

Airborne gamma-ray emitters from Fukushima detected in New York State

Michael E. Kitto · Traci A. Menia · Douglas K. Haines · Shaun E. Beach · Clayton J. Bradt · Eileen M. Fielman · Umme-Farzana Syed · Thomas M. Semkow · Abdul Bari · A. J. Khan

Received: 16 July 2012 / Published online: 17 August 2012
© Akadémiai Kiadó, Budapest, Hungary 2012

Abstract An air-sampling network that operates continuously as part of New York State's environmental surveillance program collected radionuclides emitted as a result of the Fukushima nuclear accident. Samples were collected, typically for 7 days each, by drawing $\sim 600 \text{ m}^3$ of air through a particulate-collecting filter followed in series by a canister containing activated charcoal. Additional air sampling was implemented at ~ 3 -day intervals at two locations. Gamma-ray spectroscopy was used to confirm the detection of ^{131}I , ^{137}Cs , ^{134}Cs , and ^7Be in the particulate phase at all sites, with maximum concentrations near 1,260, 160, 160, and 5,200 $\mu\text{Bq}/\text{m}^3$, respectively. Gas-phase ^{131}I , collected on activated charcoal, exhibited a maximum concentration of 3,400 $\mu\text{Bq}/\text{m}^3$ at the sites. Assessment of radionuclide levels in the air samples suggests that there were minimal health impacts from the airborne radionuclides as the activities contributed an insignificant amount to the annual human dose.

Keywords Surveillance · Dose · Gamma-ray spectroscopy · Cesium · Iodine

Introduction

While the majority of atmospheric radioactivity can be accounted for by cosmogenic production and decay products of the ^{238}U and ^{232}Th decay series, some originates from nuclear power reactors, nuclear accidents, and past nuclear-weapon testing. Airborne radioactivity from these natural and man-made sources can enter the human body through inhalation and through settling on foodstuffs. For example, an isotope of beryllium, ^7Be , produced by cosmic-ray bombardment of nitrogen and oxygen atoms in the troposphere, is commonly observed in ground-level air and on vegetation due to its lengthy half-life (53 days). Likewise, deposited radiostrontium (^{90}Sr) and ^{137}Cs from past atomic detonations has been observed in milk throughout New York State (NYS) for several decades [1]. In addition, studies have documented the release and deposition of alpha- and beta-emitting isotopes from the ^{238}U and ^{232}Th decay series during coal combustion.

To detect airborne radioactivity, the NYS Department of Health initiated a sampling and analysis program for scheduled surveillance around reactor and nonreactor sites over 25 years ago. This continuous surveillance provides a method for the determination of normal radio-activity levels and for monitoring of reactor emissions, and provides a basis for post-incident dose reconstruction should an emission event or accident occur. Airborne activities of gross-beta particles, tritium, and gamma-ray emitters, determined as part of the surveillance program, are often near or below the analytical detection limits. A summary of results of the surveillance program are presented elsewhere [2]. In this paper, activities of gamma-ray emitters that were observed in NYS following the atmospheric release of radionuclides from the Fukushima Daiichi complex of nuclear power reactors in Japan are provided.

M. E. Kitto (✉) · T. A. Menia · D. K. Haines · S. E. Beach · C. J. Bradt · E. M. Fielman · U.-F. Syed · T. M. Semkow · A. Bari · A. J. Khan
Wadsworth Center, New York State Department of Health,
P.O. Box 509, Albany, NY 12201, USA
e-mail: kitto@wadsworth.org

M. E. Kitto · T. M. Semkow
School of Public Health, State University of New York,
Albany, NY 12203, USA

As a result of an earthquake and subsequent tsunami affecting the east coast of Japan on March 11, 2011, fission-product radionuclides were released from the nuclear power reactors to the atmosphere. Although the radioactive plume traveled across the Pacific Ocean, primarily in the direction of the Arctic, and underwent dispersion and deposition, measurable amounts of gamma-ray emitters were detected at several locations in NYS. Our data are provided for comparison to concentrations of atmospheric radioactivity typically observed at the sampling locations, as well as provide an estimate of the radiation dose to the citizens of NYS as a result of the release from the Japanese reactors.

Experimental

Locations of the six air-monitoring sites in six counties in NYS are shown in Fig. 1. The sites at Albany (A) are not affected by the operation of reactor facilities that are known to release radionuclides to the atmosphere. Samples analyzed from these locations exhibit normal concentrations of naturally occurring radionuclides, plus the usual influence from past nuclear detonations and global use of nuclear energy. The six nuclear power reactors operating in NYS are located near three sites (B, E, F). Additional air-monitoring sites are located near Brookhaven National Laboratory (G) and a small training reactor operated by the United States (US) Department of Energy (D).

At the air-monitoring surveillance sites, airborne particulates were collected for ~ 7 days each, by drawing air through a cellulose-fiber filter (5-cm dia; Grade 54; Ahlstrom Corp., Mt. Holly Springs, PA) at a typical flow rate of ~ 60 L/min (Table 1). The air samplers are enclosed in

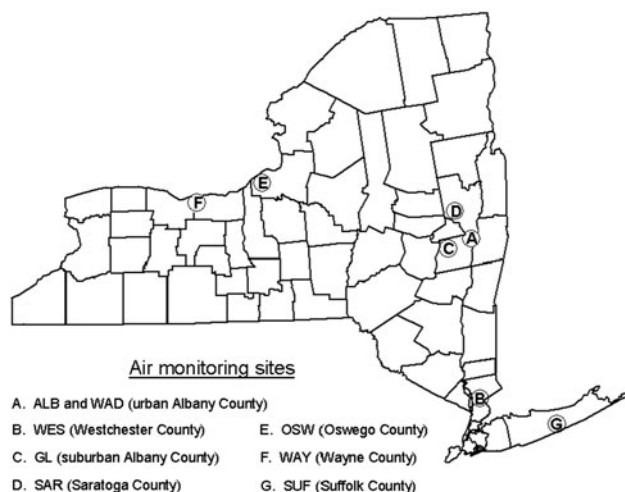


Fig. 1 Map of New York State (NYS), showing the locations of the sampling sites

galvanized steel housings ~ 1.5 m above the ground. Staff from county health departments changed the filters approximately weekly and delivered the samples to the laboratory within days of collection. Gas-phase ^{131}I was collected by placement of a 5.8-cm dia canister (model BG-300; Radeco, Plainfield, CT) containing activated charcoal in series after the particulate-collecting filter. Charcoal canisters were packaged separately, but delivered with the air-particulate filters.

For the period of March to May 2011, additional air sampling was conducted at site C (~ 15 km from site A) using identical equipment as that deployed at the surveillance air-monitoring sites. Samples were changed twice weekly, thus the air volume was only half that collected at the surveillance sites. In contrast, additional sampling at a location ~ 1 km from site A (denoted as A*) utilized a high volume (~ 1.2 m³/min) pump, a 10- μm particle cutoff head, and large (20×25 cm²) polycarbonate membrane (1 μm ; Poretics Corp., Livermore CA) or quartz (Whatman QMA, Clifton, NJ) prefilters. A large charcoal-impregnated filter (20×25 cm²; Schleicher & Schuell Inc., Keene, NH) was placed in series after the prefilter to collect gaseous airborne components. In addition to airborne samples, fallout samples were collected in open buckets exposed to atmospheric precipitation and dust at the two ALB sites (A and C). Buckets containing no liquid precipitation were washed with distilled water to obtain the dry fallout sample.

Gamma-ray spectrometry measurements of the filters, charcoals, and fallout were conducted using high-purity germanium (HPGe) detectors in low-background lead shields. Detector efficiencies ranged from 20 to 133 %, relative to a 3" \times 3" NaI(Tl) detector, and resolutions were ~ 2.0 keV at 1,333 keV. During standardization of the HPGe detectors, efficiencies for ^{134}Cs were corrected for coincidence summing. The filter and charcoal samples were counted directly on the detector surface for 400–1,000 min, while fallout samples were counted for 300–1,000 min. Data were acquired with a multiplexed system coupled to an Ethernet network. Spectra were collected and analyzed using the Genie 2000 gamma spectroscopy system (Canberra Industries Inc., Meriden, CT). Results, errors, and detection limits for gamma-ray spectrometry data are reported at the 95 % confidence level (~ 2 sigma). Concentrations of ^{131}I and ^7Be were corrected for decay from the midpoint of the collection period to the midpoint of the counting period. For all analyses, quality control was ensured by the use of NIST-traceable standards and participation in external proficiency-testing studies.

Results and discussion

Although a wide variety of radionuclides were emitted from the damaged reactors at Fukushima, in this study we

Table 1 Characteristics of air sampling conducted at various locations in New York State

Site	Dates (2011)	Duration (h)	Volume (m ³)	Flow rate (L/min)	Filter area (cm ²)	Filter type
<i>Surveillance</i>						
A ALB	Mar 16–Apr 22	144–192	480–740	60	20	CF/C
E OSW	Mar 8–Apr 26	168	440–560	53	20	CF/C
D SAR	Mar 9–May 3	142–192	520–750	63	20	CF/C
G SUF	Mar 14–Apr 18	168–173	580–590	58	20	CF
F WAY	Mar 7–Apr 25	148–187	490–620	55	20	CF/C
B WES	Mar 4–Apr 18	168–241	630–880	63	20	CF/C
<i>Special</i>						
C GL	Mar 17–May 2	72–96	230–320	55	20	CF/C
A* WAD	Mar 18–Apr 20	48–119	3,180–8,020	1,240	540	PC/C
A* WAD	Apr 20–May 16	51–144	3,580–11,000	1,260	540	GF/C

GF glass fiber, CF cellulose fiber, PC polycarbonate, C charcoal backup

report on the detection of ¹³¹I (*t*_{1/2} = 8.03 days), ¹³⁴Cs (*t*_{1/2} = 2.07 years) and ¹³⁷Cs (*t*_{1/2} = 30.1 years), as well as naturally occurring ⁷Be (*t*_{1/2} = 53.2 days), in the atmospheric samples. Detection of additional reactor-produced radionuclides in the collected air samples has been reported elsewhere [3]. Figure 2a illustrates that only the annihilation peak (511 keV) existed in the spectra of a typical detector background, while photopeaks for the isotopes of interest were prevalent on a typical 5-cm prefilter (Fig. 2b) collected during the sampling period. ¹³¹I, ⁷Be, and ¹³⁷Cs each emit a single primary gamma-ray photopeak (364, 477, and 662 keV, respectively), while ¹³⁴Cs has major photopeaks at 605 and 796 keV.

Fukushima-related radionuclides were first detected in NYS on filters collected more than a week after the reactor accident, and activities continued to rise for several weeks before diminishing. As shown in Fig. 3, similar concentration patterns were observed for ¹³⁴Cs at the six surveillance

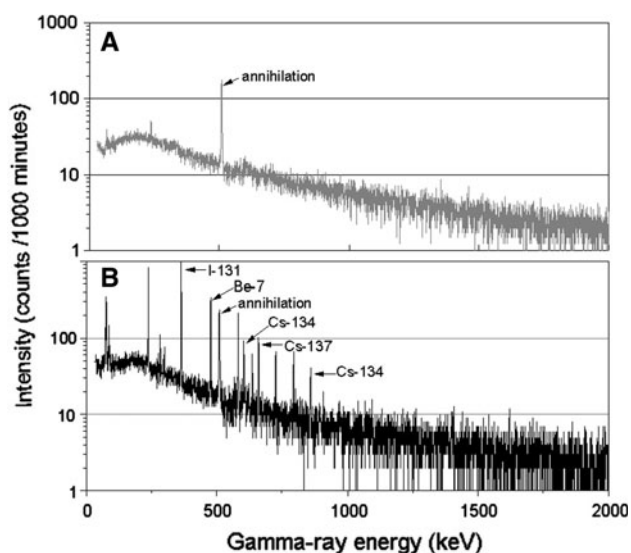


Fig. 2 Comparison of gamma-ray spectra for typical detector background (a) and air-particulate filter (b)

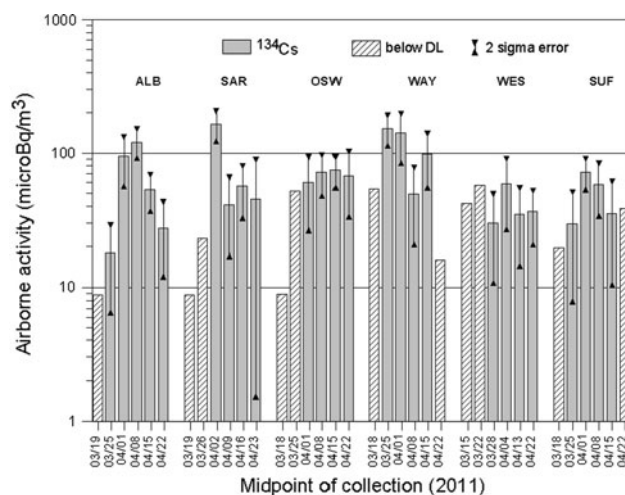


Fig. 3 Activities of airborne ¹³⁴Cs determined at six surveillance sites in NYS

sites that employed identical sampling apparatus. Slight differences are likely due to local rainout by precipitation. While it is evident that ¹³⁴Cs was not detected on filters collected immediately after the accident and prior to arrival of the plume, the high detection limits reported for some filters (e.g., OSW 4/22) are a result of reduced counting times (e.g., 400 min) and/or use of a lower-efficiency detector. Considering the low levels, activities calculated based on the two gamma-rays of ¹³⁴Cs were somewhat correlated (*r*² = 0.67). For the sampling periods given in Table 1 in which activities were above detection limits, the site-averaged ¹³⁴Cs varied from 47 to 94 μBq/m³ at the six sites and was 69 μBq/m³ overall. In comparison of results above detection limits, the overall average for NYS is somewhat less than the average (95 μBq/m³) reported for Washington State [4], equivalent to the average (72 μBq/m³) observed in Europe [5], and greater than the average activity (39 μBq/m³) measured in Greece [6]. These differences in activity are likely the result of variations in weather (rainout and

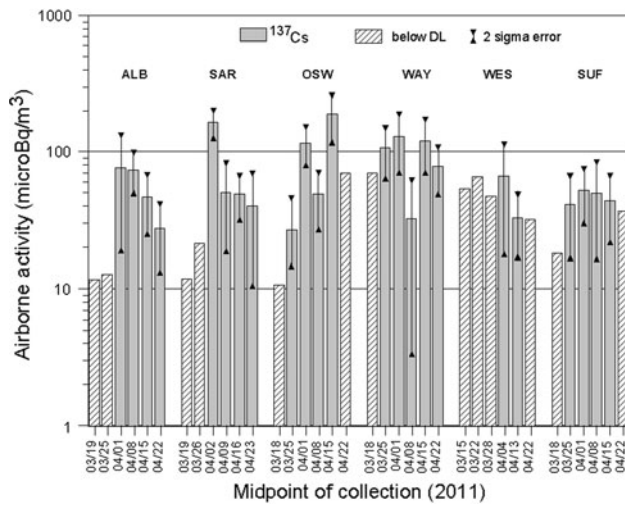


Fig. 4 Activities of airborne ^{137}Cs determined at six surveillance sites in NYS

dispersion) and wind patterns impacting the sites, and proximity of the sites to Japan.

The activities and distribution for ^{137}Cs on the filters (Fig. 4) were similar to that observed for ^{134}Cs . For the sampling periods given in Table 1 in which activities were above detection limits, the site-averaged ^{137}Cs varied from 40 to 111 $\mu\text{Bq}/\text{m}^3$ at the six sites and was 68 $\mu\text{Bq}/\text{m}^3$ overall. The latter is 24 times less than the average observed on the east coast of the US following the Chernobyl accident [7]. As with ^{134}Cs , the overall average of results above detection limits determined in this study is about half of the average (130 $\mu\text{Bq}/\text{m}^3$) reported for Washington State [4], equivalent to the average (76 $\mu\text{Bq}/\text{m}^3$) reported for northern Europe [5], and greater than the average activity (44 $\mu\text{Bq}/\text{m}^3$) measured in Greece [6]. As with ^{134}Cs , differences in reported values are likely the result of variations in weather and wind patterns, and proximity of the sites to Japan. The similarity of ^{134}Cs and ^{137}Cs activities in these studies implies that the ratios of $^{137}\text{Cs}/^{134}\text{Cs}$ are near unity. For filters from the six sites, in which both primary gamma-rays of ^{134}Cs were detected, the ratio of $^{137}\text{Cs}/^{134}\text{Cs}$ averaged 1.13 ± 0.25 . The two isotopes were well correlated ($r^2 = 0.82$). In this study no decay correction was applied for ^{134}Cs (2 % over three weeks) since this adjustment was well within the measurement errors.

Of the radioisotopes reported here, only ^{131}I exists in both particulate ($^{131}\text{I}_p$) and gaseous ($^{131}\text{I}_g$) phases in the atmosphere. Historically, no $^{131}\text{I}_p$ has been observed above the detection limit from samplers surrounding the NYS reactor sites. For the current study, $^{131}\text{I}_p$ was measurable at all routine sampling sites (Fig. 5), with an overall average of 460 $\mu\text{Bq}/\text{m}^3$ for the sampling periods listed in Table 1 in which results exceeded detection limits. Site averaged $^{131}\text{I}_p$

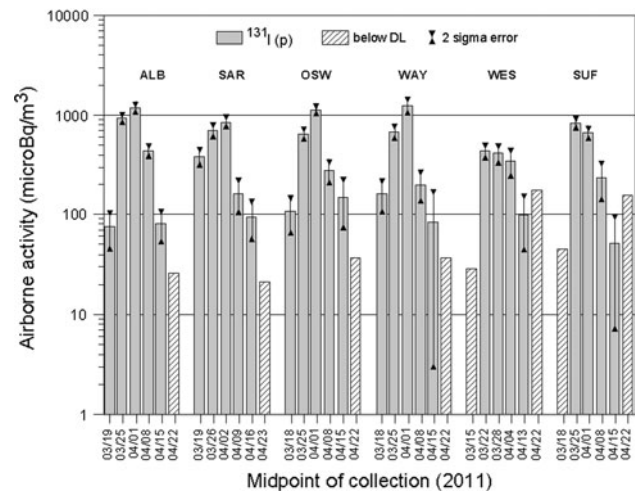


Fig. 5 Activities of airborne particulate ^{131}I determined at six surveillance sites in NYS

activities ranged from 320 to 550 $\mu\text{Bq}/\text{m}^3$, with maximum measured activities provided in Table 2. At each sampling site, the $^{131}\text{I}_p$ activity peaked near the beginning of April. As reported elsewhere [5], and apparent by comparing Figs. 4 and 6, both particulate and gaseous ^{131}I were detected before ^{137}Cs at every surveillance air-monitoring site (except SUF). This observation is likely due to the greater airborne concentrations and lower detection limit for ^{131}I relative to the radiocesium isotopes. The overall average of the $^{131}\text{I}_p$ activities were about half of the values (1,000 $\mu\text{Bq}/\text{m}^3$) reported for Washington State [4], comparable to results measured in most European countries [5], and about double the average activity (240 $\mu\text{Bq}/\text{m}^3$) that was measured in Greece [6] during the same period. Both peak and average $^{131}\text{I}_p$ levels determined in this study were about one-third of values determined in the eastern US following the Chernobyl accident [7].

The gaseous component ($^{131}\text{I}_g$) was collected on activated charcoal canisters at the six surveillance sites. Except

Table 2 Maximum airborne activities ($\mu\text{Bq}/\text{m}^3$) from sampling sites in New York State

Site		^{131}I (particle)	^{134}Cs	^{137}Cs	^7Be	^{131}I (gas)
A	ALB	1,190	77	120	4,520	3,030
E	OSW	1,140	120	75	4,270	2,690
D	SAR	860	160	160	5,180	2,990
G	SUF	840	53	73	3,540	N/A
F	WAY	1,260	130	150	4,750	3,420
B	WES	440	67	60	3,120	1,880
C	GL ^a	1,460	200	240	4,740	3,360
A*	WAD ^a	360	29	29	1,270	510

N/A results not available

^a Sampling protocol different at this site (see text)

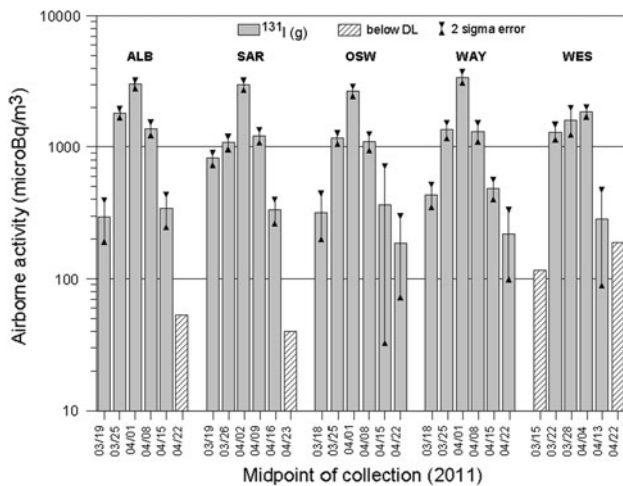


Fig. 6 Activities of airborne gaseous ¹³¹I determined at five surveillance sites in NYS

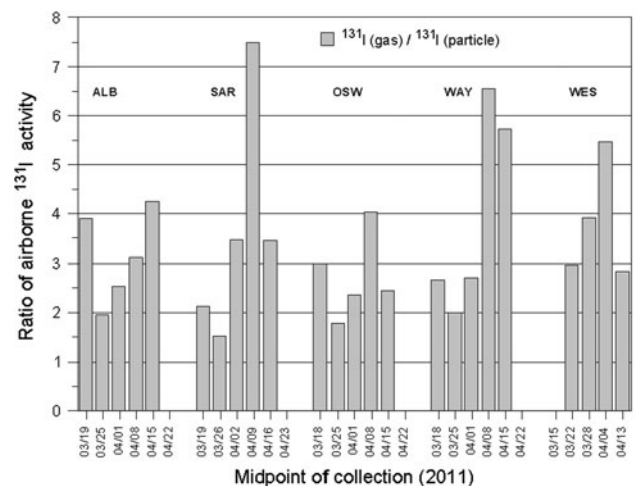


Fig. 7 Activity ratios of gaseous ¹³¹I to particulate ¹³¹I determined at five surveillance sites in NYS

for the ⁴⁰K inherent to the charcoal and the short-lived progeny of collected radon, no other radionuclides were observed on the charcoal. As with ¹³¹I_p, no gaseous ¹³¹I has been detected from near the NYS reactor sites during several decades of sampling. However, a decade ago, ¹³¹I_g activity was detected on the charcoal canisters at the Albany (A) background site [2], presumably due to emissions from incineration of dried sewage sludge containing excrement from radioiodine therapy treatments. During an 8-month period when the sampler was adjacent to the incinerator, the airborne ¹³¹I_g concentrations averaged 1.1 ± 0.9 mBq/m³ (¹³¹I_p was not measured). At the current air-sampler location (~1.5 km from the incinerator), airborne ¹³¹I_g concentrations are consistently below the detection limit (roughly 0.04 mBq/m³). For the current study, ¹³¹I_g was collected at five of the six sampling sites (Fig. 6), with a range of 0.2–3.4 mBq/m³, and an overall average of 1.2 mBq/m³. Few Fukushima-related studies have reported ¹³¹I_g levels, thus comparisons are sparse. Though no individual results were provided, Masson et al. [5] reported peak ¹³¹I_g levels in European countries that exceeded those measured in NYS, possibly due to transport mechanisms and/or particle-to-gas conversion during transport and collection. The average ¹³¹I_g level determined in the current study was about half of that measured in the eastern US following the Chernobyl accident [7]. A comparison of Figs. 5 and 6 indicates that ¹³¹I_p and ¹³¹I_g levels peaked at the same time. Activities of ¹³¹I_g and ¹³¹I_p in the air samples were well correlated (*r*² = 0.80).

Over a three-year sampling period in the eastern US, gaseous iodine (stable) comprised nearly 90 % of the total atmospheric iodine [8], while sampling at the location following the Chernobyl accident found ¹³¹I_g comprised ~65 % of the total ¹³¹I [7]. For samples from the current study in which both phases were measured above the

detection limit, ¹³¹I_g comprised, on average, 75 % of the total ¹³¹I (range = 60–88 %). This average is identical to the 77 % (±14 %) measured in some European countries [5]. In contrast to ratios of ¹³⁷Cs/¹³⁴Cs near unity, ratios of ¹³¹I_g/¹³¹I_p (Fig. 7) at the six sampling sites averaged 3.4 and varied greatly (1.5–7.5). In comparison, ¹³¹I_g/¹³¹I_p ratios in Washington state [4] varied between 2 and 20, with an average of ~5. The range of radioiodine ratios is likely a result of variations in weather (deposition, rainout, and dispersion) incurred by the emissions while transported from Japan. Of the radioisotopes determined in this study, poor correlations existed between ¹³¹I_g and ¹³⁷Cs (*r*² = 0.18) and between ¹³¹I_p and ¹³⁷Cs (*r*² = 0.14). Similarly poor correlations of these isotopes in the emissions from Japan have been reported elsewhere [5, 6].

Naturally occurring ⁷Be was above the detection limit on all of the particulate filters, with values ranging from 1.4 to 5.2 mBq/m³ and an average of 3.2 ± 0.9 mBq/m³. As summarized elsewhere [2], ⁷Be activities are historically similar among the sites over a 13-year period, with an average activity that was nearly identical to that measured during the current study. A seasonal pattern of elevated ⁷Be levels occurs during the summer months due to the increased solar flux that occurs during this period, with concomitant increased production of ⁷Be. Due to the different sources, one would not expect ⁷Be levels to correlate with the released fission products, and correlations with the measured activities (¹³¹I_p *r*² = 0.36; ¹³⁷Cs *r*² = 0.23) supports this conclusion.

In addition to the air-filter and charcoal samples collected from the six surveillance sites, supplemental sampling was implemented at two sites (A* and C) from March to May. Since these samples did not mimic the sampling schedule of the routine monitoring sites, the results are discussed separately. Samples from site C were collected

for ~3 days each, resulting in reduced air volumes and fewer activities above detection limits. Ratios of $^{137}\text{Cs}/^{134}\text{Cs}$ at site C averaged 1.13, a value identical to that determined at the reactor surveillance sites. $^{131}\text{I}_p$ was detected on 70 % of the filters, with an average ($0.58 \pm 0.46 \text{ mBq/m}^3$; range = $0.04\text{--}1.46 \text{ mBq/m}^3$) which is identical to that (0.55 mBq/m^3) determined at site A. $^{131}\text{I}_g$ was detected on 85 % of the charcoal canisters, with an average ($1.3 \pm 1.1 \text{ mBq/m}^3$; range = $0.1\text{--}3.4 \text{ mBq/m}^3$) which is nearly identical to the average (1.4 mBq/m^3) determined at site A. These similarities are expected based on the close proximity (~15 km) of the two samplers. The ratios of $^{131}\text{I}_g/^{131}\text{I}_p$ averaged 2.6 (range = 1.2–4.9). On average, $^{131}\text{I}_g$ comprised 65 % of the total ^{131}I . ^7Be was detected on all of the particulate filters, with values from 1.9 to 4.7 mBq/m^3 and an average ($3.4 \pm 0.8 \text{ mBq/m}^3$) that was comparable to activities determined at the reactor surveillance sites.

About 1 km from site A, high-volume sampling was conducted at ~3-day intervals with an average volume of ~5,500 m^3 . While the large volume allowed the detection of several radioisotopes [3], only a few are discussed here. Activity concentrations of ^{137}Cs , which averaged $13 \pm 9 \text{ } \mu\text{Bq/m}^3$ and ranged from 1 to $29 \text{ } \mu\text{Bq/m}^3$, were above detection limits on 90 % of the filters (Fig. 8). These concentrations were significantly less than those observed on the open-faced filters that were used at the six surveillance sites, possibly due to the 10- μm particle cutoff inlet used at site A* and/or inclusion of additional sampling periods containing low activities. Concentrations of ^{134}Cs averaged $14 \pm 10 \text{ } \mu\text{Bq/m}^3$ (range = $1\text{--}29 \text{ } \mu\text{Bq/m}^3$) and correlated well with ^{137}Cs ($r^2 = 0.97$). For filters on which both isotopes were detectable, the ratios of $^{137}\text{Cs}/^{134}\text{Cs}$ averaged 1.05. During the first month of sampling at the site, $^{131}\text{I}_p$ was measured on every filter (Fig. 9), with an

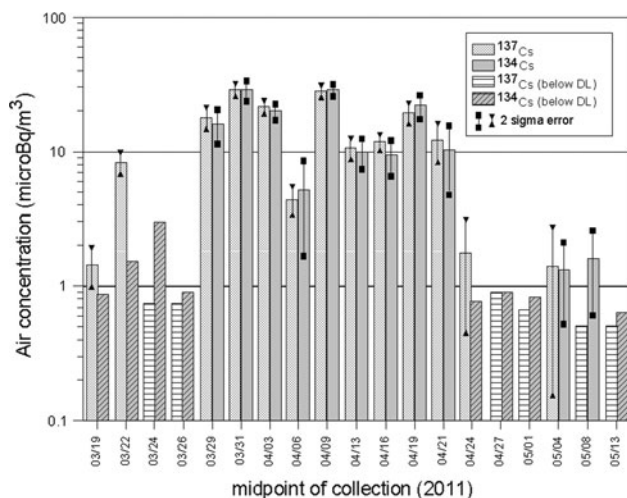


Fig. 8 Airborne activities of ^{134}Cs and ^{137}Cs determined from high-volume sampling at site A*

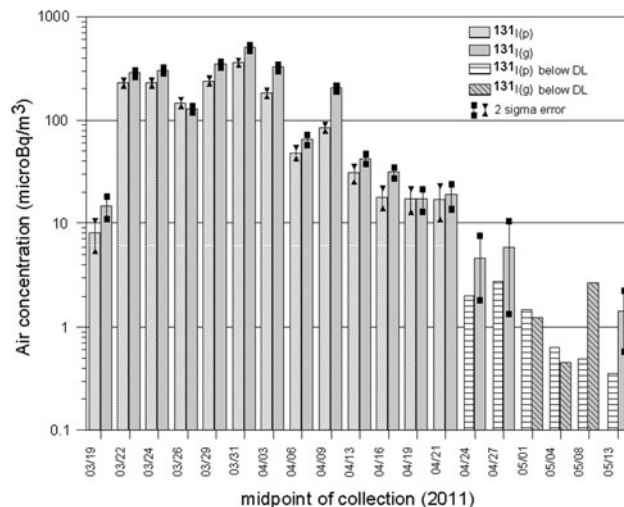


Fig. 9 Airborne activities of particulate and gaseous ^{131}I determined from high-volume sampling at site A*

average of $125 \text{ } \mu\text{Bq/m}^3$ (range = $8\text{--}360 \text{ } \mu\text{Bq/m}^3$), but $^{131}\text{I}_p$ was not detectable ($<1 \text{ } \mu\text{Bq/m}^3$) after April 22. This 30-day time frame of exposure to the Fukushima-related radioisotopes was also observed at the other sampling locations in NYS. Concentrations of $^{131}\text{I}_g$ were detected on 84 % of the charcoal filters, and for a time period that exceeded that for $^{131}\text{I}_p$. The collected $^{131}\text{I}_g$ activity averaged $155 \text{ } \mu\text{Bq/m}^3$ (range = $1\text{--}510 \text{ } \mu\text{Bq/m}^3$). The ratios of $^{131}\text{I}_g/^{131}\text{I}_p$ averaged 1.5 ± 0.4 , with $^{131}\text{I}_g$ comprising, on average, 58 % of the total ^{131}I . Lastly, naturally-occurring ^7Be was detected on all of the particulate filters, with activities from 250 to $1,270 \text{ } \mu\text{Bq/m}^3$ and an average of $750 \text{ } \mu\text{Bq/m}^3$. A comparison of average activities determined at site A* with those from nearby site A shows the latter's concentrations were 4.0–4.8 times greater for ^{134}Cs , ^{137}Cs , $^{131}\text{I}_p$, and ^7Be , and nearly 9 times greater for $^{131}\text{I}_g$. The greater discrepancy for $^{131}\text{I}_g$ may be an artifact of the high flow rates (low contact time) through the charcoal filter paper used at site A* relative to lower flow through the thick charcoal canisters used at site A.

Gamma-ray emitting radioisotopes originating from Japan were also measured in fallout (wet and dry deposition) collected at sites A and C. Four of the six weekly samples collected at site A from March 19 to April 22 had ^{131}I concentrations ranging from 220 to $3,800 \text{ mBq/m}^2\text{-day}$. Considering the volume of rain ($0.2\text{--}1.9 \text{ L}_w$) collected with the fallout samples, the liquid concentrations of ^{131}I ranged from 110 to $1,470 \text{ mBq/L}_w$. In comparison, ^{131}I activities in two deposition samples in Greece [6] were near 100 and 700 mBq/L_w . Deposition of ^{137}Cs ($n = 2$) averaged $160 \text{ mBq/m}^2\text{-day}$ (46 mBq/L_w), while ^{134}Cs ($n = 1$) deposition was $220 \text{ mBq/m}^2\text{-day}$ (400 mBq/L_w). Naturally occurring ^7Be was detected in all six deposition samples from site A, with an average of $4.5 \text{ Bq/m}^2\text{-day}$

(3.1 Bq/L_w). Collection of fallout ($n = 8$) at site C, at intervals of 3–4 day each, had ¹³¹I concentrations above detection limits on half of the samples, with activities ranging from 350 to 5,100 mBq/m²-day (70–1,800 mBq/L_w). Deposition of ¹³⁷Cs ($n = 2$) at site C averaged 180 mBq/m²-day (63 mBq/L_w), while ¹³⁴Cs ($n = 1$) deposition was 140 mBq/m²-day (49 mBq/L_w). ⁷Be was detected in seven deposition samples at an average of 12 Bq/m²-day (4 Bq/L_w).

Dose assessment

Human exposure to the measured radioisotopes occurred through three pathways: inhalation, ingestion, and external irradiation. Only the radiological dose associated with inhalation of the observed radionuclides is provided, as similar calculations for ingested and external (submersion) radiation showed these two exposure pathways to be much less in magnitude than the inhalation dose. For a given time period, the committed effective dose equivalent ($E_{(\tau)}$) from inhaled gamma-ray emitters can be estimated, using Eq. 1, by factoring the measured concentrations (C_i) of the airborne radioisotopes during the period, an inhalation (I) rate (1.5 m³/h) [9], and adult dose coefficients (D_i) of 20, 20, and 39 nSv/Bq for ¹³¹I, ¹³⁴Cs, and ¹³⁷Cs [10], respectively.

$$E_{(\tau)} = S(D_i * C_i * I). \quad (1)$$

For the sampling periods in which the isotopes were detected at the six air-monitoring surveillance sites in NYS, the resulting committed effective dose values averaged 0.33, 0.05, and 0.10 nSv/day to the total body from the inhaled ¹³¹I_p, ¹³⁴Cs, and ¹³⁷Cs, respectively. Similarly, the committed effective dose values from ¹³¹I_g averaged 0.87 nSv/day, with a range of 0.13–2.46 nSv/day at the surveillance sites. The total committed effective dose values for the 6-week sampling period ranged from 4.9 nSv at WES to 7.9 nSv at WAY. These two sites are located ~400 km apart. Comparison of the total derived committed effective dose from inhalation of the airborne radioisotopes to that (~7 μSv/day) received from natural sources (primarily radon and its decay products), demonstrates that the activities of the Fukushima-related radiation provided an insignificant additional dose.

The absorbed dose to the thyroid gland from ¹³¹I is dependent on age, with infants being the most affected. For this sensitive population (1 year olds), the inhalation D_i is 3.2 μSv/Bq [10] for I₂ uptake to the thyroid. When ¹³¹I_p was detected at the six air-monitoring surveillance sites in NYS, the committed dose equivalent to the thyroid from ¹³¹I_p averaged 49 nSv/day, with a range of 6–136 nSv/day. Using an identical approach, the committed dose equivalent from ¹³¹I_g averaged 130 nSv/day, with a range of 20–370 nSv/day at the sites. The total absorbed dose to the

thyroid gland, determined by combining the contribution from ¹³¹I_p and ¹³¹I_g at each site for the 6 week sampling period, ranged from 690 nSv at WES to 1,040 nSv at ALB. This dose is trivial when compared to the US average daily radiation dose of ~17 μSv (6,200 μSv/year) from all sources.

Conclusions

Results of airborne samples collected following the Fukushima reactor accident and analyzed as part of an environmental surveillance network in NYS have been presented. Weekly filters of collected airborne-particulate matter had detectable ¹³⁴Cs, ¹³⁷Cs, ¹³¹I, and ⁷Be activity at all sites. Airborne activities of gaseous ¹³¹I, collected on activated charcoal, surpassed those collected on particulate matter. Airborne activity levels in NYS were considerably less than those measured in the eastern US following the Chernobyl accident. During the six-week sampling period, the health impact of Fukushima-related radionuclides in NYS was negligible, as demonstrated by comparing the increase in committed effective dose (<0.2 nSv/day) to that (~7 μSv/day) received from natural sources during the period, and posed no concern to public health.

Acknowledgments The authors wish to thank staffs of the Bureau of Environmental Radiation Protection, regional and local health departments, and power utilities that support the sampling for the environmental surveillance network.

References

1. New York State Department of Health (1994) Environmental radiation in New York State: Annual Report, 23
2. Kitto ME, Fielman EM, Hartt GM, Gillen EA, Semkow TM, Parekh PP, Bari A (2006) Long-term monitoring of radioactivity in surface air and deposition in New York State. *Health Phys* 90:31–37
3. Khan AJ, Semkow TM, Haines DK, Beach SE, Bari A, Bradt CJ, Fielman EM, Menia TA, Kitto ME, Syed UF (2011) Progress in low-background gamma spectrometry at New York State Department of Health. Presented at Radiobioassay & Radiochemical Measurements Conference, Sandestin, FL
4. Diaz LJ, Jaffe DA, Kaspar J, Knecht A, Miller ML, Robertson RGH, Schubert AG (2011) Arrival time and magnitude of airborne fission products from the Fukushima, Japan, reactor incident as measured in Seattle, WA, USA. *J Environ Radioact* 102: 1032–1038
5. Masson O, Baeza A, Bieringer J et al (2011) Tracking of airborne radionuclides from the damaged Fukushima Dai-ichi nuclear reactors by European networks. *Environ Sci Technol* 45:7670–7677
6. Manolopoulou M, Vagena E, Stoulos S, Ioannidou A, Papa-stefanou C (2011) Radioiodine and radiocesium in Thessaloniki, Northern Greece due to the Fukushima nuclear accident. *J Environ Radioact* 102:796–797

7. Kitto ME, Faller SH, Anderson DL, McCarthy LE (1991) Airborne Chernobyl radioactivity in College Park, Maryland. *Radiochim Acta* 55:43–45
8. Kitto ME (1987) Receptor modeling of atmospheric and acidic gases. PhD Thesis, University of Maryland, College Park
9. Eckerman KF, Wolbarst AB, Richardson ACB (1988) Limiting values of radionuclide intake and air concentration and dose conversion factors for inhalation, submersion, and ingestion. EPA-520/1-88-020, Federal guidance report no. 11, Environmental Protection Agency, Washington DC
10. US Nuclear Regulatory Commission website. <http://www.nrc.gov/about-nrc/regulatory/research/radiological-toolbox.html>. Accessed 1 Feb 2012