. This original map was created using ArcGIS 9.3 software.

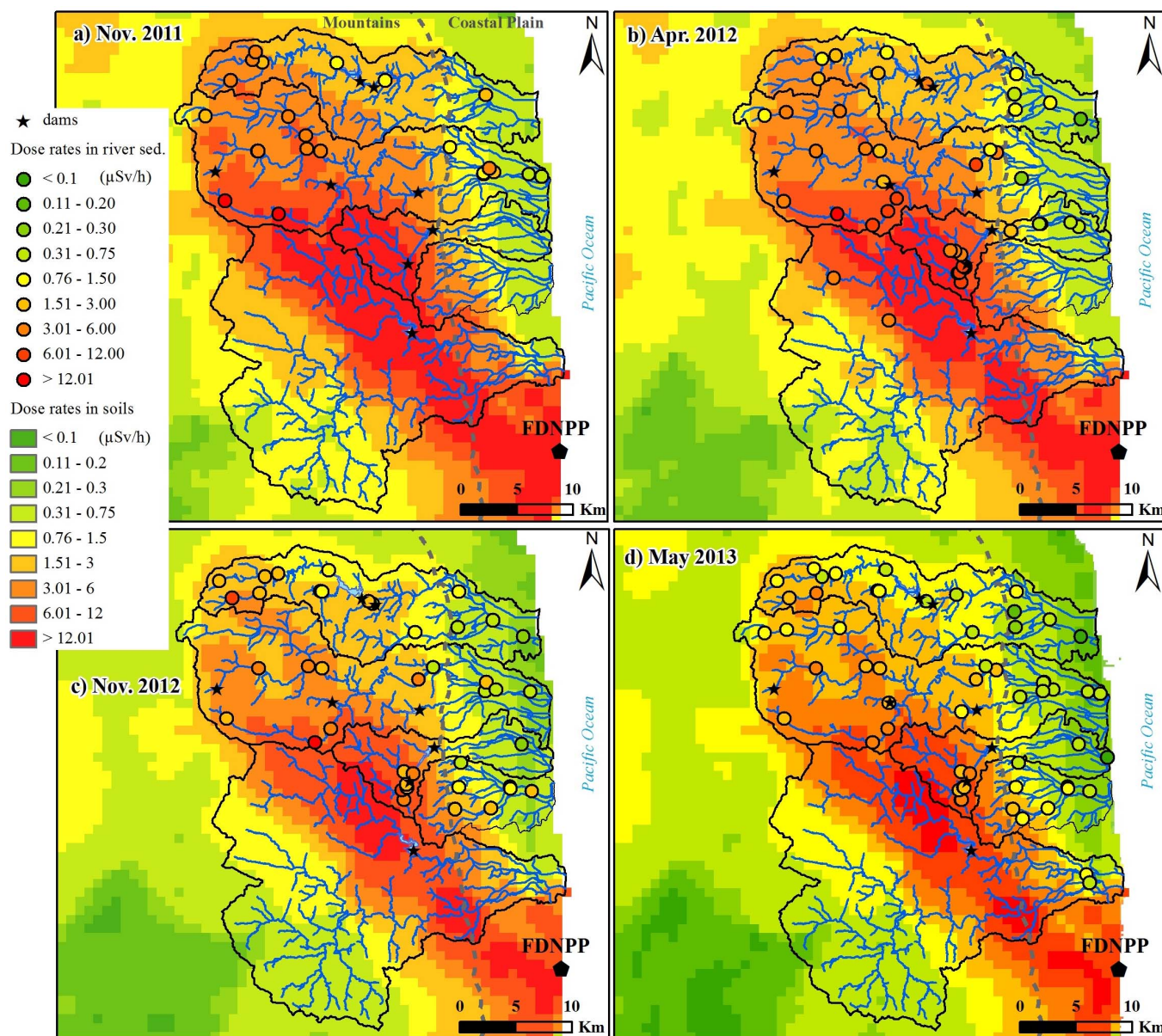


Figure 2 | Evolution of dose rates ($\mu\text{Sv h}^{-1}$) measured in soils (MEXT) and fresh riverbed sediment (TOFU project) along rivers draining the main contamination plume in (a) November 2011, (b) April 2012, (c) November 2012 and (d) May 2013. This original map was created using ArcGIS 9.3 software.

occurred in 2012. Summer typhoons may trigger intense soil erosion and supply contaminated material to the river, but this material would remain at least partly stored in the river channel, before being resuspended and exported to the coastal plains during the subsequent spring snowmelt. In upper parts of the catchments and during the last two field surveys (Fig. 2c–d; Fig. 3c–d), lower dose rates were almost systematically measured in sediment drape deposits than in nearby soils. Overall, doses rates logically decreased at similar measurement locations because of radioactive decay (mainly of ^{134}Cs) throughout the study period but, more interestingly, we observed a general relative decrease in dose rates measured in sediment drape deposits that may not be attributed to the single radioactive decay (Fig. 2d).

Impact of dams and reservoirs on dispersion of contamination.

The main dams are found in upper parts of Mano and Ota River catchments. Their reservoirs therefore constitute a potential sink of radioactive sediment originating from the most contaminated parts

of the catchments. Mano Dam was opened during the typhoons in 2011, leading to the propagation of contaminated sediment to the downstream sections of the river (Fig. 3a). This sediment was subsequently flushed by the snowmelt and replaced with sediment coming from local – much less contaminated – sources. A similar situation is suspected in the Ota River catchment, even though we could not get access to this area in November 2011. Nevertheless, previously published data⁹ showed that strongly contaminated sediment deposited behind the Ota River upstream reservoir between the accident and April 2012 (i.e. most likely by November 2011), which would indicate that a rapid export of contaminated material also occurred in this area, but that this material remained trapped in the reservoir. In contrast, in catchments where no dam is found (i.e., Uda and Nitta R. catchments), there has been a continuous supply of contaminated material from the upper parts to the downstream sections, even though storage of this material within the river channels is very likely because no extreme event occurred after the November 2011 typhoons (Figure S2).

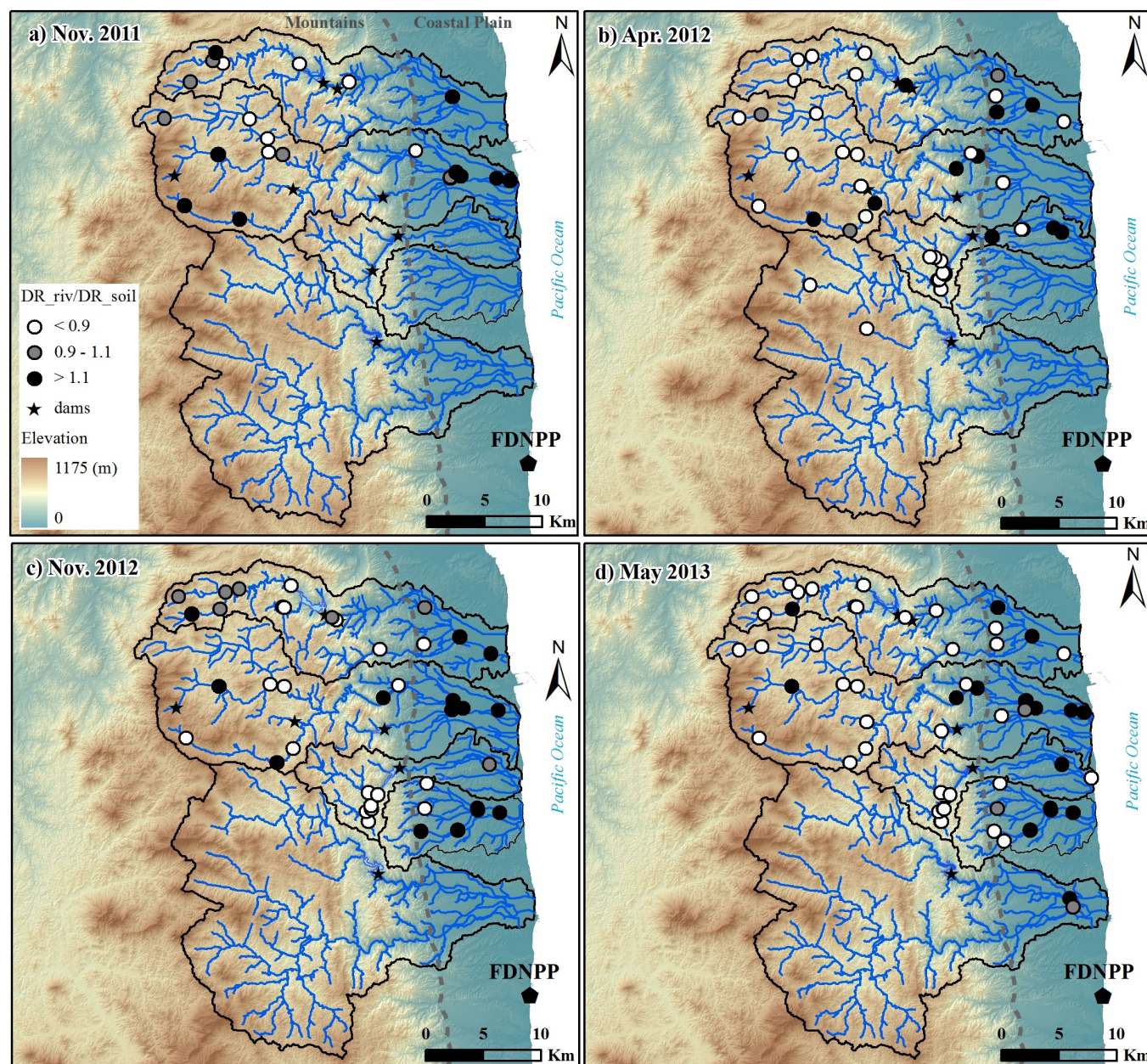


Figure 3 | Ratio between dose rates measured in fresh riverbed sediment (this study) and nearby soils (MEXT data) along rivers draining the main contamination plume in (a) November 2011, (b) April 2012, (c) November 2012 and (d) May 2013. This original map was created using ArcGIS 9.3 software.

Discussion

Our work relies on measurement of dose rates in soils and sediment drape deposits. This material consists of sediment freshly deposited in the river network that was shown to be representative of the suspended sediment that was conveyed by the rivers during the main previous hydro-sedimentary events in mountainous catchments exposed to heavy rainfall in summer¹¹. This alternative method of ‘contaminated sediment tracking’ provides the unique solution to address the lack of continuous river monitoring in this region during the first two years that followed FDNPP accident. Furthermore, the results presented in this study remain consistent with the ones obtained using measurements of ^{110m}Ag - ^{137}Cs ratios in soils and sediment of the Nitta R. catchment⁹, and outlining (1) erosion of contaminated particles in upper parts of the catchment and their supply to rivers as soon as by November 2011, (2) amplification of this export of material to the coastal plain during spring in 2012.

Overall, the percentage of sampling sites where dose rates measured in sediment drape deposits were significantly higher than in nearby soils (i.e., with a ratio > 1.1) evolved differently in mountainous upper parts of the catchments and in the coastal plains throughout the investigation period (Fig. 4). In mountains, this percentage was rather low in November 2011 (40%) and it subsequently decreased linearly during our study, down to 13% in November 2013. This likely reflects the low connectivity – except in areas located directly downstream of paddy fields (Figure S1) – between the sampling sites and the nearby hillslopes. Moreover, this connectivity has likely further decreased during our study, because of the regrowth of vegetation in abandoned farmland located in upper parts of the most contaminated catchments. In contrast, in the coastal plains, this percentage remained high from one survey to the next, with higher values recorded after the typhoons (73–83%) than after the snowmelt (56–63%) during which contaminated sediment was

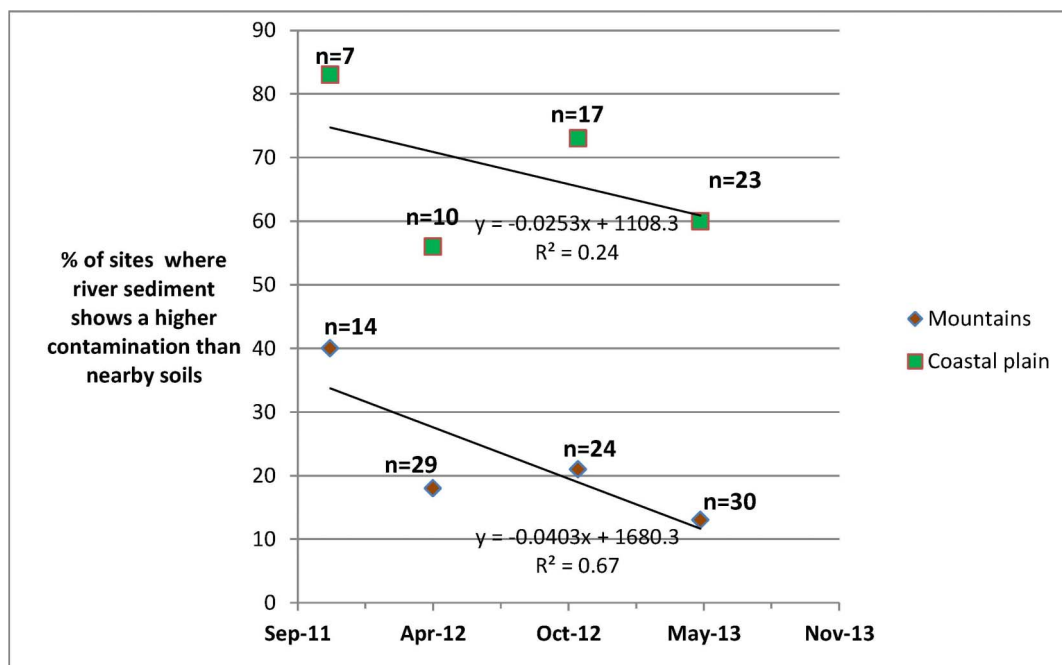


Figure 4 | Evolution of the percentage of sampling sites where dose rates on drupe sediment deposits were higher than on nearby soils between November 2011 and May 2013.

probably diluted with uncontaminated material originating from river bank collapse. However, the decreasing trend in contamination is less clear in the coastal plains than in the mountainous part of the catchments and it likely reflects a different behaviour depending on the fact that the river is dammed or not, with a continuous dispersion of contaminated sediment along the undammed Nitta River, whereas this process might only take place after the 2011 typhoons in the catchments equipped with a dam.

Our results outline the fast reactivity of river systems submitted to the succession of summer typhoon and spring snowmelt events, reactivity which is even quicker than previously expected. They also indicate that a significant part of contaminated material is currently stored in reservoirs and in coastal sections of the rivers and that it could not be remobilized after the 2011 events so far, the subsequent events having not been erosive enough.

Finally, our results have important management implications in the study areas. The impact of future dam releases should be monitored, as they could enhance the natural dispersion of contaminants in downstream sections of rivers, notably along Mano and Ota Rivers. Our findings should be considered carefully to manage fishing and recreational activities along the entire course of investigated coastal rivers and particularly in contamination storage areas. Finally, cultivation of floodable plains should be evaluated in light of our results to meet the requirements of the Japanese Food Sanitation Law¹² and to limit the contamination of the food chain.

Methods

Background maps of total cesium activity (¹³⁴Cs, ¹³⁷Cs) and corresponding dose rates from soils (¹³⁴Cs, ¹³⁷Cs) were computed based on data provided for a set of 2200 soil samples analyzed by the Japanese Ministry of Education, Culture, Sports, Science and Technology (MEXT) and corresponding to the situation of June 2011¹³. Dose rates from soils for November 2011 (Fig. 2a), April 2012 (Fig. 2b) and November 2012 (Fig. 2c) were corrected based on MEXT measurements (Fig. S3). Dose rates from soils in May 2013 (Fig. 4c) were decay-corrected from the ones measured in November 2012 by MEXT.

Topographic data were derived from a Digital Elevation Model (DEM) with a 10-m regular grid provided by the Geospatial Information Authority of Japan (GSI) from the Ministry of Land, Infrastructure, Transport and Tourism (<http://www.gsi.go.jp/>). Land uses were mapped based on a multitemporal and multispectral classification of images from SPOT-4 and SPOT-5 satellites. Differences of spectral properties (surface reflectances) between the different land uses of interest (i.e., forests, cropland,

built-up areas and surface water areas) allowed their spatial discrimination using a supervised classification methodology, under ENVI4.8 software. A hillslope-to-river connectivity index was computed to outline the spatial linkages and the potential connection between the sediment eroded from hillslopes by runoff and water erosion processes and the river channels^{14,15}. This landscape-based connectivity index combines two types of algorithms usually used separately in many geomorphological studies to assess hydrological and sediment connectivity within catchments, i.e., the upslope approach which integrates the morphology and land use of the drainage area, and the downslope approach which accounts for the flow path characteristics (length, slope, land use) that a particle has to follow from its eroded source to the nearest sink.

Fieldwork started in the Mano R., Nitta R. contaminated catchments that were already accessible in November 2011, and the study area was enlarged to the Ota R., Udaka R. and Ukedo R. catchments as soon as access to those areas was permitted by the Japanese authorities (between April 2012 and April 2013¹⁶). Campaigns were organized after the main hydro-sedimentary events that occurred in the region (November 2011, April 2012, November 2012, May 2013; Fig. S2). Radiation dose rates were systematically measured in the field using a NaI portable detector (Inspector 1000 INK 1KN-3, Canberra) and a radiometer (LB123 D-H10, Berthold Technologies) in fresh sediment drupe deposits and in nearby soils along rivers (i.e. in a 10 to 20 m wide area along rivers).

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Author contributions

O.E. and Y.O. designed research and wrote the main manuscript; C.C. conducted spatial analyses and field measurements; J.P., H.L., L.L., P.B. conducted field measurements and C.O. conducted satellite image analysis and interpretation. S.A. participated to the redaction and reviewed the manuscript.

Additional information

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