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Surficial redistribution of fallout ^{131}I in a small temperate catchment

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Isotopes of iodine play significant environmental roles, including a limiting micronutrient (^{127}I), an acute radiotoxin (^{131}I), and a geochemical tracer (^{129}I). But the cycling of iodine through terrestrial ecosystems is poorly understood, due to its complex environmental chemistry and low natural abundance. To better understand iodine transport and fate in a terrestrial ecosystem, we traced fallout ^{131}I throughout a small temperate catchment following contamination by the 11 March 2011 failure of the Fukushima Daiichi nuclear power facility. We find that radioiodine fallout is actively and efficiently scavenged by the soil system, where it is continuously focused to surface soils over a period of weeks following deposition. Mobilization of historic (pre-Fukushima) ^{137}Cs observed concurrently in these soils suggests that the focusing of iodine to surface soils may be biologically mediated. Atmospherically deposited iodine is subsequently redistributed from the soil system via fluvial processes in a manner analogous to that of the particle-reactive tracer ^7Be , a consequence of the radionuclides' shared sorption affinity for fine, particulate organic matter. These processes of surficial redistribution create iodine hotspots in the terrestrial environment where fine, particulate organic matter accumulates, and in this manner regulate the delivery of iodine nutrients and toxins alike from small catchments to larger river systems, lakes and estuaries.

beryllium-7 | fluvial transport | iodine cycling | particle tracer | radioiodine

The iodine isotope ^{131}I (half-life of 8.02 d) is a by-product of nuclear fission, is highly radioactive and acutely toxic, and presents a health risk (1) upon its release to the environment from fuel reprocessing or industrial accidents. But assessments of radioiodine exposure incorporate large uncertainties (2, 3) because iodine environmental behavior is not well understood (4, 5, 6). In the Earth near-surface, iodine exists in multiple oxidation states ranging from -1 to $+5$, which form inorganic (e.g., iodide, I^- , and iodate, IO_3^-) and organic (e.g., CH_3I) species in gaseous (e.g., iodine, I_2), particle-reactive or soluble phases, all with widely divergent chemical behaviors (7) and facile interconversions. The most important iodine species in terrestrial systems are “organoiodine” (5, 8), thought to be incorporated in aromatic moieties (9) but otherwise poorly characterized as to origin, reactivity, or fate. Coupled with the very low natural abundance of iodine (10), the complexity of iodine chemistry has historically challenged analytical capabilities (10, 7, 11) and continues to limit our understanding of diverse problems including the true hazards of nuclear energy production or amelioration of human iodine deficiency (12).

Following the M_w 9.0 Tōhoku earthquake of 11 March 2011 (13) and subsequent failure of the Fukushima Daiichi nuclear power plant (FDNPP), sufficient radioactive ^{131}I was released into the global atmosphere (14) to generate measurable fallout even at the great transport distance required to reach the northeastern United States. While regrettable, this event created an opportunity to observe the cycling of iodine across a natural landscape, as the radioactivity of ^{131}I , when measured by gamma spectroscopy, provides a highly sensitive iodine tracer. Two additional environmental radionuclides, if measured concurrently, provide a context for comparison of iodine behavior. Beryllium-7 (half-life

54 d) is a natural, cosmogenic tracer commonly used in particle- and contaminant-transport studies, is also atmospherically derived, and is known to be highly surface-reactive, to accumulate efficiently in surface vegetation and topsoil, and to associate irreversibly with fine particles in fluvial transport (15). Cesium-137 is another fission product released to the atmosphere in large quantities by the FDNPP disaster (14). The relatively long half-life of ^{137}Cs (30.3 a) has permitted detailed study of its fate in terrestrial ecosystems following historical nuclear events (16).

Here, we use gamma spectroscopic measurements of ^{131}I contamination, in conjunction with measurements of ^7Be and ^{137}Cs , to develop a broad perspective of iodine transport and fate in a terrestrial ecosystem. Our observations of ^{131}I , summarized in Fig. 1 and elaborated in the following sections, yield a catchment-scale view of iodine behavior that both complements and validates current interpretations of iodine environmental chemistry.

Results and Discussion

Experimental Setting and Sample Collection. At the onset of the FDNPP crisis we began a weekly regime of collection and analysis of precipitation, terrestrial, and fluvial-sediment samples in the Mink Brook catchment of Hanover, New Hampshire, USA ($43^\circ42'18''\text{ N}$, $72^\circ17'08''\text{ W}$; Fig. S1). Elevation of the Mink Brook catchment ranges from ca. 500 m above mean sea level in the headwaters to 120 m at its confluence with the Connecticut River. At the start of our sampling central New Hampshire was entering spring, which is a period of peak stream flow. Decimeters of snowpack remained at elevations above ca. 400 m, while terrain below ca. 200 m was generally snow free. Frequent rainfall amounted to 70 mm during the period March 11–April 11 and to 120 mm for the period April 11–May 11. On April 8, near the observed peak in local ^{131}I deposition and in the lee of a subsiding flood event, we sampled fluvial sediments at eight sites along the 15-km length of Mink Brook.

Stream sampling locations at each site included, in close juxtaposition, active channel sediments as well as surficial channel margin sediments which were deposited at the falling waterline of a recent high-water event (Fig. S1B). Suspended sediments were collected by filtration of 20 L stream water samples. Terrestrial samples, including vegetation, litter, soil, snowpack (where present), were collected concurrently at lawn, forest, and agricultural sites in the Mink Brook catchment. Multiday cumulative precipitation samples were collected at the Dartmouth College Observatory in an open collector. For all samples we measured, using gamma spectroscopy, fallout radionuclides ^{131}I and $^{134,137}\text{Cs}$, and naturally occurring ^7Be .

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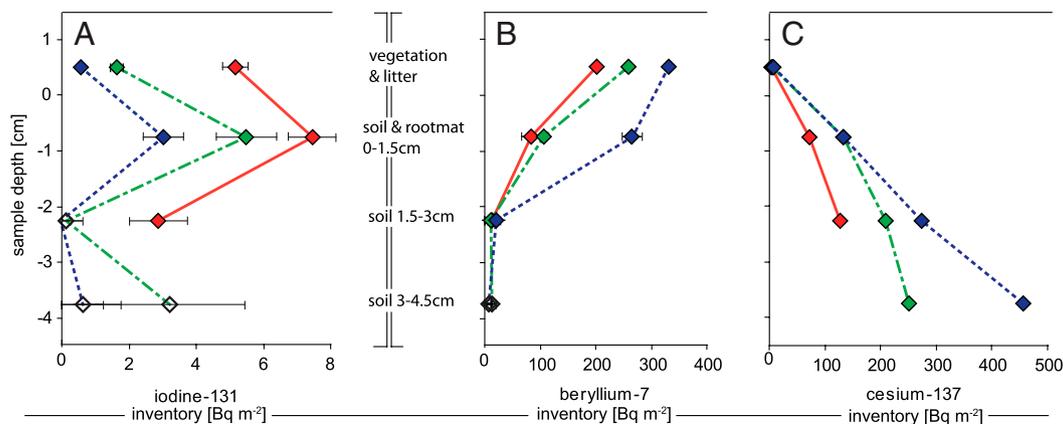


Fig. 3. Soil depth profiles of radionuclide terrestrial inventories at one site for a number of sampling times, for radionuclides: (A) ^{131}I iodine, (B) ^7Be beryllium, (C) ^{137}Cs cesium. Samples collected on 6 April 2011 are shown in red, 21 April in green, 5 May in blue. Open symbols indicate measurements did not satisfy a 2σ detection criterion. Error bars depict 1σ analytical uncertainty. ^{131}I peaks in surface soil and declines with time due to decay. ^7Be peaks in vegetation or litter and increases with time due to ongoing atmospheric deposition. ^{137}Cs is a bomb-test legacy from ca. 1965 (1), peaking at depth and increasing during the study period through biological activity (see text).

from subsurface to surface soil via either (ii) root uptake (22) or (iii) volatilization (23, 24) of iodine that, upon initial deposition, penetrated to soil depths greater than 2.5 cm. Details of the upward transport of volatilized iodine can include diffusion to surface soil and entrapment in the rhizosphere, or escape to the atmosphere with subsequent stomatal uptake (23) or redeposition from dew or fog (25) as in mechanism 1 above.

Of these possible explanations, secondary deposition from an undetected process (mechanism 1) is unlikely given the frequency and efficiency of rain in removing atmospheric iodine (18, 26). Mechanisms 2 and 3 are plausible; of the former we find additional evidence as follows. Radiocesium has previously been demonstrated to cycle seasonally in surface soils (27), entrained as a potassium analog by virtue of similar charge/mass ratios. As a consequence of biological activity it is actively retained in surface soils (28, 16). In our inventory collections, ubiquitous bomb-test-legacy ^{137}Cs (1) became enriched in subsurface soils over a period of weeks at our two uncultivated sites (Fig. 3C and Fig. S2). This cesium source is clearly distinguishable from recent FDNPP emissions by the noted absence of short-lived ^{134}Cs at depth. Instead, we attribute the increase in subsurface ^{137}Cs inventory to its translocation from a deeper reservoir (27) during the seasonal transition from winter dormancy to spring growth. Iodine is a micronutrient in its own right, widely implicated in soil microbial metabolism (29, 30). While the enrichment pattern or process may not be identical for iodine as for cesium, as the former is derived from a new, atmospheric source and the latter from an old, subsurface source, we propose that the active enrichment of iodine in surface soils or root mat is not unexpected (31).

Iodine Transport and Fluvial Redistribution. The focusing and storage of ^{131}I in a soil reservoir enhances its exposure to surficial transport processes such as mechanical erosion and overland flow. Accordingly, to track the postdepositional fate of ^{131}I , we sampled stream waters and sediments at various times throughout the Mink Brook catchment. While ^{131}I activity in filtered waters of Mink Brook was too low for direct gamma spectroscopic measurement ($<0.03 \text{ Bq L}^{-1}$), it was readily measurable in stream sediments. Indeed, in several locations, channel margin sediment ^{131}I inventories to a depth of 2 cm (thereby representing a *minimal* estimate) exceeded the corresponding terrestrial inventory by a factor of about two (Tables S2, S3). These observations confirm that, following atmospheric deposition, ^{131}I is redistributed by streams in association with a particulate phase.

The surficial redistribution of ^{131}I follows that of ^7Be , as evidenced by correlation of the two radionuclides across the variety

materials we have measured (Fig. 4A). Among terrestrial samples this correlation is a consequence of a common source for the radionuclides via precipitation, but also, we contend, of their shared particle-reactivity. The strength and persistence of the correlation between ^{131}I and ^7Be among channel margin sediments (Fig. 4A; $\text{adj } R^2 = 0.92, p < 10^{-4}$) demonstrates that the particle-reactivity of both ^{131}I and ^7Be remains stable during transport to the fluvial system and subsequent deposition. In this regard, despite radically different elemental chemistry, ^7Be is an effective tracer of ^{131}I redistribution and fate.

Commonality of the redistribution of ^{131}I and ^7Be implicates a common transport phase for the radionuclides. In particle size-fractions isolated from stream bed sediments by sieving, or from suspended sediments by filtration, decreasing particle size is correlated both with increasing loss-on-ignition (LOI) and with increasing radionuclide content (Fig. 4B). LOI is a measure of a sample's combustible mass that is inferred to be organic in nature. This relationship is similarly demonstrated for bulk sediment samples in Fig. 5, where LOI also is positively correlated with radionuclide activity and inversely correlated with median grain size (d_{50}). From these covariate relationships we confirm that the dominant transport phase of ^{131}I , as for ^7Be , is very-fine, organic particulate matter with which the radionuclides rapidly associate following deposition from the atmosphere.

The relative activities of ^{131}I and ^7Be in sediment are dependent on the differential decay rates of ^{131}I and ^7Be during particle transport, deposition, and storage, as well as on inheritance of ^7Be from previous or extended atmospheric exposure predating the FDNPP disaster. The strong correlation of ^{131}I and ^7Be in channel margin deposits along the entire length of Mink Brook suggests uniform inheritance and residence time of ^{131}I in this depositional environment. In this circumstance, the differential decay rates of ^{131}I and ^7Be can be exploited to constrain the age of the iodine-bearing sediment (32). Here we assume an initial ^{131}I - ^7Be composition equivalent to that of topsoil from our headwater floodplain site, where the soil was found to be saturated, overland flow appeared active, and lateral connectivity of the surrounding hill slope with the stream channel appeared in this manner to be qualitatively high. The corresponding maximum (no inheritance) transport-plus-storage age for channel margin sediments are thus calculated as $5.4 \pm 2.9 \text{ d}$ (mean ± 1 standard deviation).

In contrast, within the active stream channel, in both riffle and pool environments, sediments exhibit pronounced and variable departures of radionuclide activities from the ^{131}I - ^7Be correlation that distinguishes channel margin sediments (Fig. 4C). The channel margin environment is significantly enriched in ^{131}I ver-

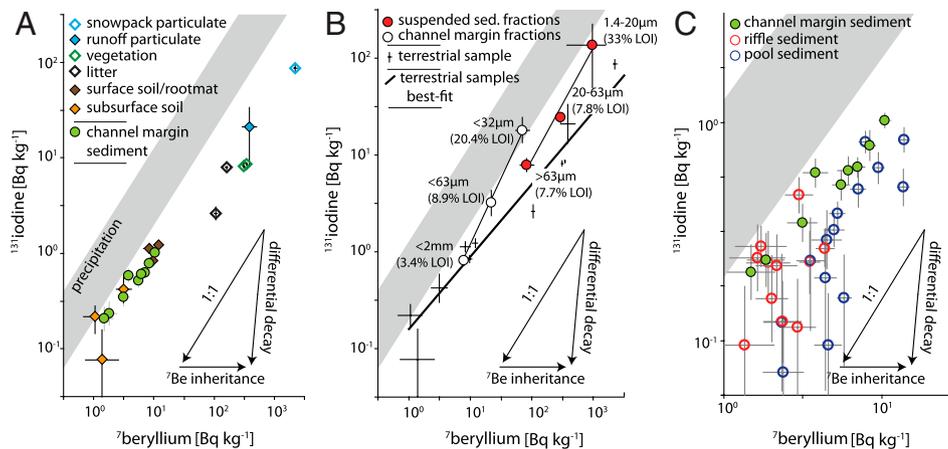


Fig. 4. ^{131}I and ^7Be activity-concentrations for the materials sampled in this study. Shaded region shows precipitation composition during the period which preceded sample collection. Error bars depict 1σ analytical uncertainty. (A) terrestrial materials (see legend), and waterline stream sediments. Arrows depict the effect of processes including: linear (1:1) mixing, differential decay of ^{131}I and ^7Be , and dilution by ^7Be inherited from previous or extended atmospheric exposure. (B) size fractions isolated from both stream channel sediment (white) and suspended sediment (red), with organic content and particle size included as data labels. Terrestrial sample best-fit is included for context. (C) inset of waterline sediment ^{131}I and ^7Be activities, with comparison to fast-water riffle and slow-water pool sediments. Channel margin sediments are shown in closed green, fast-water riffle sediments in open red, slow-water pool samples in open blue.

sediment ^7Be relative to both riffle and pool ($p = 0.031$ and $p < 10^{-4}$, respectively, by post hoc Tukey HSD test). This departure among riffles and pools indicates the role of site specific histories and different stream microenvironments in regulating the radionuclide burden of corresponding sediment deposits. Pool environments, for example, may be inferred to have lower exposure to fine particulate exchange, longer mean radionuclide residence times, and larger volumes of pre-Fukushima sediment. While the stream bed as a whole is shown here to sequester radioiodine fallout, channel margin deposits appear to play a significant and underappreciated role in regulating the short-term export of suspended organic matter, contaminants, and nutrients.

Toward an Integrated Perspective of Iodine Terrestrial Cycling. The Fukushima Daiichi nuclear disaster, though tragic, provides insight to the fate of radioiodine fallout in a terrestrial ecosystem. Our observations reveal the complexities of iodine behavior at a scale and in a context of utmost poignancy. We confirm that the

soil system of a temperate, low-order catchment actively and quantitatively scavenges ^{131}I upon its deposition from the atmosphere. The very rapid and stable association of radioiodine with organic carbon, together with the transport processes we describe here using the ^7Be tracer (Fig. 1), determines the subsequent fate of atmospherically derived iodine in terrestrial and low-order fluvial ecosystems.

Fluvial redistribution of iodine-bearing particulates produces “hotspots” within active channels and depositional zones where fine, organic matter accumulates. We demonstrate that these zones, even in low-order streams, may sequester significant quantities of fine particulate material, contaminants, and nutrients (33). The continuous processes of fluvial redistribution in small catchments are, in turn, important in supplying organic material (34, 35, 36) and soil carbon (37, 38) to larger river systems, lakes, wetlands, and estuaries (39, 40). By extrapolation the same may be expected of iodine (41, 42, 4). To the extent that iodine remains particle-bound during long-term cycling in the soil system, its continued removal from a landscape will follow mechanisms and time scales of soil erosion previously described using ^7Be , ^{210}Pb , and ^{137}Cs radionuclide tracers (43, 38, 44). However, the active focusing of iodine to surface soils, likely mediated by microbial activity, may enhance the susceptibility of iodine to surficial transport relative to other tracers.

The focusing and concentrating of radioiodine fallout on a landscape carries some implication for human ^{131}I exposure, though this risk is mitigated by the brevity of the ^{131}I half-life relative to the time scale of fluvial redistribution, and, in the present case, by the low radioiodine deposition observed at our site. More important are implications for ^{129}I (half-life 15.6 Ma), which has a fission yield approximately one third that of ^{131}I , and which, due to its long half-life and continued release from ongoing nuclear energy production, is perpetually accumulating in the environment (45, 1) and poses a growing radiological risk. Iodine-129 has also been proposed as a geochemical tracer for “young” carbon where explicit dating as by carbon-14 fails (5), and our findings support this use of ^{129}I measurements.

The ^{131}I behavior described here also serves as an analog for that of the vital micronutrient ^{127}I . Radioiodine emitted during reactor accidents is predominately gaseous (18, 26, 19), presumed to be some combination of I_2 and organic iodine, e.g., CH_3I (46, 47). These iodine species are also principle constituents of natural iodine atmospheric cycling (48), but both photo-dissociate rapidly and are recycled through numerous

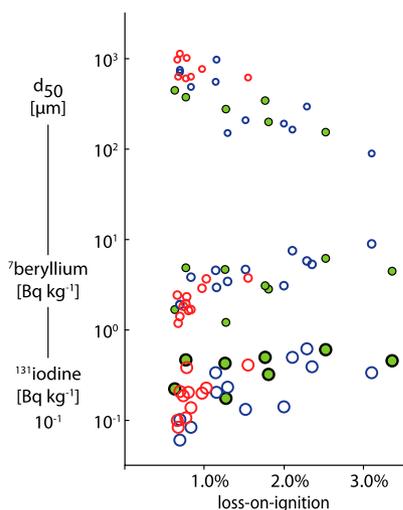


Fig. 5. Bulk sediment loss-on-ignition (LOI) as a predictor of median grain diameter (d_{50}), ^7Be activity and ^{131}I activity, respectively, for all bulk stream sediments. Ordinate axis is shared. Channel margin sediments are shown in solid green, riffle sediments in open red, and pool sediments in open blue.

intermediates in both gas and aerosol phases over time scales of hours to days (48, 49). The reactions of iodine with the atmosphere and its aerosol components favor an equilibrium gas-particle distribution (50, 51, 52), and for both natural and fission-derived iodine, progress at comparable rates (53) and yield similar gross speciation (52). From these prior observations we conclude that the deposition of radioiodine via precipitation, thousands of kilometers and weeks removed from the Fukushima Daiichi nuclear disaster, follows that of natural iodine, and therefore conforms to the primary natural pathway of iodine entry to the terrestrial environment (54). We must also conclude that the postdepositional redistribution of radioiodine, as observed and reported here, manifests the natural cycling of stable ^{127}I iodine in a temperate, terrestrial ecosystem.

Methods

Catchment Characteristics. The Mink Brook catchment area is 48 km² and typical base flow discharge is ca. 0.01 m³ s⁻¹. Bankfull-channel width of Mink Brook is ca. 1 m at its headwaters, increasing to ca. 10 m at its confluence with the Connecticut River. Mean annual precipitation in Hanover, New Hampshire is ca. 100 cm.

Gamma Spectroscopy. Each of the radionuclides ^{131}I , ^7Be and $^{134,137}\text{Cs}$ are gamma emitters. The high energy of their gamma photons (364.5 keV, 477.6 keV, and 661.7 keV, respectively) permits measurement of very low abundances (<10⁻¹⁸ grams per gram soil) by standard gamma spectroscopic methods employed at the Dartmouth Short-lived Radionuclide Laboratory (55). For this study large samples (110 cm³ for solids, 1 L for waters) were measured in bulk and field-wet to avoid losses of ^{131}I resulting from handling or interactions with equipment materials, or by decay during prolonged processing. Sediment size fractions, isolated either by sieving or filtration, were

gamma-counted on filter paper. Analyses were performed within hours of collection for terrestrial samples or within hours to days for fluvial samples. All samples were stored at 4 °C in sealed plastic bags during any delay between collection and spectroscopic analysis. To minimize analytical uncertainty, sample gamma emissions were counted for at least 24 h. All measurements were decay-corrected to the time of collection.

Precipitation collections were acidified upon retrieval by addition of HCl to pH ~ 1 to ensure desorption of ^7Be from the collection vessel walls, and were presented immediately for gamma measurement. Given the short half-life of ^{131}I relative to the duration of our precipitation-collection campaign (Fig. 2), we corrected measured depositional inventories for decay that occurred during deployment of the collector. This correction was achieved by attributing each sample's cumulative nuclide deposition to daily wet precipitation totals:

$$J_{\text{deposition}} = \frac{J_{\text{collection}}}{\sum_i^n f_i e^{-\lambda t_i}}, \quad [1]$$

where J is activity inventory [Bq m⁻²], f_i is the known fraction of precipitation during a collection period that fell on day i , λ is the ^{131}I decay constant [d^{-1}] and t is time elapsed [days] between the precipitation event and closure of the collection period.

Sediment Characterization. Mass loss-on-ignition (LOI), a measure of sample organic matter content, was evaluated for oven-dried samples by combusting them at 550 °C for 4 h. Sediment particle-size analyses were performed by the laser scattering method using a Coulter LS230.

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