



# **Fission Products in National Atmospheric Deposition Program—Wet Deposition Samples Prior to and Following the Fukushima Dai-Ichi Nuclear Power Plant Incident, March 8–April 5, 2011**

Open-File Report 2011–1277

U.S. Department of the Interior  
U.S. Geological Survey

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By Gregory A. Wetherbee, Timothy M. Debey, Mark A. Nilles, Christopher M.B. Lehmann, and David A. Gay

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**U.S. Department of the Interior  
U.S. Geological Survey**

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## Conversion Factors

### SI to Inch/Pound

Multiply	By	To obtain
Length		
millimeter (mm)	0.03937	inch (in.)
meter (m)	3.281	foot (ft)
kilometer (km)	0.6214	mile (mi)
meter (m)	1.094	yard (yd)
Area		
square meter (m <sup>2</sup> )	0.0002471	acre
Volume		
liter (L)	1.057	quart (qt)
Mass		
gram (g)	0.03527	ounce, avoirdupois (oz)
kilogram (kg)	2.205	pound avoirdupois (lb)
Energy		
1,000 electron volts (keV)	$1.602 \times 10^{-16}$	joules
1,000,000 electron volts (MeV)	$1.602 \times 10^{-13}$	joules
Activity		
microCurie (μCi)	$1 \times 10^6$	Curies
Becquerels per liter (Bq/L)	27.03	picoCuries per liter (pCi/L)
Deposition (flux to Earth's surface)		
Becquerels per square meter (Bq/m <sup>2</sup> )	27.03	picoCuries per square meter (pCi/m <sup>2</sup> )

Vertical coordinate information is referenced to the North American Vertical Datum of 1988 (NAVD 88).

Horizontal coordinate information is referenced to the North American Datum of 1983 (NAD 83).

Altitude, as used in this report, refers to distance above the vertical datum.

Activities of isotopes in water are given either in picoCuries per liter (pCi/L) or Becquerels per liter (Bq/L).

Deposition (flux) of isotopes to Earth's surface is given in Becquerels per square meter (Bq/m<sup>2</sup>).

## Abbreviations and Acronyms

<b>Abbreviation</b>	<b>Full name</b>
ACM	AeroChem Inc. Metrics Model 301 precipitation collector
BrCl	bromine monochloride
Bq/m <sup>2</sup>	Becquerels per square meter
BQS	Branch of Quality Systems
CAL	Central Analytical Laboratory
Cs-134	cesium-134 isotope
Cs-137	cesium-137 isotope
Eu-152	europium-152 isotope
GIS	Geographic Information System
HAL	Mercury Analytical Laboratory
HCl	hydrochloric acid
HPGe	high-purity germanium
I-131	iodine-131 isotope
keV	thousand electron volts
MDN	Mercury Deposition Network
NADP	National Atmospheric Deposition Program
NIST	National Institute of Standards and Technology
NTN	National Trends Network
pCi/L	picoCuries per liter
pCi/m <sup>2</sup>	picoCuries per square meter
QA	quality assurance
SAP	sampling and analysis plan
USGS	United States Geological Survey

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## Abstract

Radioactive isotopes I-131, Cs-134, or Cs-137, products of uranium fission, were measured at approximately 20 percent of 167 sampled National Atmospheric Deposition Program monitoring sites in North America (primarily in the contiguous United States and Alaska) after the Fukushima Dai-Ichi Nuclear Power Plant incident on March 12, 2011. Samples from the National Atmospheric Deposition Program were analyzed for the period of March 8–April 5, 2011. Calculated 1- or 2-week radionuclide deposition fluxes at 35 sites from Alaska to Vermont ranged from 0.47 to 5,100 Becquerels per square meter during the sampling period of March 15–April 5, 2011. No fission-product isotopes were measured in National Atmospheric Deposition Program samples obtained during March 8–15, 2011, prior to the arrival of contaminated air in North America.

## Introduction

On March 11, 2011, a magnitude 9.0 earthquake centered off the Pacific Coast of Japan triggered a tsunami and a 14-meter (m)-high tidal wave that inundated the Fukushima Dai-Ichi Nuclear Power Plant near Sendai, Japan (Wakeford, 2011). The facility was left without electric power, which resulted in failure of the cooling systems for its six nuclear reactors (Wakeford, 2011). The resultant loss of containment due to steam explosions and the breach of fuel elements in reactors and cooling ponds lead to the discharge of radioactive materials into the atmosphere, including fission-product isotopes iodine-131 (I-131, half-life 8.0 days), cesium-134, and -137 (Cs-134 half-life 2.1 years; Cs-137 half-life 30.2 years) (Lide, 1990). These isotopes were reported in many different countries in the Northern Hemisphere at variable but low levels (Wakeford, 2011).

In response to the incident, the United States Geological Survey (USGS), lead agency within the National Atmospheric Deposition Program<sup>4</sup> (NADP), collaborated with NADP personnel to quickly develop sampling and analysis plans and quality assurance protocols to monitor the radioactive fallout

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<sup>4</sup> The NADP is a focused and coordinated effort of many different organizations, including federal, state, tribal, and local governmental agencies, educational institutions, private companies, and non-governmental agencies, to monitor atmospheric deposition throughout North America (<http://nadp.sws.uiuc.edu/>).

over North America. NADP sample management and handling protocols were adapted to archive both filter samples and excess whole-water wet-deposition samples during the period of March 8–April 5, 2011. USGS measured 456 samples by gamma spectrometry for radioactive isotopes for both filterable solids from the precipitation (280 samples) and whole-water wet-deposition (176 samples).

### National Atmospheric Deposition Program Sites

Sites owned by the NADP in both the NADP’s National Trends Network (NTN) and Mercury Deposition Network (MDN) were used to monitor fission-product isotope fallout. The NTN is described by Nilles (2001), and the MDN is described by Prestbo and Gay (2009). Standard operating procedures for NTN are available at <http://nadp.sws.uiuc.edu/lib/manuals/opman.pdf>, and for MDN at <http://nadp.sws.uiuc.edu/lib/manuals/mdnopman.pdf>, both accessed October, 2011.

As of summer 2011, precipitation was being collected weekly from 251 sites in the NTN and 105 sites in the MDN within Canada and the United States, including Alaska, Puerto Rico, and the U.S. Virgin Islands (figs. 1 and 2). Both NTN and MDN sites are equipped with automatic precipitation collectors that open to the atmosphere at the onset of precipitation and close at cessation of precipitation to collect wet-only atmospheric deposition. While the precipitation collectors obtain representative samples for chemical analysis, they do not provide precipitation depth measurements of consistent accuracy. Therefore, each site is equipped with a rain gage to measure precipitation depth for calculation of the mass of selected constituents per unit area (flux) to the landscape. Radioactive isotope deposition was calculated by equation 1.

$$\text{Deposition, in pCi/m}^2 = A_{pCi/L} \times Ppt_{mm}, \quad (1)$$

where:  $A_{pCi/L}$  is activity of radioactive isotope, in pCi/L, and  $Ppt_{mm}$  is precipitation depth, in millimeters (mm) as measured by the rain gage applied over a 1-square-meter area.

Comparison of NADP measurements to data obtained from other monitoring entities requires conversion of deposition in pCi/m<sup>2</sup> to Becquerels per square meter (Bq/m<sup>2</sup>). Because  $3.7 \times 10^{10}$  Bq equals  $1 \times 10^{12}$  pCi, the units are converted as follows in equation 2.

$$\text{Deposition, in pCi/m}^2 \times 0.037 = \text{Deposition, in Bq/m}^2 \quad (2)$$



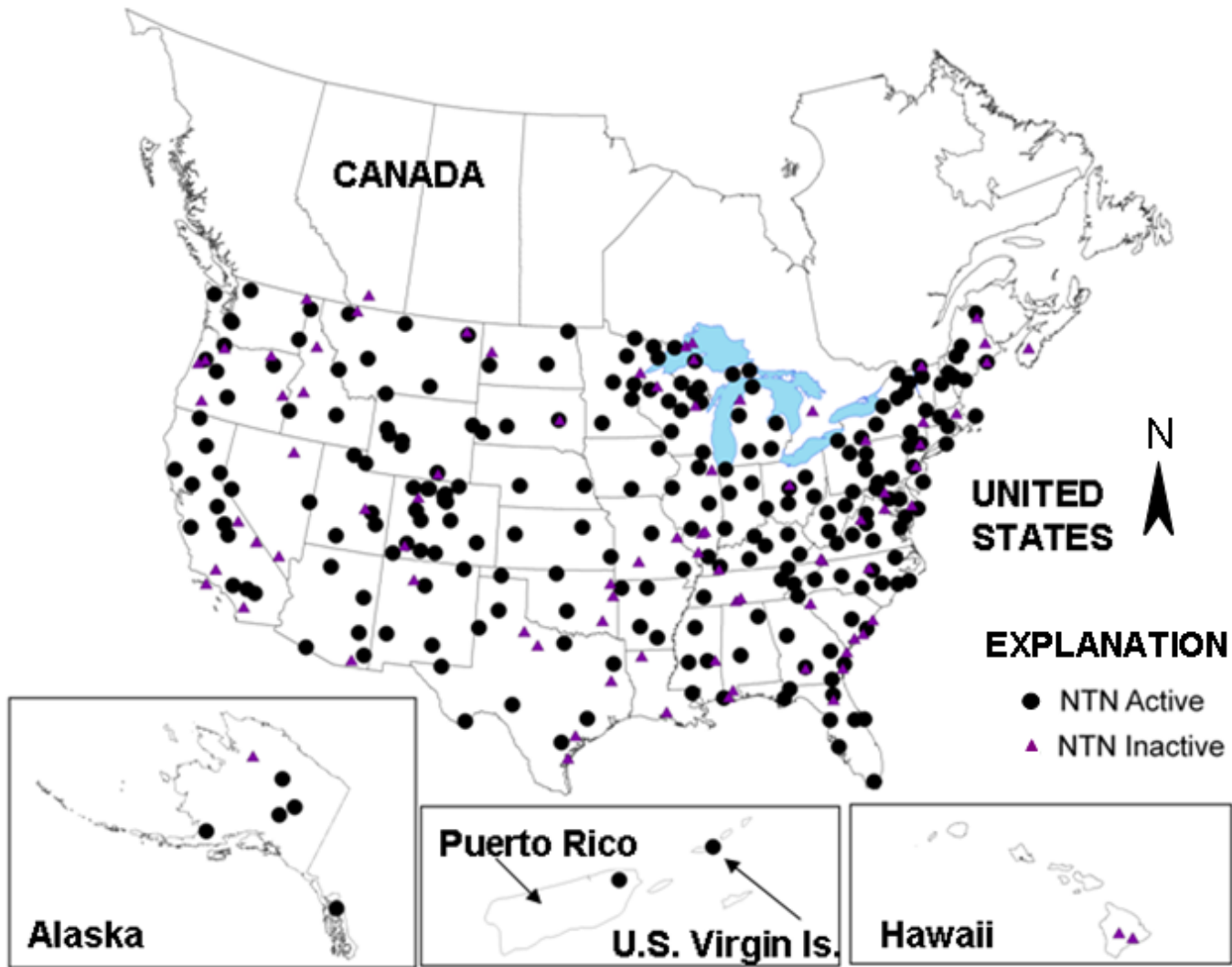


Figure 1. Active (2011) and formerly active National Atmospheric Deposition Program/National Trends Network (NTN) sites.

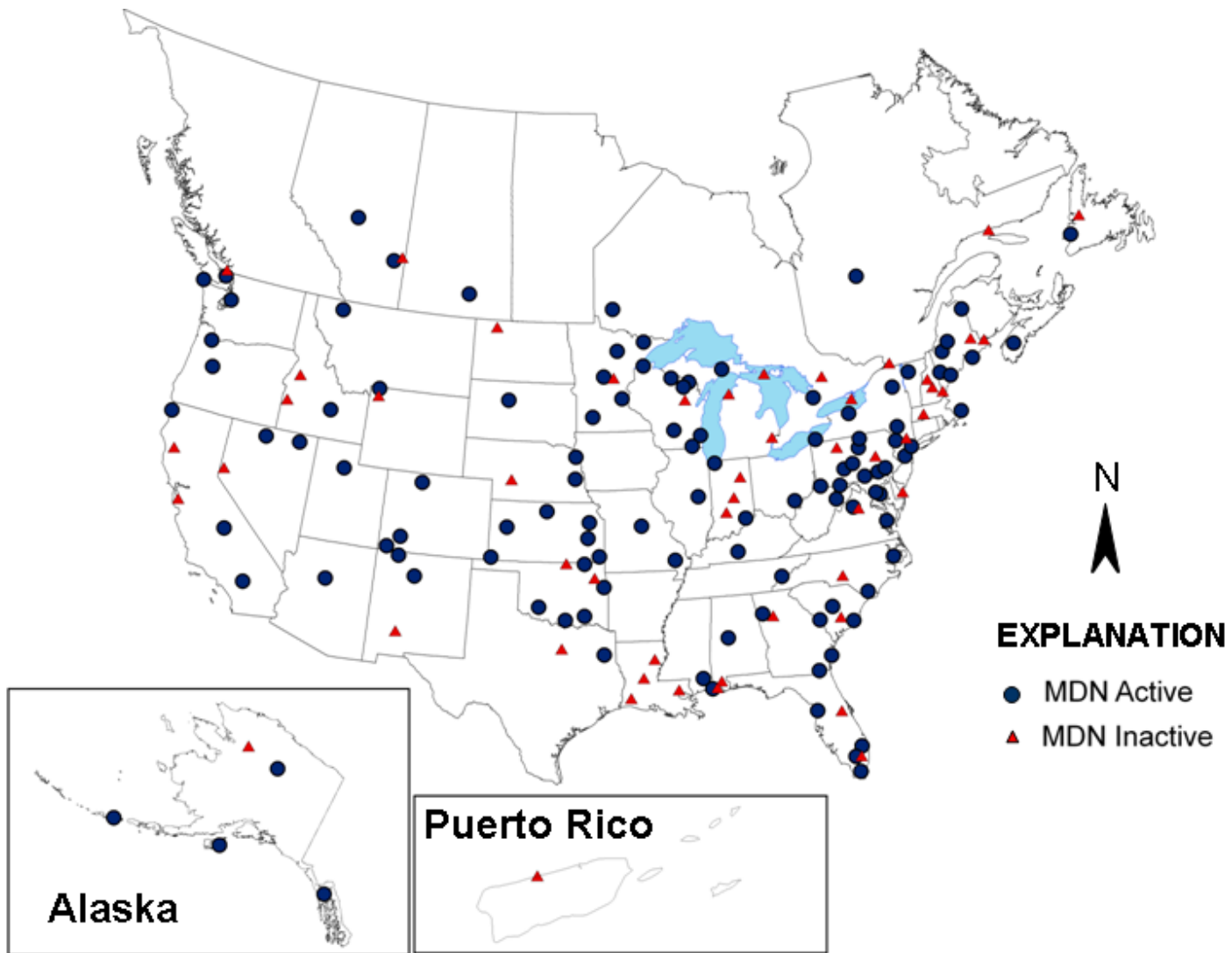


Figure 2. Active (2011) and formerly active National Atmospheric Deposition Program Mercury Deposition Network (MDN) sites.

Weekly composite NTN and MDN whole-water precipitation samples are removed from the automated collectors every Tuesday morning at approximately 9:00 a.m. local time. In this report, NADP site identifiers are used for brevity. The identifiers consist of the state or country abbreviation and a one- or two-digit number. For example, site CO90 is in Colorado (CO) and CAN5 is in Quebec, Canada.

## **Purpose and Scope**

The purpose of this report is to present the methods and results from sampling and analysis of the radioactive isotopes iodine-131 (I-131), cesium-134 (Cs-134), and cesium-137 (Cs-137) in NADP wet-deposition samples during the period of March 8–April 5, 2011. These isotopes are collectively referred to as “fission products” hereinafter. This report describes sample collection, processing, and analysis methods for unfiltered wet-deposition samples and filtered particulates (filter samples). The report also presents quality assurance (QA) protocols and results.

Gamma-ray spectra for naturally occurring gamma-emitting isotopes were obtained as part of this study, but these spectral components were not quantified for this report. Health risk assessments and comparison of measured activities to water-quality or health-related standards are beyond the scope of this report. This report provides no interpretation with respect to atmospheric transport processes.

Deposition of fission products is estimated using the measured activities and precipitation-depth measurements obtained from the rain gages at each NADP site. Selected sites in the southern and southeastern regions of the Nation were excluded from this study because samples from these sites were used for a plant pathogen study, for which the entire sample volume is filtered and needed for analysis in that study. An example of a similar NADP plant pathogen study is described by Barnes and others (2009).

## **Methods**

### **Adaptation of National Atmospheric Deposition Program Networks**

On March 14, 2011, within 48 hours (hr) after the release of radioactive material into the atmosphere at the Fukushima Dai-Ichi Nuclear Power Plant in Japan on March 12, 2011, the NADP coordinated with the USGS to put a plan in place to measure radionuclide fallout in NADP wet-deposition samples. NADP/NTN began saving excess unfiltered wet-deposition samples with special handling and analysis protocols, beginning with the sample-collection week of March 8–15, 2011, prior to any documented measurement of radioactive fallout in North America (Leon and others, 2011). Sampling and analysis plans (SAPs) were developed and reviewed by USGS and NADP personnel. These plans defined methodology for sample archive, preservation, analysis, and quality assurance for both the filter samples and wet-deposition samples. The SAPs delineated North America into three regions: western, central, and eastern. Samples were prioritized for analysis by region with the western region given first priority, and the eastern region given last priority. The goal of the prioritization scheme was to analyze samples with the greatest likelihood of containing fission products first, with a particular focus on short-lived radionuclides such as I-131. It was assumed that radionuclide concentrations in wet-deposition samples would be proportional to the west-to-east (prevailing wind direction; Takemura and others, 2011) distance from Japan.

The NTN samples are collected in clean, 29.4-centimeter (cm) diameter, 13.2-liter (L), polyethylene buckets. The MDN samples are collected using a borosilicate glass sampling train consisting of a 120.1- mm diameter funnel and thistle tube that directs the precipitation to a 2-L borosilicate bottle pre-charged with 20 milliliters (mL) of 1-percent (volume : volume) hydrochloric

acid (HCl) for mercury preservation. Aerochem Metrics, Inc. Model 301(ACM) precipitation collectors (modified for MDN) have an auxiliary collection chimney that may be fitted with polyethylene funnels and bottles for collection of additional samples that are not preserved. Excess samples from both networks were obtained for gamma spectrometry analysis of radioactive isotopes.

Archive of MDN excess samples in Washington and Alaska was implemented on March 18, 2011. MDN samples were treated with 1 percent (volume : volume) bromine monochloride (BrCl) for mercury analysis, and the unused portion was split into clean borosilicate glass bottles for gamma spectrometry analysis of radioactive isotopes. In addition, polyethylene bottles were shipped to MDN site AK00 at Dutch Harbor, Alaska, and 18WA in Seattle, Wash. These bottles were installed in the auxiliary sample collection chimneys of the precipitation collectors. Samples collected in the polyethylene bottles were not preserved, and they were shipped first to the Mercury Analytical Laboratory (HAL) at Frontier Global Sciences, Inc. in Seattle, Wash. and then to Denver, Colo. for gamma spectrometry analysis.

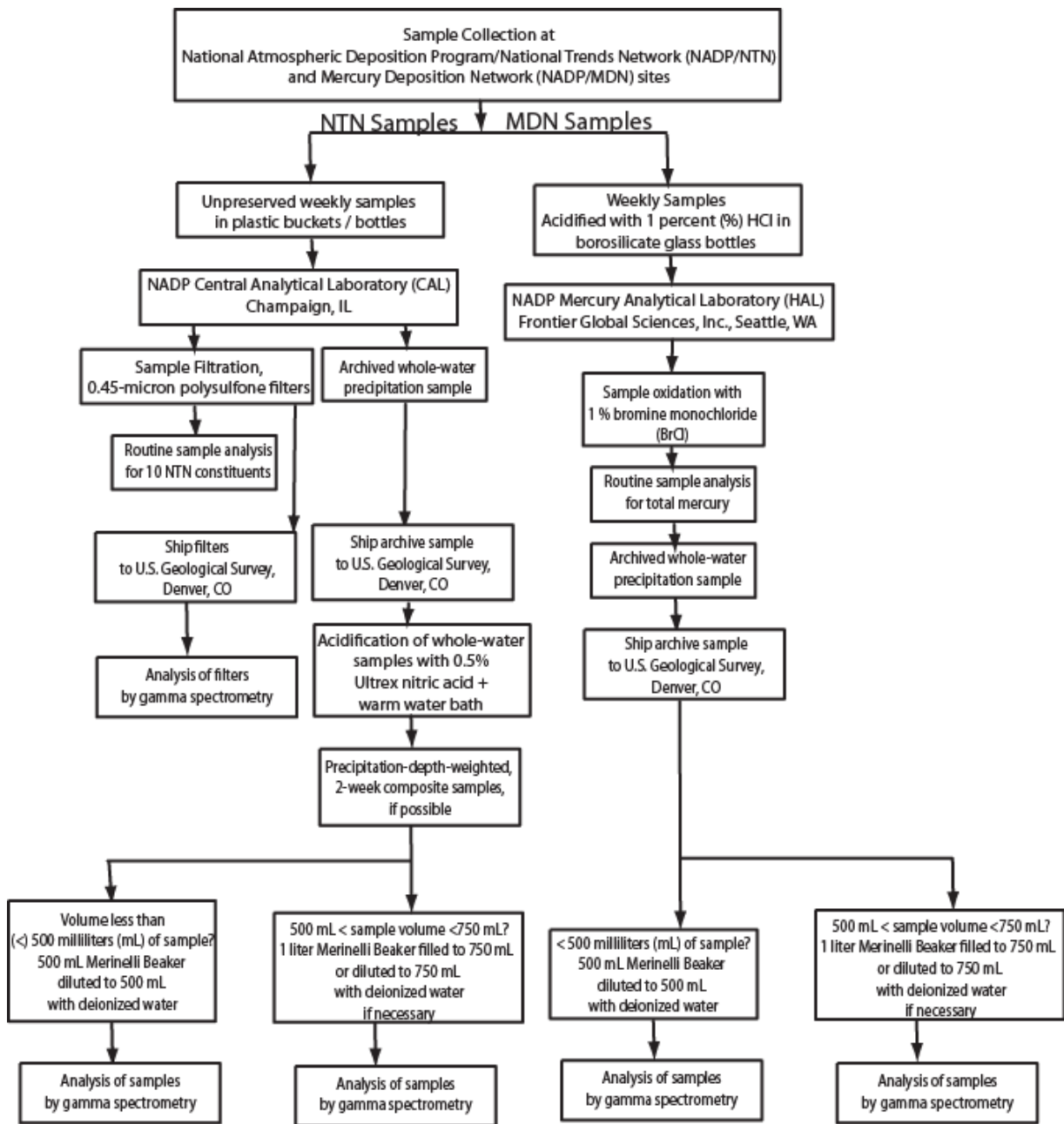
### **Analysis of Filtered Particulates**

Samples from NTN are decanted from the collector buckets to 1-L Nalgene bottles in the field and shipped without preservation to the NADP Central Analytical Laboratory (CAL) located in Champaign, Ill. CAL filters approximately 250 mL of NTN samples through 47-mm diameter, 0.45-micron ( $\mu\text{m}$ ) pore-size, polysulfone filters. CAL saved the filters from NTN samples for the weeks ending March 15, March 22, and March 29, 2011 in labeled plastic petri dishes and shipped them to Denver, Colo., for gamma spectrometry analysis. Radioactive isotopes released at the Fukushima Dai-ichi Nuclear Power Plant were assumed to have affinity for adsorption onto aerosol particles, which are washed out of the atmosphere in wet deposition. Therefore, the NTN filters were initially targeted for gamma spectrometry analysis. As shown later herein, this assumption proved to be invalid.

### **Analysis of Whole-Water Wet Deposition Samples**

After analysis of the filter samples was completed, the archived, unfiltered NTN wet-deposition samples for the weeks ending March 22 and March 29, 2011 were shipped to the USGS Branch of Quality Systems (BQS), located in the USGS National Water Quality Laboratory in Denver, Colo. BQS added Ultrex nitric acid, obtained from J.T. Baker, Center Valley, Penn., to each 1-L Nalgene container according to sample volume (estimated by sample weight) such that each sample contained approximately 0.5 percent nitric acid by volume. The samples were shaken and then placed in a warm-water bath for 1 hr to desorb radioactive isotopes from the bottle walls. The acidified samples were removed from the bath and allowed to leach for 24 hr prior to compositing of samples by site.

Precipitation-depth-weighted, 2-week composite samples were made for each site by measuring aliquots from the acidified weekly samples in proportion to the measured precipitation depths for each week. For example, if 6 mm and 4 mm of precipitation were measured for the weeks ending March 22 and March 29, respectively, then 60 percent of the 2-week composite volume was obtained from the March 22 sample and 40 percent was obtained from the March 29 sample. For some sites, no precipitation was measured during one of the weeks, in which case the 2-week composite was obtained exclusively by the sample for the week with precipitation. There were also instances where precipitation was sampled for both weeks, but insufficient volume was available for archive. In these cases, the entire sample volumes were analyzed, and it was noted that no compositing was possible. Figure 3 illustrates how NADP samples were processed for analysis of radionuclides. Table 1 provides information for the representation and composition of each sample. For convenience, table 1 also lists any fission-product isotopes detected at each site.



**Figure 3.** Flow diagram of sample processing for analysis of radionuclides in National Atmospheric Deposition Program / National Trends Network and Mercury Deposition Network samples.

**Table 1.** NADP weekly wet-deposition samples, March 8, 2011–April 5, 2011, for gamma spectrometry analysis. [NADP, National Atmospheric Deposition Program; NTN, National Trends Network; MDN, Mercury Deposition Network; Trace, auxiliary trace metal sample; mm, millimeters; mL, milliliters; Cs-134, cesium-134; Cs-137, cesium-137; I-131, iodine-131]

NADP site	Network	Represented sampling period	Total precipitation (mm)	Radioactive isotopes detected	Analyzed sample volume (mL)
<b>Samples for NTN sites with 1 dry week and standard weekly MDN samples</b>					
18WA	MDN / Trace	3/15–3/22/11	15.5	-	789
18WA	MDN / Trace	3/22–3/29/11	13.7	-	10
18WA	MDN / Trace	3/29–4/05/11	34.3	-	66
AK00	MDN	3/16–3/23/11	86.0	Cs-134, Cs-137	302
AK00	MDN	3/23–3/29/11	14.5	-	21
AK02	NTN	3/15–3/22/11	11.2	Cs-137	303
AK05	MDN	3/08–3/17/11	22.4	-	221
AK05	MDN	3/29–4/05/11	32.8	-	271
AK98	MDN	3/22–3/29/11	16.5	Cs-137	57.4
AK98	MDN	3/29–4/06/11	113	-	1,167
AL03	NTN	3/22–3/29/11	11.7	-	457
CA20	MDN	3/22–3/29/11	126	-	1,325
CA20	MDN	3/29–4/05/11	15.2	-	59
CA88	NTN	3/22–3/29/11	58.6	-	634
CAN5	NTN	3/15–3/22/11	12.7	-	520
CO98	NTN	3/22–3/29/11	54.9	Cs-134, Cs-137	222
CT15	NTN	3/15–3/22/11	22.1	-	622
IL11	NTN	3/15–3/22/11	9.1	-	317
IL46	NTN	3/22–3/29/11	4.8	-	128
IL63	NTN	3/22–3/29/11	20.6	Cs-134, Cs-137	500
IL78	NTN	3/22–3/29/11	13.0	Cs-134, Cs-137	549
KY10	NTN	3/22–3/29/11	7.4	-	276
KY99	NTN	3/22–3/29/11	10.7	-	473
MA08	NTN	3/15–3/23/11	26.2	-	629
ME00	NTN	3/15–3/22/11	12.4	-	40

**Table 1.—Continued**

NADP site	Network	Represented sampling period	Total precipitation (mm)	Radioactive isotopes detected	Analyzed sample volume (mL)
<b>Samples for NTN sites with 1 dry week and standard weekly MDN samples</b>					
ME02	NTN	3/15–3/22/11	16.0	-	408
ME04	NTN	3/15–3/23/11	10.1	-	289
ME08	NTN	3/15–3/22/11	10.7	-	256
ME96	NTN	3/15–3/22/11	23.4	-	638
MI48	NTN	3/15–3/22/11	22.9	-	613
MI51	NTN	3/15–3/22/11	13.9	-	562
MI98	NTN	3/15–3/22/11	23.1	-	300
MN08	NTN	3/15–3/22/11	17.3	-	625
MN18	NTN	3/15–3/22/11	7.9	-	89
MN99	NTN	3/15–3/22/11	16.3	-	426
MO03	NTN	3/22–3/29/11	10.7	Cs-134, Cs-137	401
MO05	NTN	3/22–3/29/11	26.4	Cs-134, Cs-137	649
MT00	NTN	3/22–3/29/11	7.1	-	148
NC34	NTN	3/22–3/29/11	17.8	-	673
NE15	NTN	3/22–3/29/11	13.7	-	602
NH02	NTN	3/15–3/22/11	27.9	-	666
NJ00	NTN	3/15–3/22/11	28.7	-	694
NY20	NTN	3/15–3/22/11	22.6	-	583
NY22	NTN	3/15–3/22/11	17.2	-	679
NY29	NTN	3/15–3/22/11	16.5	-	539
PA72	NTN	3/17–3/25/11	34.3	-	657
SC05	NTN	3/22–3/29/11	44.5	-	553
SD04	NTN	3/22–3/29/11	21.6	Cs-134, Cs-137	682
SD99	NTN	3/21–3/29/11	14.4	Cs-137	362
TN04	NTN	3/22–3/29/11	48.5	-	610
TN11	NTN	3/21–3/29/11	40.6	-	724
TN14	NTN	3/22–3/29/11	20.0	Cs-134, Cs-137	577
VI01	NTN	3/15–3/22/11	5.1	-	36
VT01	NTN	3/15–3/22/11	18.0	Cs-134, Cs-137	694
WA03	MDN	3/08–3/15/11	226.	-	1,197
WA03	MDN	3/15–3/22/11	39.4	-	315
WA03	MDN	3/22–3/29/11	45.7	-	293
WA03	MDN	3/29–4/05/11	159.	Cs-134, Cs-137	989
WI99	NTN	3/15–3/23/11	50.8	-	723

Table 1.—Continued

NADP site	Network	Represented sampling period	Total precipitation (mm)	Radioactive isotopes detected	Analyzed sample volume (mL)
<b>Samples for NTN sites with 2 wet weeks, but insufficient volume for 2-week composite.</b>					
01MA	NTN	3/15–3/22/11	20.1	-	448
AZ03	NTN	3/15–3/22/11	18.8	-	170
CA42	MDN	3/22–3/29/11	170.	Cs-134, Cs-137	164
CA67	NTN	3/15–3/22/11	9.7	-	261
CO02	NTN	3/22–3/29/11	109.	Cs-134, Cs-137	290
CO08	NTN	3/15–3/22/11	50.5	-	574
CO10	NTN	3/22–3/29/11	21.8	Cs-134, Cs-137	900
CO19	NTN	3/15–3/22/11	17.5	-	494
CO94	NTN	3/22–3/29/11	8.6	Cs-137	95
CO97	NTN	3/15–3/22/11	23.1	-	342
ID11	NTN	3/24–3/31/11	9.7	-	448
IN22	NTN	3/15–3/22/11	9.1	-	247
KY19	NTN	3/15–3/22/11	12.4	-	538
MA01	NTN	3/15–3/22/11	19.1	-	666
MN23	NTN	3/22–3/29/11	34.3	Cs-137	640
MT98	NTN	3/22–3/29/11	5.3	-	139
NC03	NTN	3/22–3/29/11	27.4	-	711
NC29	NTN	3/22–3/29/11	17.5	-	554
NC35	NTN	3/22–3/29/11	13.5	-	454
NC41	NTN	3/22–3/29/11	16.0	-	355
NJ99	NTN	3/22–3/29/11	16.8	-	522
NV03	NTN	3/15–3/23/11	18.3	-	533
NY98	NTN	3/15–3/22/11	14.0	-	467
OH09	NTN	3/15–3/22/11	12.7	-	458
OH49	NTN	3/15–3/22/11	15.2	-	414
OR18	NTN	3/15–3/22/11	21.8	-	641
SC06	NTN	3/22–3/29/11	60.5	-	678
TN00	NTN	3/22–3/29/11	26.9	-	673
UT01	NTN	3/16–3/22/11	20.8	-	378
UT99	NTN	3/22–3/29/11	14.5	-	204
VA99	NTN	3/22–3/29/11	8.6	Cs-137	111
VT99	NTN	3/15–3/22/11	11.2	-	220
WI09	NTN	3/15–3/22/11	9.7	-	360
WI25	NTN	3/15–3/22/11	8.1	-	221
WI28	NTN	3/15–3/22/11	17.0	-	94
WV05	NTN	3/15–3/22/11	44.7	-	636
WY00	NTN	3/22–3/29/11	106.	-	244



Table 1.—Continued

NADP site	Network	Represented sampling period	Total precipitation (mm)	Radioactive isotopes detected	Analyzed sample volume (mL)
<b>Samples for NTN sites with 2 wet weeks, but insufficient volume for 2-week composite.</b>					
WY08	NTN	3/15–3/22/11	32.8	-	652
WY99	NTN	3/22–3/29/11	9.4	Cs-134, Cs-137	239
<b>NTN precipitation-depth-weighted 2-week composite samples.</b>					
50CA	NTN	3/15–3/29/11	246.	-	1,000
CA28	NTN	3/15–3/29/11	246.	-	775
CA45	NTN	3/15–3/29/11	212.	-	1,000
CA50	NTN	3/15–3/29/11	189.	-	1,000
CA66	NTN	3/15–3/29/11	107.	Cs-134, Cs-137, I-131	1,000
CA75	NTN	3/15–3/29/11	128.	-	1,000
CA76	NTN	3/15–3/29/11	42.9	-	485
CA94	NTN	3/16–3/29/11	60.2	Cs-134, Cs-137	870
CA96	NTN	3/15–3/29/11	231.	-	821
CA99	NTN	3/15–3/29/11	235.	Cs-134, Cs-137, I-131	1,000
CO15	NTN	3/15–3/29/11	29.2	-	481
CO89	NTN	3/15–3/29/11	92.7	Cs-134, Cs-137	303
CO90	NTN	3/15–3/29/11	48.5	Cs-134, Cs-137, I-131	1,000
CO91	NTN	3/15–3/29/11	34.6	-	717
IA08	NTN	3/15–3/29/11	58.9	-	1,000
IA23	NTN	3/15–3/29/11	18.3	-	548
ID02	NTN	3/14–3/29/11	50.1	-	947
ID03	NTN	3/15–3/29/11	20.8	-	687
IL18	NTN	3/15–3/29/11	20.8	-	589
IN20	NTN	3/15–3/29/11	40.6	-	575
KY03	NTN	3/15–3/29/11	12.4	-	210
KY22	NTN	3/15–3/29/11	27.4	-	735
KY35	NTN	3/15–3/29/11	20.1	-	258
MD07	NTN	3/15–3/29/11	36.6	Cs-137	794
MD08	NTN	3/15–3/29/11	38.1	-	715
MD13	NTN	3/15–3/29/11	40.9	-	786
MD15	NTN	3/15–3/29/11	28.5	-	1,000
MD18	NTN	3/15–3/29/11	32.5	-	1,000
MD99	NTN	3/15–3/29/11	31.0	-	660
ME98	NTN	3/15–3/29/11	43.4	-	771
MI26	NTN	3/15–3/29/11	45.2	Cs-137	930
MI52	NTN	3/15–3/29/11	49.2	-	891
MI53	NTN	3/15–3/29/11	34.5	-	733
MN01	NTN	3/15–3/29/11	45.5	Cs-134, Cs-137	1,000
MN27	NTN	3/15–3/29/11	32.8	-	853

Table 1.—Continued

NADP site	Network	Represented sampling period	Total precipitation (mm)	Radioactive isotopes detected	Analyzed sample volume (mL)
<b>NTN precipitation-depth-weighted 2-week composite samples.</b>					
MN28	NTN	3/15–3/29/11	27.2	-	167
MT07	NTN	3/15–3/29/11	14.7	Cs-137	308
MT97	NTN	3/15–3/29/11	48.7	-	751
NC06	NTN	3/15–3/29/11	18.1	-	236
NC25	NTN	3/15–3/29/11	103.	-	620
NC45*	NTN	3/08–3/29/11	88.2	-	1,000
NY01	NTN	3/15–3/29/11	48.5	Cs-134, Cs-137	1,000
NY08	NTN	3/15–3/29/11	24.6	-	552
NY10	NTN	3/15–3/29/11	47.0	Cs-134, Cs-137	1,000
NY68	NTN	3/15–3/29/11	33.5	-	740
NY96	NTN	3/15–3/29/11	17.5	-	252
NY99	NTN	3/15–3/29/11	38.8	-	297
OH17	NTN	3/15–3/29/11	20.9	-	855
OH54	NTN	3/15–3/29/11	14.0	-	606
OH71	NTN	3/15–3/29/31	16.3	-	527
OR10	NTN	3/15–3/29/11	173.	Cs-137	1,002
OR97	NTN	3/15–3/29/11	68.6	-	753
PA00	NTN	3/15–3/29/11	26.7	-	340
PA15	NTN	3/15–3/29/11	48.8	-	801
PA18	NTN	3/15–3/29/11	52.6	-	1,000
PA29	NTN	3/15–3/29/11	50.0	-	1,000
PA42	NTN	3/15–3/29/11	49.1	-	1,000
PA47	NTN	3/15–3/29/11	30.0	-	1,000
PR20	NTN	3/15–3/29/11	60.5	-	583
VA00	NTN	3/15–3/29/11	26.4	-	1,000
VA13	NTN	3/15–3/29/11	31.7	-	1,000
VA24	NTN	3/15–3/29/11	28.7	-	529
VA28	NTN	3/15–3/29/11	31.3	-	982
VA98	NTN	3/15–3/29/11	30.2	-	102
WA14	NTN	3/15–3/29/11	97.5	-	1,000
WA19	NTN	3/15–3/29/11	78.0	-	639
WA21	NTN	3/15–3/29/11	45.4	-	838
WA24	NTN	3/15–3/29/11	38.8	-	901
WA98	NTN	3/15–3/29/11	126.	Cs-134, I-131	1,000
WA99	NTN	3/16–3/29/11	54.6	Cs-134, I-131	1,000
WI10	NTN	3/15–3/29/11	49.7	-	930

\*3-week composite sample for NC45

**Table 1.—Concluded**

NADP site	Network	Represented sampling period	Total precipitation (mm)	Radioactive isotopes detected	Analyzed sample volume (mL)
<b>NTN precipitation-depth-weighted 2-week composite samples.</b>					
WI35	NTN	3/15–3/29/11	56.2	-	807
WI36	NTN	3/14–3/29/11	24.8	-	818
WI37	NTN	3/15–3/29/11	40.1	Cs-137	978
WI98	NTN	3/15–3/29/11	57.1	-	1,000
WV04	NTN	3/15–3/29/11	35.5	-	981
WV18	NTN	3/15–3/29/11	59.2	-	842
WY02	NTN	3/15–3/29/11	11.4	-	401

Weekly MDN samples were not composited into 2-week precipitation-depth-weighted composite samples. MDN samples are already acidified when they are returned from the field, and they are treated with BrCl at the HAL. Therefore, MDN samples were analyzed by gamma spectrometry with no pre-processing by BQS. The MDN samples analyzed for radioactive isotope content are also identified in table 1.

### Sample Analysis by Gamma Spectrometry

Samples were analyzed by USGS at the National Reactor Facility at the Denver Federal Center, Denver, Colo., using gamma spectrometry with high-purity germanium (HPGe) detector systems. Because there were only two detectors available, analysis by gamma spectrometry occurred from about 3 to 21 weeks after the samples were collected from the NADP sites. The delay between collection and analysis for the first wet-deposition sample analyzed was 20 days and 5 hr for site CA99, completed at 5:00 p.m. on April 18, 2011. The delay between collection and analysis for the last sample analyzed was 145 days, completed on August 7, 2011 for site WA98. Two HPGe detectors were used. The first was a Canberra 14-percent relative efficiency detector and the second was an Ortec 40-percent relative efficiency detector. Detector efficiency calibrations were done for the sample-to-detector geometries that were used in this study. The detectors are kept at liquid-nitrogen temperature to eliminate thermal noise problems. Background radiation is reduced with a lead shield that surrounds the sample and detector.

The energy response calibration is performed with Eu-152 check sources because it emits a wide energy range of gamma rays, with a useable range from approximately 122 keV (thousand electron volts) to 1.528 MeV (million electron volts). Energy calibration checks were performed several times a day to ensure that the gamma peaks were within 1 keV of the measured values. Efficiency calibrations were done with National Institute of Standards and Technology (NIST) traceable calibration standards manufactured by Eckert & Ziegler, Atlanta, Ga. The efficiency calibrations were performed once and those calibration curves were then stored in the analyzer's internal memory.

Two standards were used for the calibration geometry identical to the 47-mm diameter filters: (1) 0.5 microCurie of cobalt-60 and (2) 0.055 microCurie of cesium-137 plus 0.575 microCurie of barium-133. These isotopes cover a gamma energy range from 88 keV to 1,332 keV. Each weekly filter sample obtained for the weeks ending March 22 and March 29, 2011 was analyzed in its petri dish container. Weekly filter samples were also composited by stacking them on the detector by site or

grouping them together regionally in plastic bags. Finally, all of the filters were composited together in a bag, and the bag was counted for 24 hr.

For the water samples, two standards were used: (1) 500-mL volume and (2) 1-L volume in standard Marinelli beakers. These standards each contained approximately one microCurie of Eu-152 that was uniformly dispersed in the Marinelli beaker. All water samples were put into Marinelli beakers for the spectroscopy measurements. Two sizes of Marinelli beakers were used: (1) 500 mL and (2) 1 L. If the sample volume was less than 500 mL and not more than 750 mL, then a 500-mL beaker was used, and deionized water was added if necessary to achieve a 500-mL volume. If the sample volume was greater than 750 mL, then 750 mL of sample was used in a 1-L Marinelli beaker without dilution. The 750-mL volume was used instead of the full 1 L because more radioactive material is physically closer to the detector for the 750-mL volume than for 1 L. Data collection times were at least 6 hr per sample. If manual observation of the gamma spectrum revealed a potential gamma peak for a fission-product or activation-product (for example, cobalt-60, iron-59) isotope after 6 hr, then data collection was continued for a longer time until sufficient data were obtained for quantification of peak areas.

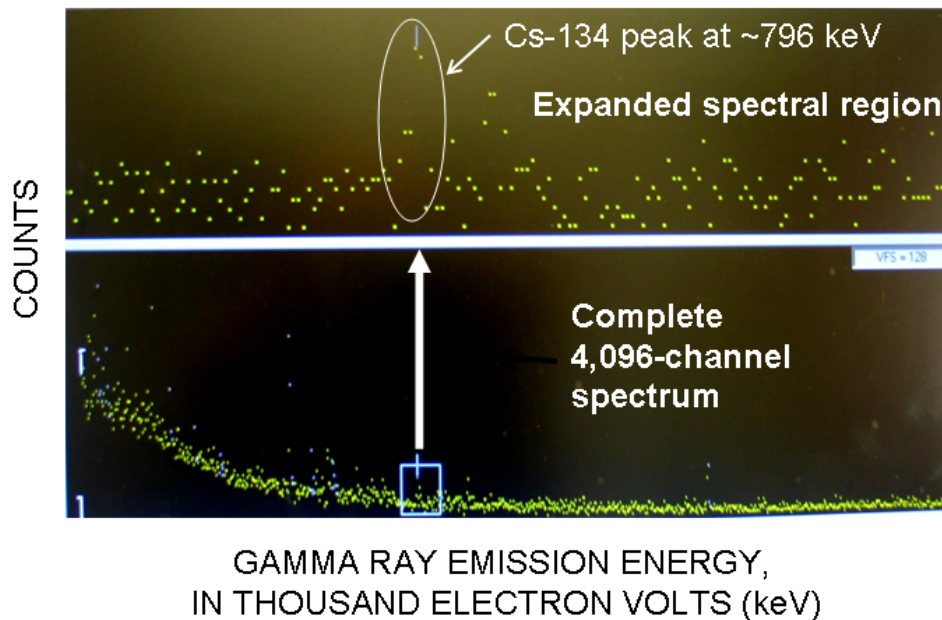
The automated gamma spectrum analyzer software uses a peak search algorithm to identify gamma ray peaks by shape and specified ratios of isotope-specific peak area to background. The energy of each peak corresponds to the radioisotope that emits the associated gamma rays, and the area of the peak correlates to the amount of the radioisotope present in the sample. The analyzer provides the identified isotopes and their radioactivity levels (referred to as “activity” in this report) as a part of its output. The automated analyzer identified Cs-134 in one sample for CO90. Subsequently, USGS personnel manually reanalyzed the gamma spectra and qualitatively identified Cs-134 peaks for an additional 23 sites. Because these peaks were manually identified, error associated with Cs-134 quantification cannot be evaluated. Therefore, these 23 manually derived Cs-134 values should be regarded as estimated. An example of a manually quantified Cs-134 peak for the sample from SD04 is shown in figure 4.

I-131 activities were adjusted for decay occurring between the sample-analysis stop time to the date and time when the collector was last open during precipitation per NADP records. The I-131 decay was calculated by determining the time between the collection and the analysis, calculating the number of half-lives that time period represented, and then dividing the analyzed I-131 activity by the value given by:

$$\text{I-131 activity at collection time} = \frac{A_{pCi/L}}{0.5^{\text{halfives}}} ; \quad (3)$$

where:  $A_{pCi/L}$  is activity of I-131 obtained at stop time of analysis, in picoCuries per liter (pCi/L), and  $halfives$  is number of half-lives calculated between stop time of sample analysis and noon of day when last 2.5 millimeters (mm) of precipitation fell.

The day and time (Mountain Standard) of the last 2.5 mm of precipitation was March 26 at 3:00 p.m. for CA66, March 27 at 12:45 a.m. at CA99, March 28 at 11:00 a.m. for CO90, March 29 at 9:30 a.m. at WA98, and March 29 at 6:45 a.m. at WA99. Cs-134 and Cs-137 activities were not adjusted for decay to the time of sample collection because radioactive decay during the 2-month period between collection and analysis was negligible for these isotopes (Wakeford, 2011).



**Figure 4.** Gamma ray spectra for wet-deposition sample from National Atmospheric Deposition Program site SD04 (March 22–29) with manually quantified cesium-134 peak.

### Quality Assurance

The following protocols were implemented to assure the quality of the data. Four blank filter samples were analyzed; two filters were rinsed by filtering 250 mL of deionized water, and the other two were rinsed with a laboratory-prepared synthetic rainwater solution. No radioactive isotopes were detected in these filters.

Four blank water samples were prepared using randomly selected, cleaned, 1-L NADP Nalgene sample bottles and filling them with the same deionized water used to dilute the samples for gamma spectrometry analysis. These samples were acidified with 5 mL of the same Ultrex nitric acid used to acidify the real samples. No radioactive isotopes were detected in these samples.

To establish that there were no fission products in samples prior to the reported arrival of air masses containing emissions from the incident, archived wet-deposition samples for the week March 8–15, 2011 for selected NTN sites were analyzed. Samples from sites CO90, IL63, NY10, VT01, and WA98—where Cs-137 was detected during the March 15–29, 2011 period—were analyzed by gamma spectrometry. No fission or activation products were detected in these samples.

Replicate samples were obtained from sites where co-located precipitation collectors are operated for comparison of instrumentation and estimation of variability in NTN measurements. Three pairs of co-located sites in operation during the study period are MA01 and 01MA at Cape Cod National Sea Shore, Mass.; CA50 and 50CA at Sage Hen Creek Field Station near Truckee, Calif; and CO98 and CO89 at Loch Vale, Rocky Mountain National Park, Colo. No fission products were detected at either of the co-located sites at MA01/01MA and CA50/50CA. Conversely, Cs-137 was detected for both CO98 (22 pCi/L) and CO89 (11 pCi/L). The CO98 and CO89 collectors are approximately 15 m apart, and Cs-137 activities differed by a factor of 2. The CO98 sample volume was obtained only from the week ending March 29, 2011 (54.9 mm precipitation depth) because the collector missed a precipitation

event due to power loss during the previous week. The CO89 sample was a true 2-week composite representing 37.8 mm of additional precipitation during the week ending March 22, 2011. Cs-134 activities were similar at CO98 (7.2 pCi/L) and CO89 (8.9 pCi/L).

## **Fission Products**

None of the filter samples contained detectable fission or activation products regardless of compositing method, including compositing by site and region. Finally, all of the filters were grouped together in a bag-type geometry that was draped over the detector and counted for 24 hr, and no fission products were detected.

Fission products were detected in whole-water precipitation samples from 35 individual NADP sites (tables 1 and 2). Active NTN and MDN sites not listed in tables 1 and 2 did not collect sufficient sample volumes for analysis. Table 1 lists all 167 sampling sites and shows those for which no fission products were detected in whole-water wet-deposition samples. Table 2 provides a summary of the results for fission products detected in the NTN and MDN wet-deposition samples collected during March 15–April 5, 2011 from 35 individual NADP sites.

Spatial distributions of measured fission product isotope activities and calculated deposition values were mapped using a geographic information system (ESRI, 2010). The spatial distributions of I-131 activities and deposition values for sites where I-131 was detected are shown in figures 5 and 6. The spatial distributions of measured Cs-137 and Cs-134 activities detected in wet-deposition samples and the calculated deposition values are shown in figures 7–10 for the 48 contiguous United States and in figure 11 for Alaska.

**Table 2.** Fission product isotope activities, associated precipitation depths, and calculated deposition values for National Atmospheric Deposition Program wet-deposition samples collected during March 15–April 5, 2011.

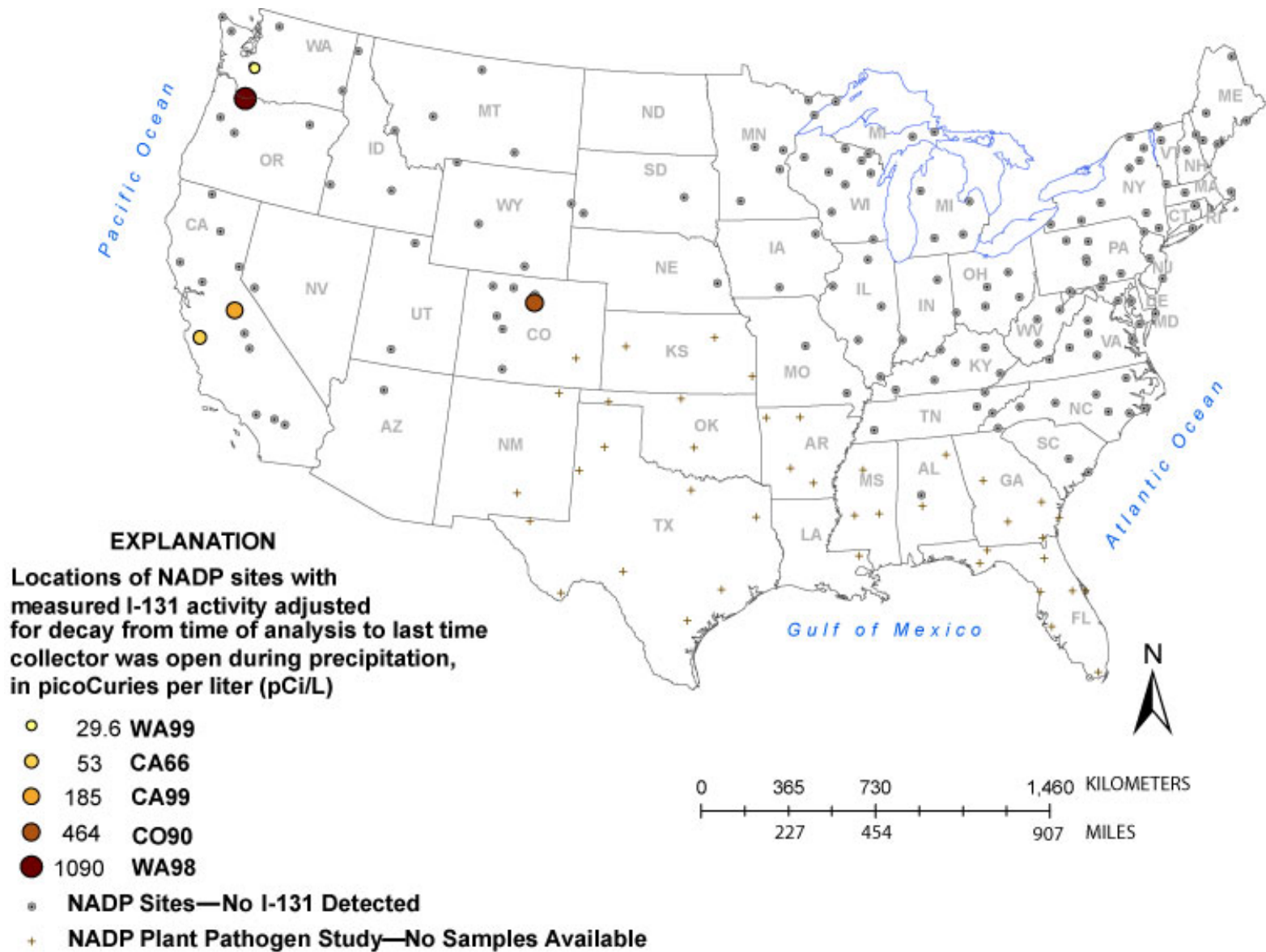
[NADP, National Atmospheric Deposition Program; mm, millimeters; C-134s, cesium-134; Cs-137, cesium-137; I-131, iodine-131; pCi/L, picoCuries per liter; Bq/m<sup>2</sup>, Becquerels per square meter; 1 pCi = 0.037 Bq; I-131 values are adjusted for radioactive decay from time of sample analysis to noon on the last day when at least 2.5 mm of precipitation was measured]

NADP Site	North Latitude (decimal degrees)	West Longitude (decimal degrees)	Sampling Period	Total Precipitation Depth (mm)	I-131 Activity (pCi/L)	Estimated Cs-134 Activity (pCi/L)	Cs-137 Activity (pCi/L)	I-131 Deposition (Bq/m <sup>2</sup> )	Estimated Cs-134 Deposition (Bq/m <sup>2</sup> )	Cs-137 Deposition (Bq/m <sup>2</sup> )
AK00	53.8454	-166.5048	3/16–3/23/11	86		55	8.6		180	27
AK02	58.5139	-134.7843	3/15–3/22/11	11			7.6			3.1
AK98	57.7189	-152.5617	3/22–3/29/11	17			26			16
CA42	34.2071	-117.7618	3/22–3/29/11	170		7.3	39		46	240
CA66	36.4834	-121.1570	3/15–3/29/11	107	53.0	2.6	9.3	211	10	37
CA94	34.1938	-116.9131	3/16–3/29/11	60.2		1.1	3.0		2.6	6.7
CA99	37.7961	-119.8581	3/15–3/29/11	235	185		3.4	1,610		30
CO02	40.0547	-105.5891	3/22–3/29/11	109		11	10		43	42
CO10	38.9581	-106.9850	3/22–3/29/11	21.8		1.0	3.9		0.81	3.1
CO89	40.2878	-105.6628	3/15–3/29/11	92.7		8.9	11		31	37
CO90	40.0358	-105.5440	3/15–3/29/11	48.5	464	14	12	833	25	22
CO94	39.9939	-105.4800	3/22–3/29/11	8.6			14			4.4
CO98	40.2878	-105.6628	3/22–3/29/11	54.9		7.2	22		15	45
IL63	37.4356	-88.6719	3/22–3/29/11	20.6		6.4	15		4.9	12
IL78	40.9333	-90.7231	3/22–3/29/11	13.0		1.0	2.6		.47	1.2
MD07	39.6470	-77.4848	3/15–3/29/11	36.6			2.5			3.4
MI26	42.4103	-85.3928	3/15–3/29/11	45.2			7.0			12
MN01	45.4017	-93.2031	3/15–3/29/11	45.5		1.5	2.8		2.5	4.7
MN23	46.2494	-94.4972	3/22–3/29/11	34.3			3.0			3.8
MO03	38.7536	-92.1994	3/22–3/29/11	10.7		2.2	9.2		.89	3.6
MO05	36.9108	-90.3186	3/22–3/29/11	26.4		.80	0.80		.78	.78
MT07	46.4850	-112.0647	3/15–3/29/11	14.7			1.9			1.1
NY01	42.2280	-77.8016	3/15–3/29/11	48.5		.40	2.4		0.72	4.3
NY10	42.2994	-79.3964	3/15–3/29/11	47.0		1.8	3.1		3.1	5.4

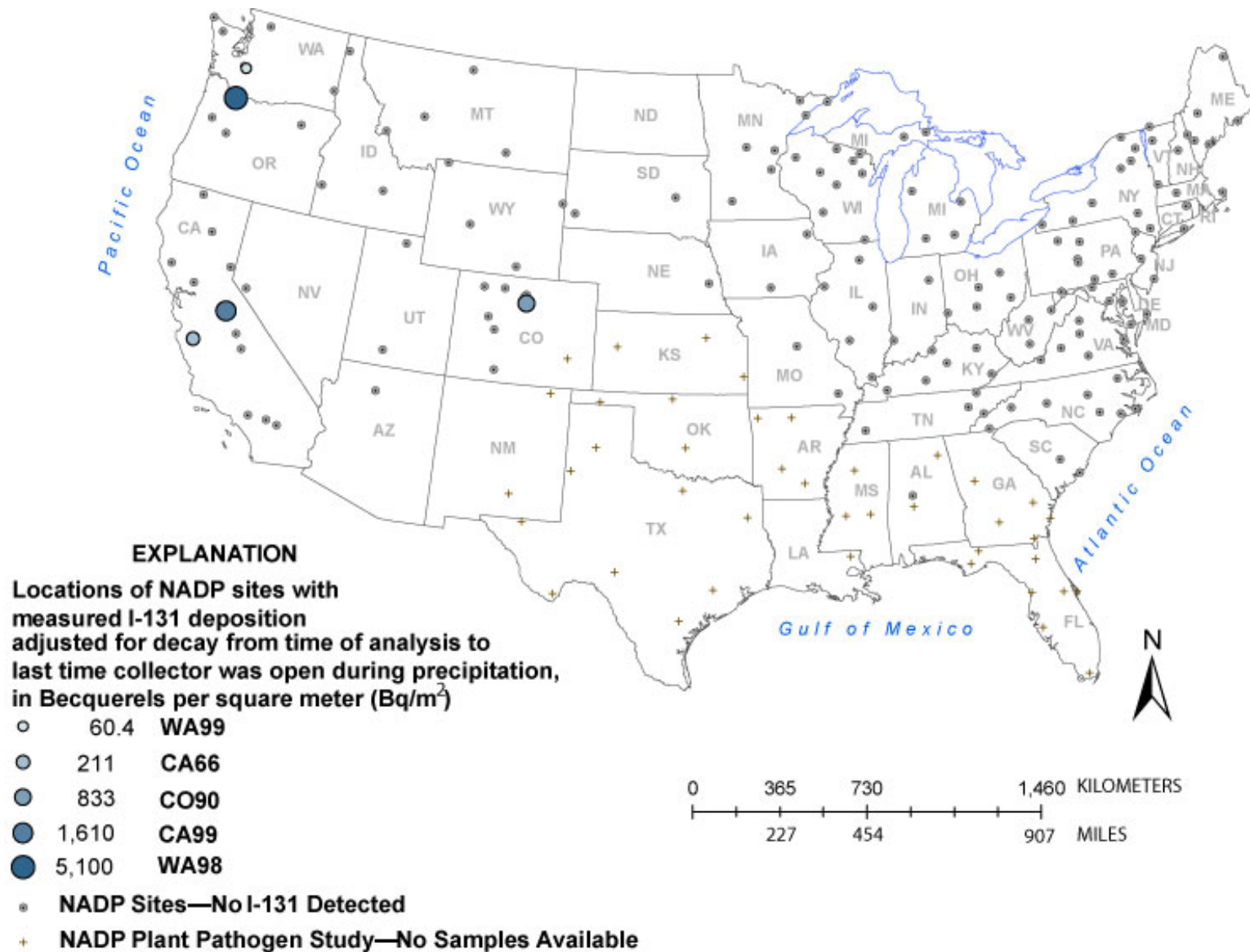
**Table 2.—Continued**

<b>NADP Site</b>	<b>North Latitude (decimal degrees)</b>	<b>West Longitude (decimal degrees)</b>	<b>Sampling Period</b>	<b>Total Precipitation Depth (mm)</b>	<b>I-131 Activity (pCi/L)</b>	<b>Estimated Cs-134 Activity (pCi/L)</b>	<b>Cs-137 Activity (pCi/L)</b>	<b>I-131 Deposition (Bq/m<sup>2</sup>)</b>	<b>Estimated Cs-134 Deposition (Bq/m<sup>2</sup>)</b>	<b>Cs-137 Deposition (Bq/m<sup>2</sup>)</b>
OR10	44.2118	-122.2560	3/15–3/29/11	173			0.70			4.5
SD04	43.5577	-103.4835	3/22–3/29/11	21.6		10	20		8.0	16
SD99	44.3550	-98.2917	3/21–3/29/11	14.4			3.9			2.1
TN14	35.4688	-89.1713	3/22–3/29/11	20.0		2.0	11		1.5	8.1
VA99	37.6265	-79.5126	3/22–3/29/11	8.6			13			4.0
VT01	42.8761	-73.1633	3/15–3/22/11	18.0		0.86	3.0		0.60	2.0
WA03	48.2892	-124.6519	3/29–4/05/11	159		5.0	1.2		29	7.1
WA98	45.5639	-122.2089	3/15–3/29/11	126	1,090	1.4		5,100	6.5	
WA99	46.7582	-122.1243	3/16–3/29/11	54.6	29.6	1.3		60.4	2.7	
WI37	45.8228	-91.8744	3/15–3/29/11	40.1			1.9			2.9
WY99	43.8730	-104.1917	3/22–3/29/11	9.4		13	24		4.4	8.4

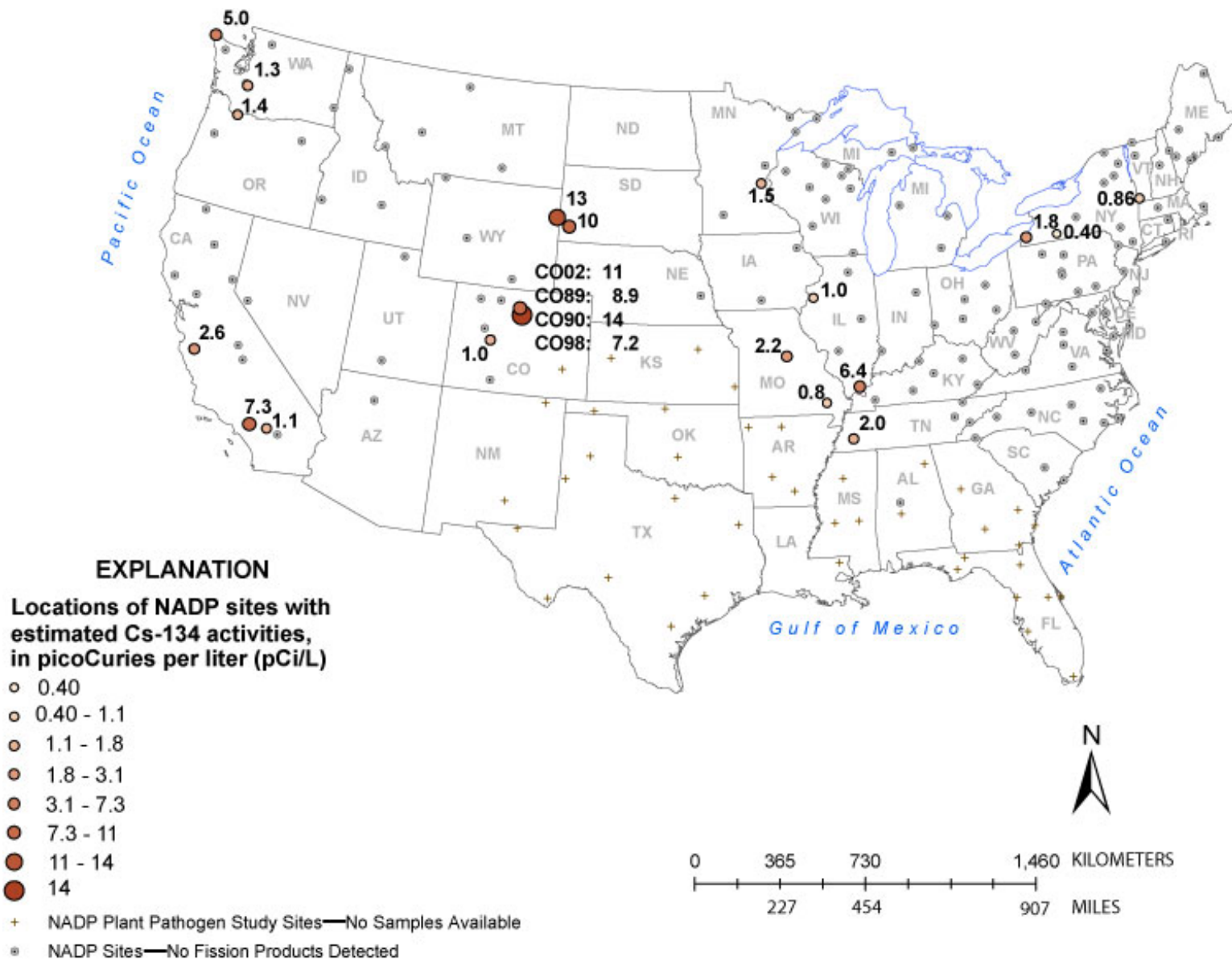




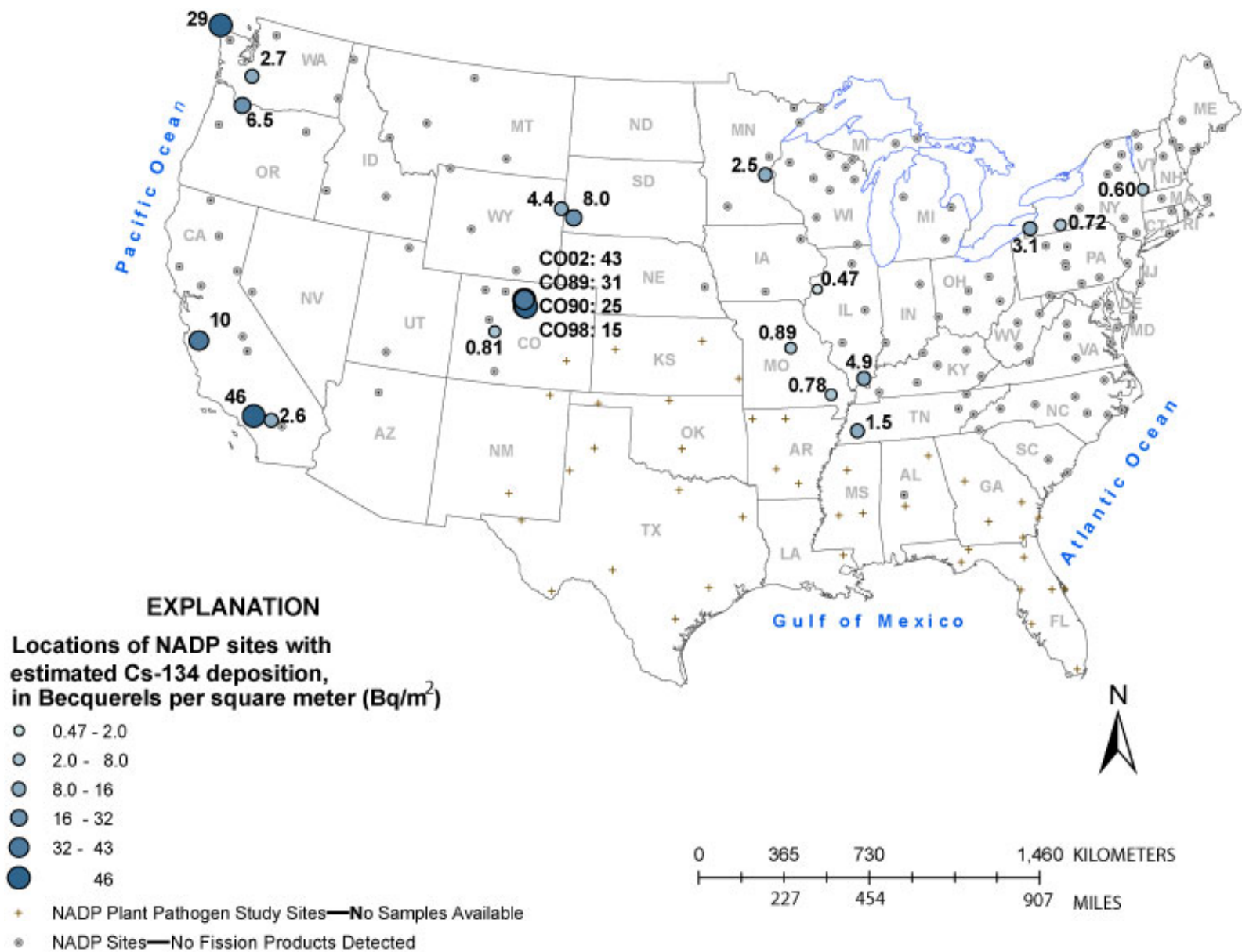
**Figure 5.** Spatial distribution of iodine-131 (I-131) activities in National Atmospheric Deposition Program (NADP) samples from the 48 contiguous United States during March 15–29, 2011.



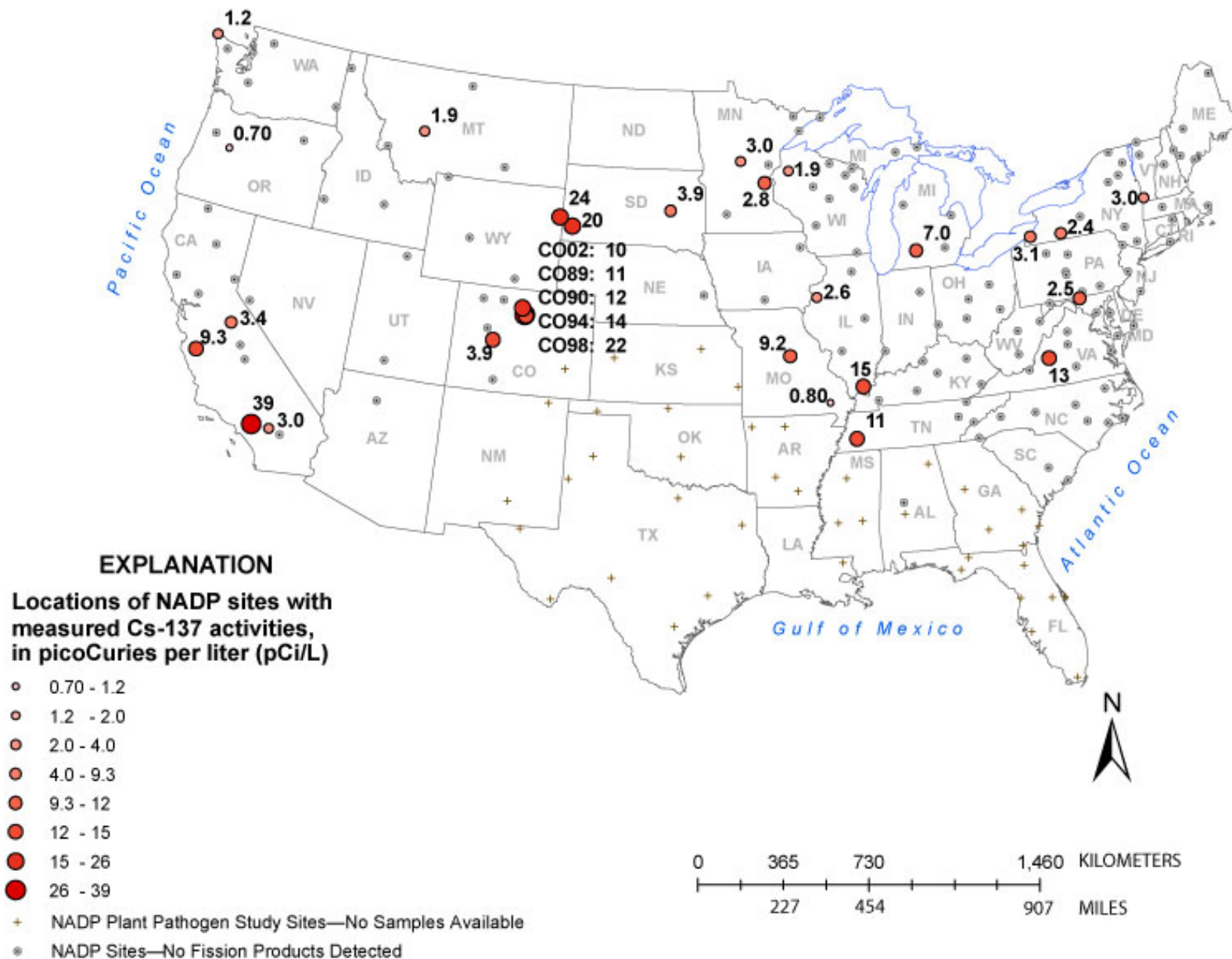
**Figure 6.** Spatial distribution of iodine-131 (I-131) deposition calculated from measured activities and rain-gage depths from National Atmospheric Deposition Program (NADP) sites in the 48 contiguous United States during March 15–April 5, 2011.



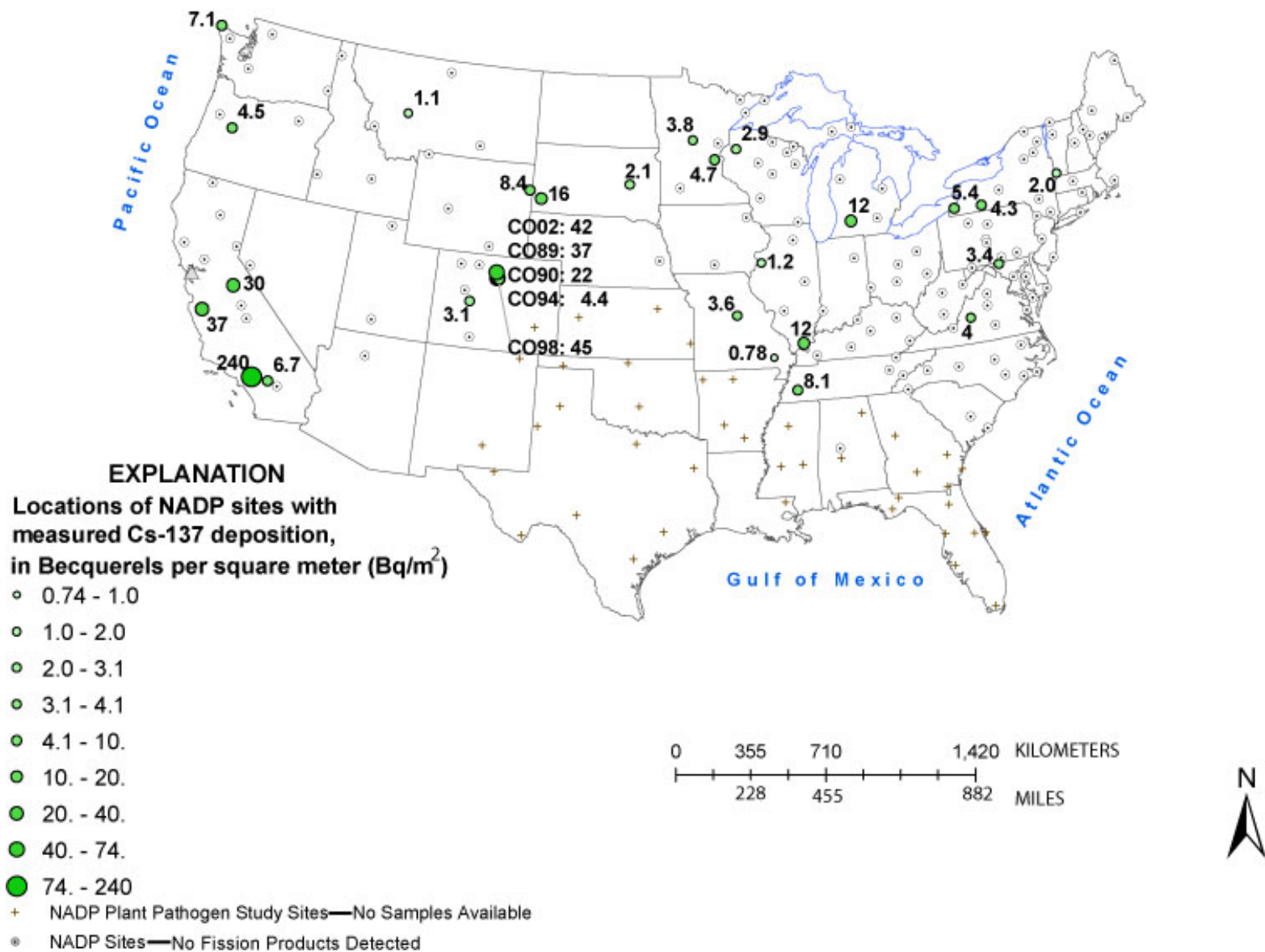
**Figure 7.** Spatial distribution of estimated cesium-134 (Cs-134) activities in National Atmospheric Deposition Program (NADP) samples from the 48 contiguous United States during March 15–29, 2011.



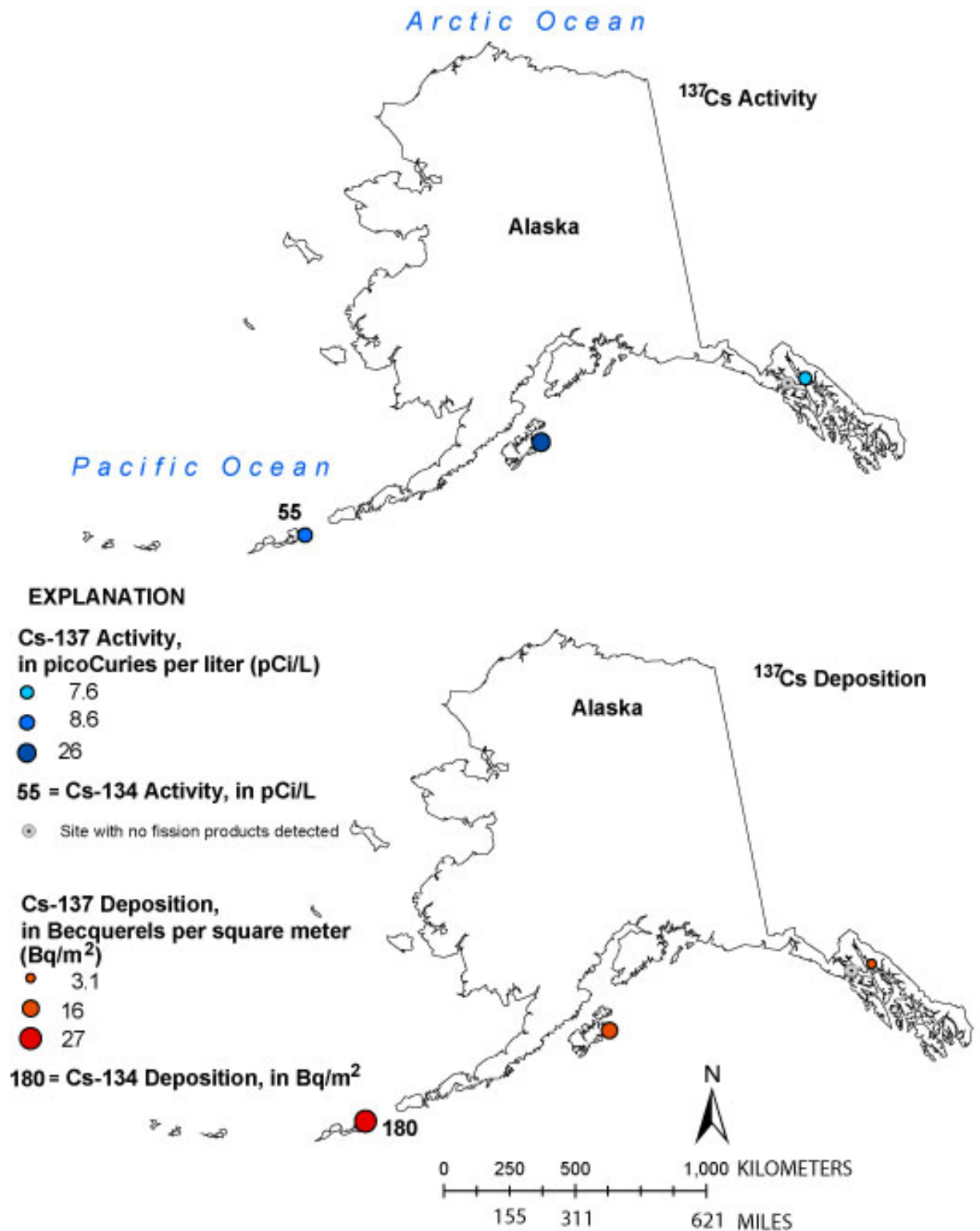
**Figure 8.** Spatial distribution of cesium-134 (Cs-134) deposition calculated from estimated activities and measured rain-gage depths from National Atmospheric Deposition Program (NADP) sites in the 48 contiguous United States during March 15–April 5, 2011.



**Figure 9.** Spatial distribution of detected cesium-137 (Cs-137) activities in National Atmospheric Deposition Program (NADP) samples from the 48 contiguous United States during March 15–29, 2011.



**Figure 10.** Spatial distribution of cesium-137 (Cs-137) deposition calculated from measured activities and rain-gage depths from National Atmospheric Deposition Program (NADP) sites in the 48 contiguous United States during March 15–April 5, 2011.



**Figure 11.** Spatial distribution of cesium-134 (Cs-134) and cesium-137 (Cs-137) measured activities and calculated deposition values from National Atmospheric Deposition Program (NADP) sites in Alaska during March 15–April 5, 2011.

## Summary and Conclusions

This study demonstrated the flexibility of the NADP in conjunction with USGS capabilities to monitor a sudden atmospheric release incident originating at the Fukushima Dai-Ichi Nuclear Power Plant near Sendai, Japan on March 12, 2011. Archived wet-only deposition samples were processed. Filtered particulates samples were archived and analyzed by gamma spectrometry for 280 NADP samples. Whole-water precipitation samples (176) were analyzed by gamma spectrometry for 161 NTN and 6 MDN sites. Samples were collected and analyzed for the week before contaminated air arrived over North America (March 8–15, 2011) for five of the same sites where fission-product isotopes were subsequently detected during the period of March 15–April 5, 2011. No fission products were detected in the pre-arrival period samples. Quality-assurance data from analysis of blank samples and co-located site replicate measurements provide confidence in the results.

Filtered particulate samples were analyzed prior to analysis of the water samples. No fission products were detected on individual filters or on composited filters. Therefore, the transported fission products were either on particles smaller than the 0.45- $\mu\text{m}$  pore size of the filter, or they were associated with soluble aerosol salts present in the collected samples.

I-131 was quantified and adjusted for decay to the time of sample collection for five whole-water wet-deposition samples from California, Colorado, and Washington. The activities of the quantified I-131 ranged from 29.6 to 1,090 pCi/L, and calculated deposition values ranged from 211 to 5,100 Bq/m<sup>2</sup>. Several weeks transpired between sample collection and analysis of the I-131 in the water samples, which were prioritized for analysis from west (high priority) to east (low priority). Therefore, I-131 activities likely decayed in most of the samples before they could be measured.

Cs-134 activities were quantified for 23 samples from Alaska, California, Colorado, Illinois, Minnesota, Missouri, New York, South Dakota, Tennessee, Vermont, Washington, and Wyoming. The Cs-134 ranged from 0.40 to 55 pCi/L, and deposition values ranged from 0.47 to 180 Bq/m<sup>2</sup>. Cs-134 values reported for all but one site (CO90 near Nederland, Colo.) have substantial error because they were manually estimated from the gamma-ray spectra. Cs-137 activities were quantified for 33 samples from Alaska, California, Colorado, Illinois, Maryland, Michigan, Minnesota, Missouri, Montana, New York, Oregon, South Dakota, Tennessee, Virginia, Vermont, Washington, Wisconsin, and Wyoming. These samples ranged from 0.70 pCi/L to 39 pCi/L, and calculated deposition values ranged from 0.78 to 240 Bq/m<sup>2</sup>. Cs-134 and Cs-137 activities were not adjusted for decay to date of sample collection because radioactive decay during the 2-month period between collection and analysis was negligible for these isotopes.

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