Radionuclide Deposition: The Fukushima Dai-ichi Nuclear Power Facility Incident

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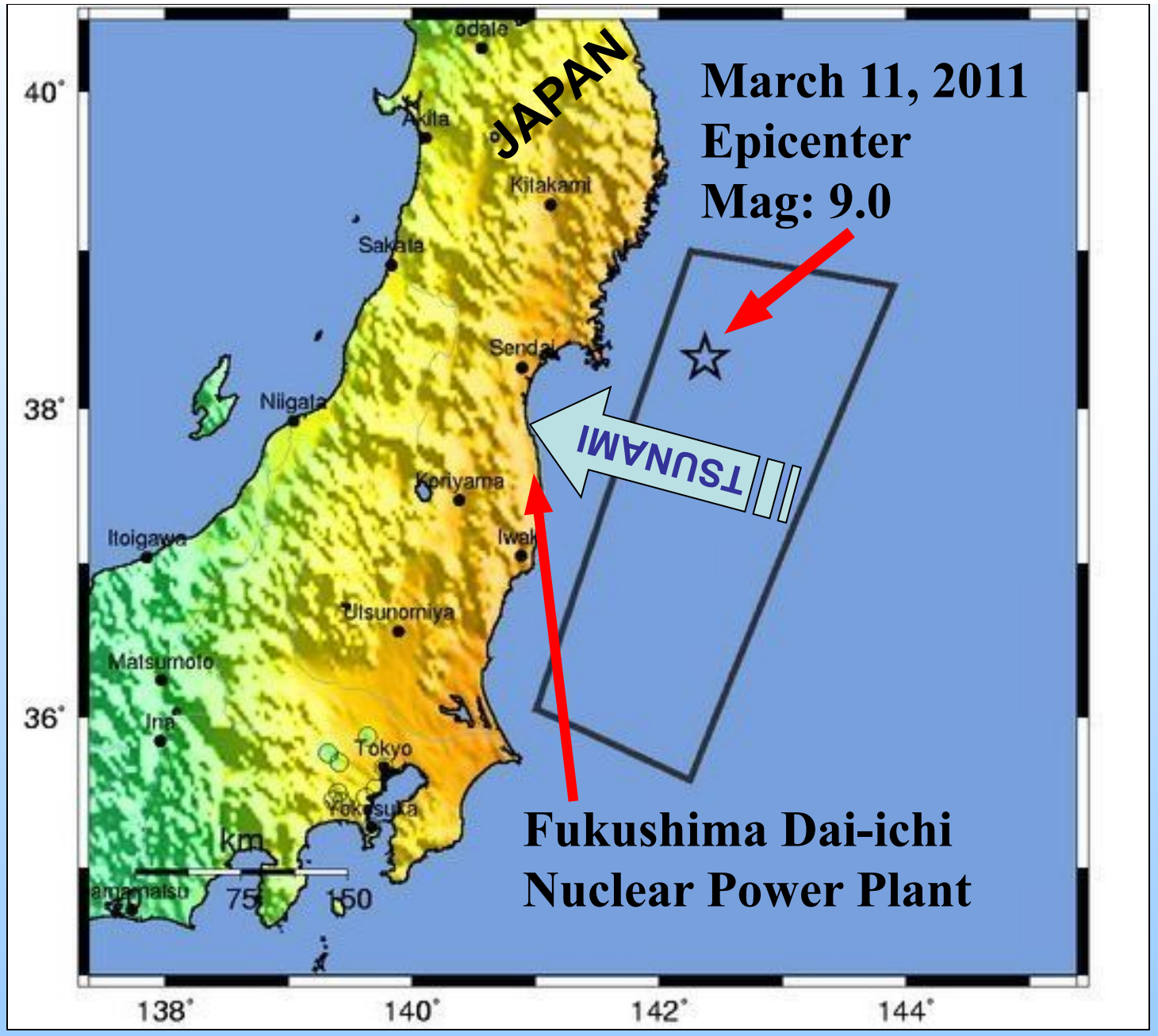
1 National Atmospheric Deposition Program

2 USGS science for a changing world

USGS SHAKEMAP, E. COAST JAPAN, MAR. 11, 2011



Not felt

Light

Extreme

Fukushima Dai-ichi Nuclear Power Plant, Near Sendai, Japan

Timeline

March 12, 2011 – Fukushima Dai-ichi Release(s)

March 14, 2011 – NADP and USGS begin preparation of Sampling and Analysis Plans.

- NADP begins saving filters and water samples (3/8 – 3/15).

March 15, 2011 - NADP/USGS contacted US EPA to offer samples for analysis.
Sent 5 samples on March 28.

March 16, 2011 - NADP/USGS contacted DHS to offer samples for analysis.
No samples sent.

Timeline

March 25, 2011 - USGS Reactor Facility Group (RFG) started analysis of filters.

April 15, 2011 - USGS completes filter analyses

April 18, 2011 - USGS begins water analyses

July 8, 2011 - USGS completes water analyses

August 26, 2011 - Initiated ES&T article and Open-File Report (OFR)

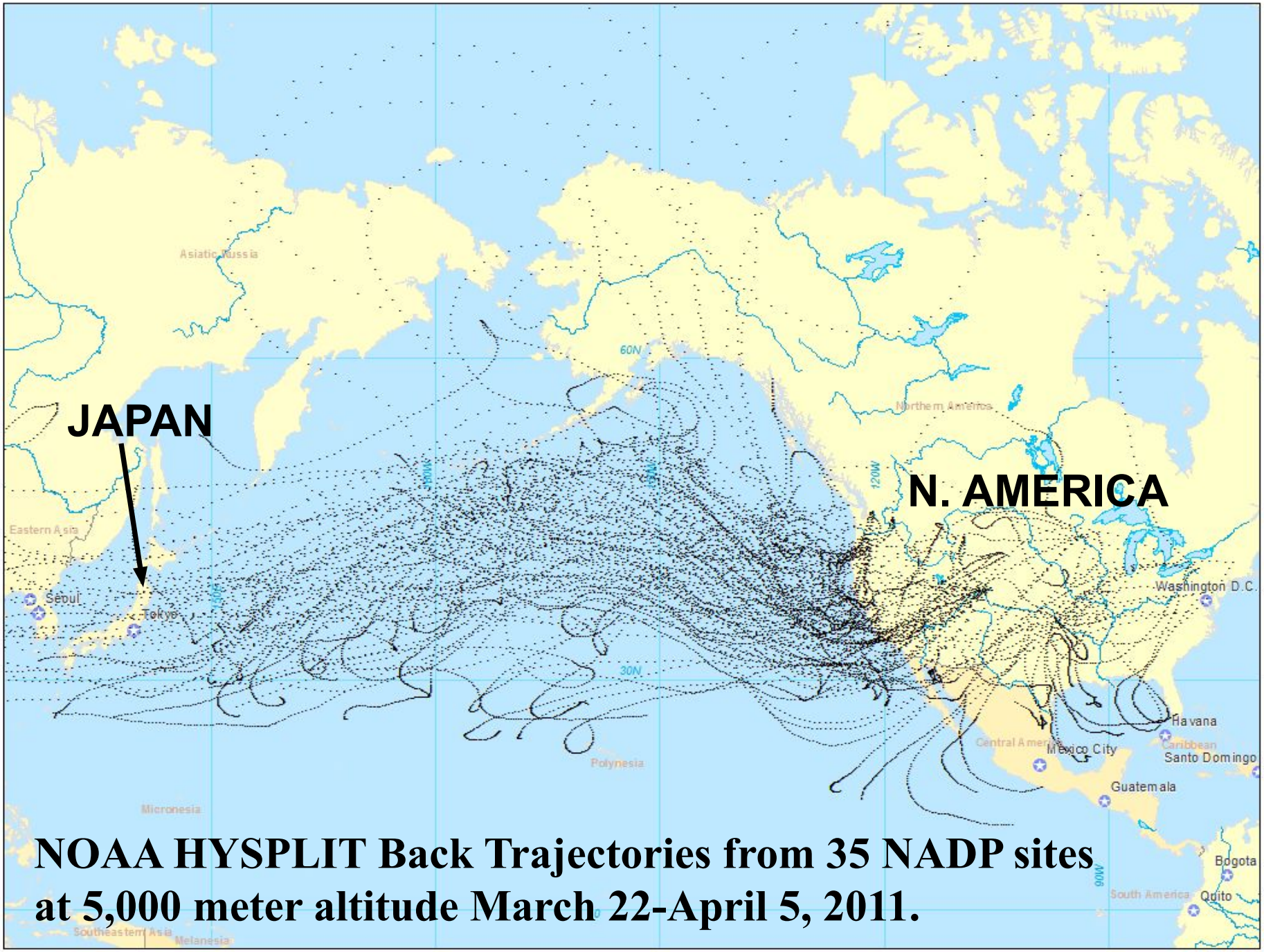
February 2012 - ES&T article and OFR Published

Objectives

1. Evaluate NADP capabilities for monitoring radionuclide activities in precipitation.

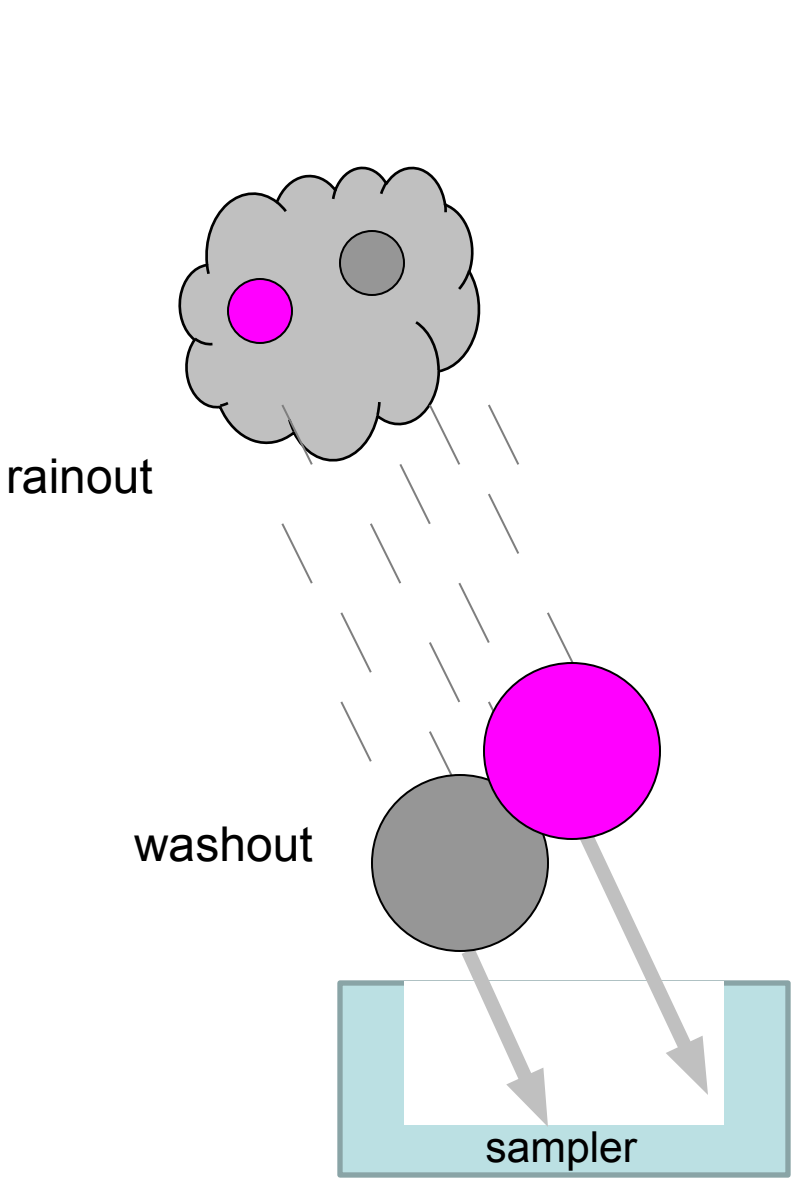
2. Evaluate NADP/USGS capabilities to monitor unexpected atmospheric events.

3. Offer NADP support to agencies responsible for monitoring radioactive fallout – US EPA, DOE, DHS, Environment Canada.



NOAA HYSPLIT Back Trajectories from 35 NADP sites at 5,000 meter altitude March 22-April 5, 2011.

Basic Wet Deposition Sampling



What is the NADP?

National Atmospheric Deposition Program

- A Cooperative Research Program *(Un. Of Illinois)*
  - Measure wet deposition and atmospheric concentrations of pollutants
  - monitor the rate of pollution movement into ecosystems
  - North America
    - Taiwan, Mexico, South America
  - Owned and operated by our members
    - started in 1978
  - “acid rain network”
  - Over 400,000 precipitation samples

National Atmospheric Deposition Program


CO98 and CO89,
Rocky Mountain National Park
3,231 meters altitude
ETI Noah-IV raingages

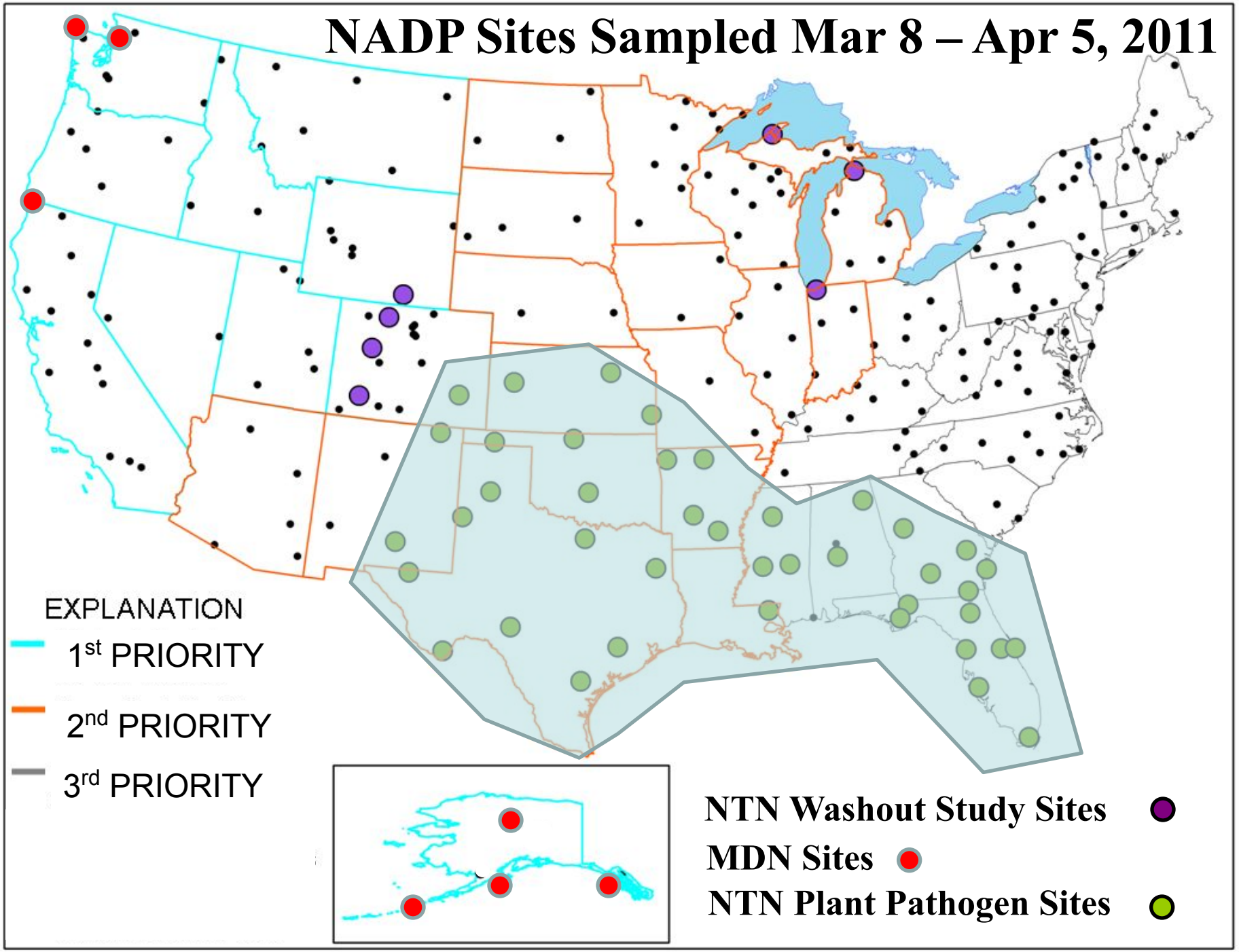
AIRMoN PA15
Penn State Univ., PA

MDN FL11
Everglades N.P., FL


Loch Vale NADP Site: April 19, 2011
After
Collectors
Gages


AK00: Dutch Harbor, Aleutian Islands
Modified ACM
MDN Collector
ETI Noah-IV
Precipitation
Gage

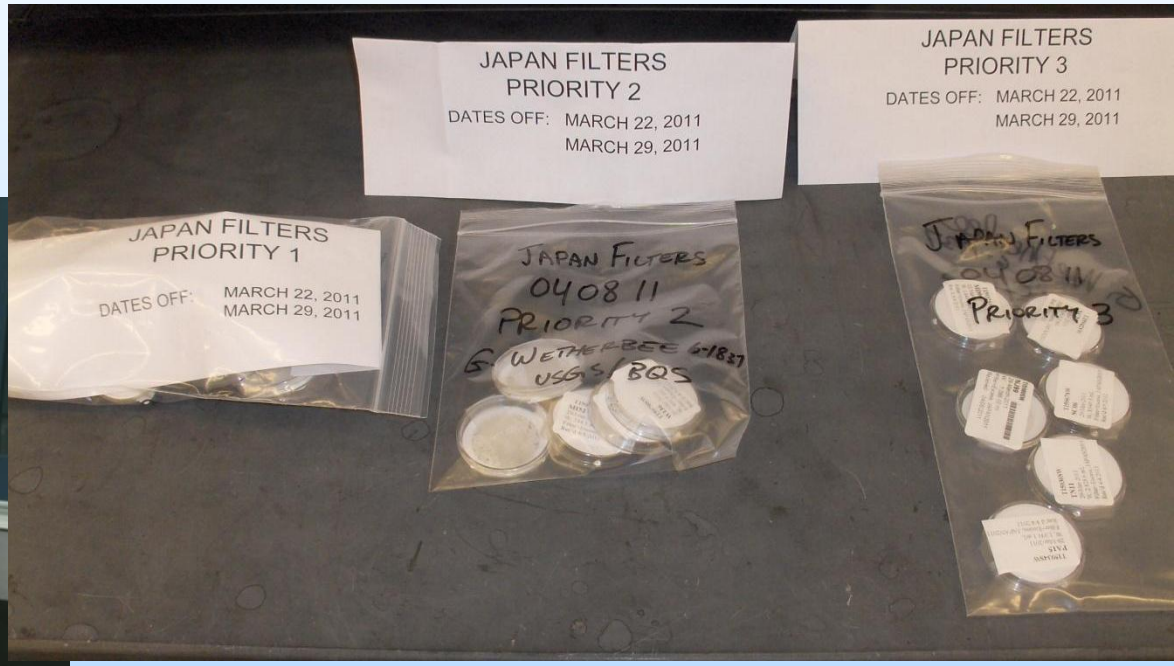
NADP Sites Sampled Mar 8 – Apr 5, 2011



Sample Processing & Filtration

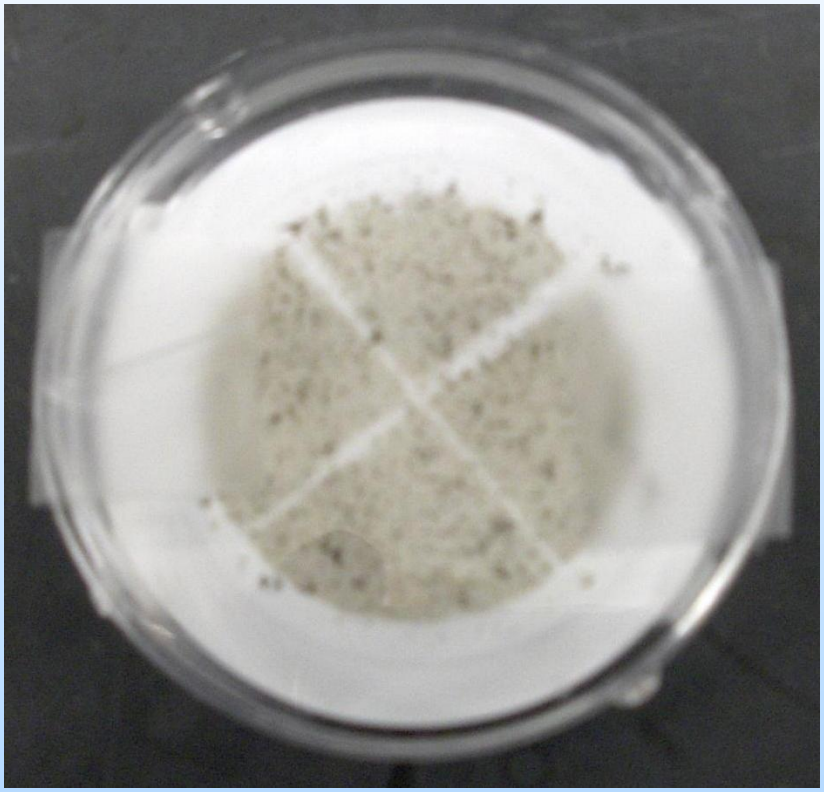
- Polyethylene buckets and bottles
- Polyethersulfone filters

Filters Sorted by Priority Regions



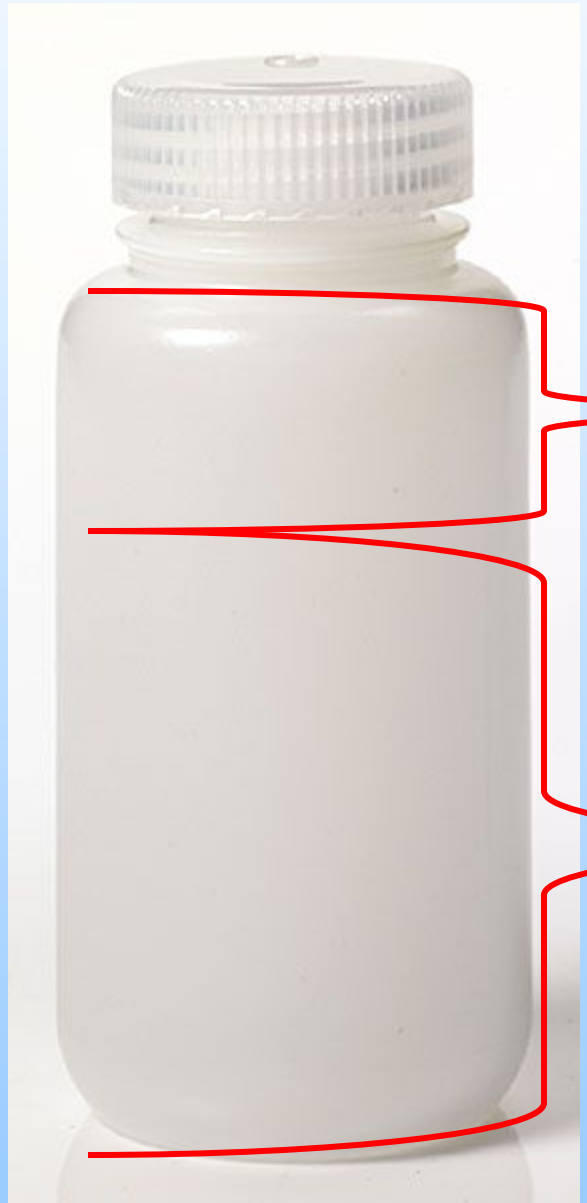
Phase 1: 47 mm Diameter Filters

Example of NTN Filter Sample



RESULTS: No Fission Products Detected on 280 Filters

Phase 2: Whole-water precipitation samples



First 250 ml, for normal NTN operations

Available for gamma spectrometry

(N = 160 sites)


WET DEPOSITION SAMPLES
NOT FILTERED
ACIDIFIED, pH < 2.0

Sampling and Analysis

1. Analyzed 280 NTN filters.

2. Weighed, acidified, composited and analyzed 176 precipitation samples.

3. QA/QC – 8 Blanks and 4 replicates.

4. Gamma Spectrometry by
   USGS National Reactor Facility, Denver, CO

Quality Assurance

1. 4 Blank Filters

   No fission products

2. 4 Blank Water Samples

   No fission products

3. Prior-Incident Samples (Mar 8-15, analyzed for ^{137}Cs)

   No fission products

4. Replicate Samples

   MA01 / 01MA @ Cape Cod NSS – both No fission products

   CA50 / 50CA @ Sage Hen Cr. FS – both No fission products

   CO98 / CO89 @ Rocky Mt. NP – both WITH fission products!

Gamma Spectroscopy

1. 2 Detectors
   16% & 40% efficient

2. Det. Efficiency Check w/^{152}Eu source

3. Range: 122 keV – 1.528 MeV

4. Calibration: +/- 1 keV

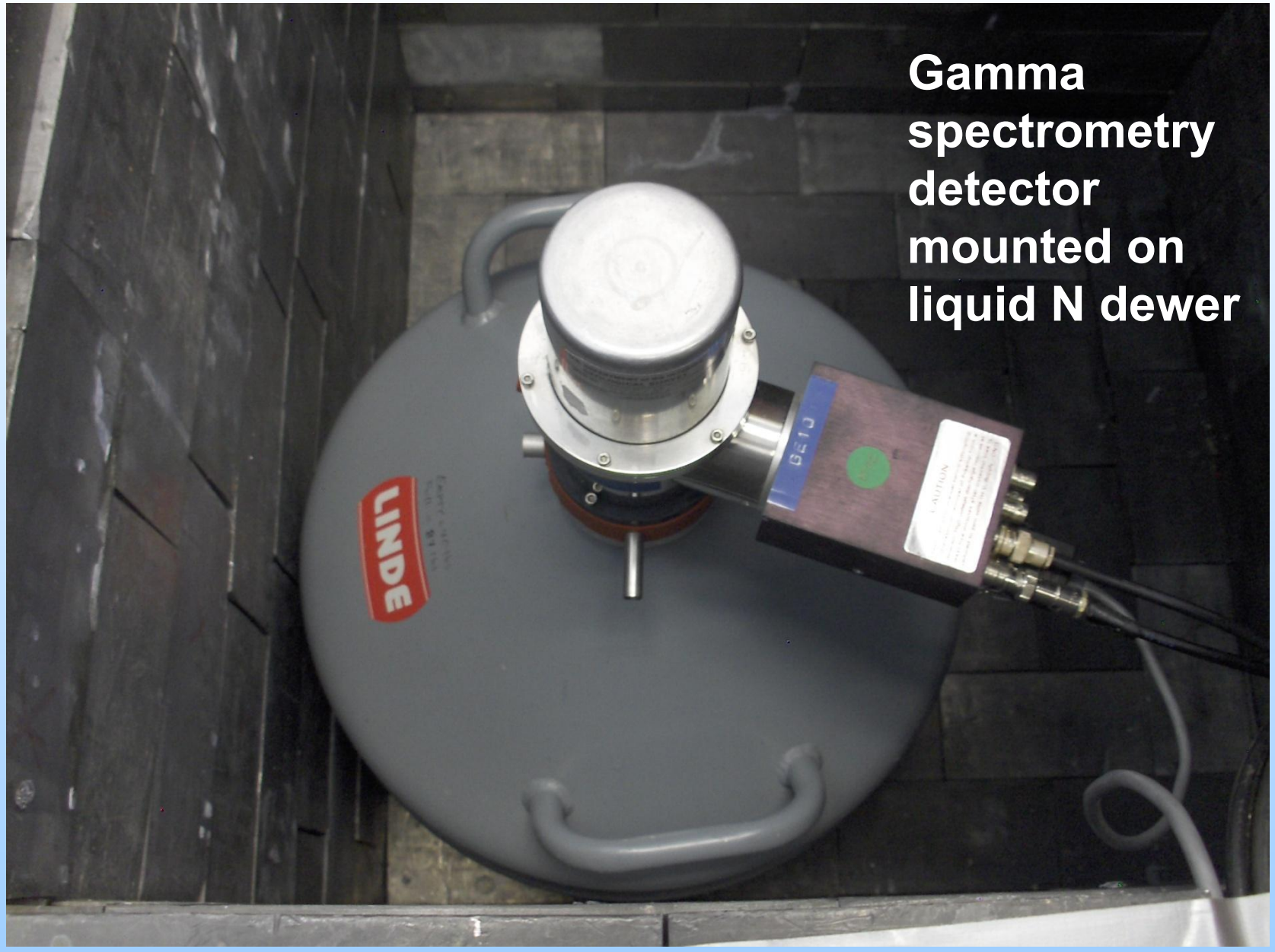
5. Filter Calibration : 0.5 µC ^{60}Co, 0.055 µC ^{137}Cs

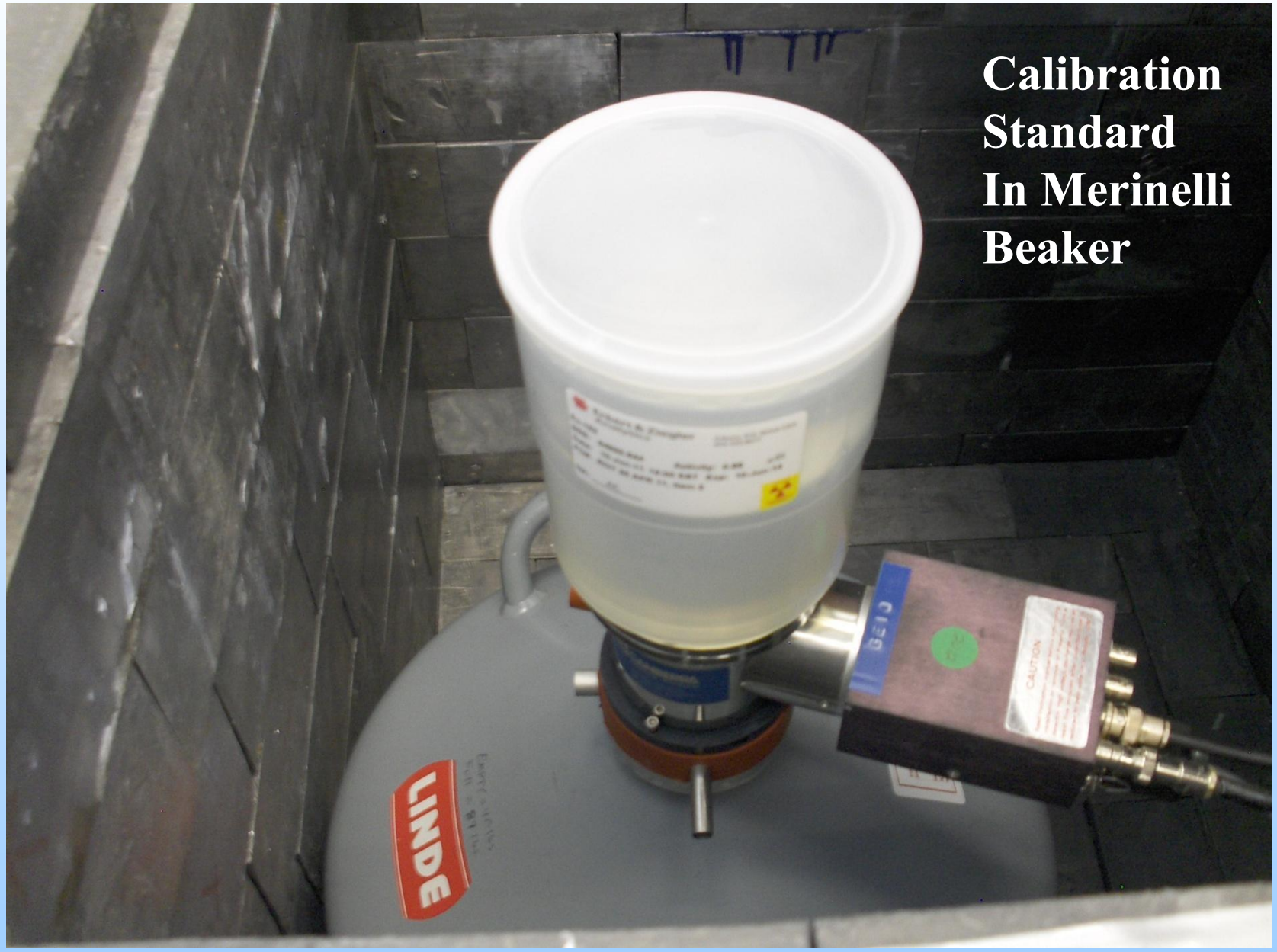
6. Water Calibration : 1 µC ^{152}Eu in 500 mL & 1,000 mL
   Merinelli Beaker Geometry

7. Count Times: 6 hr standard, up to 24 hours (reruns)

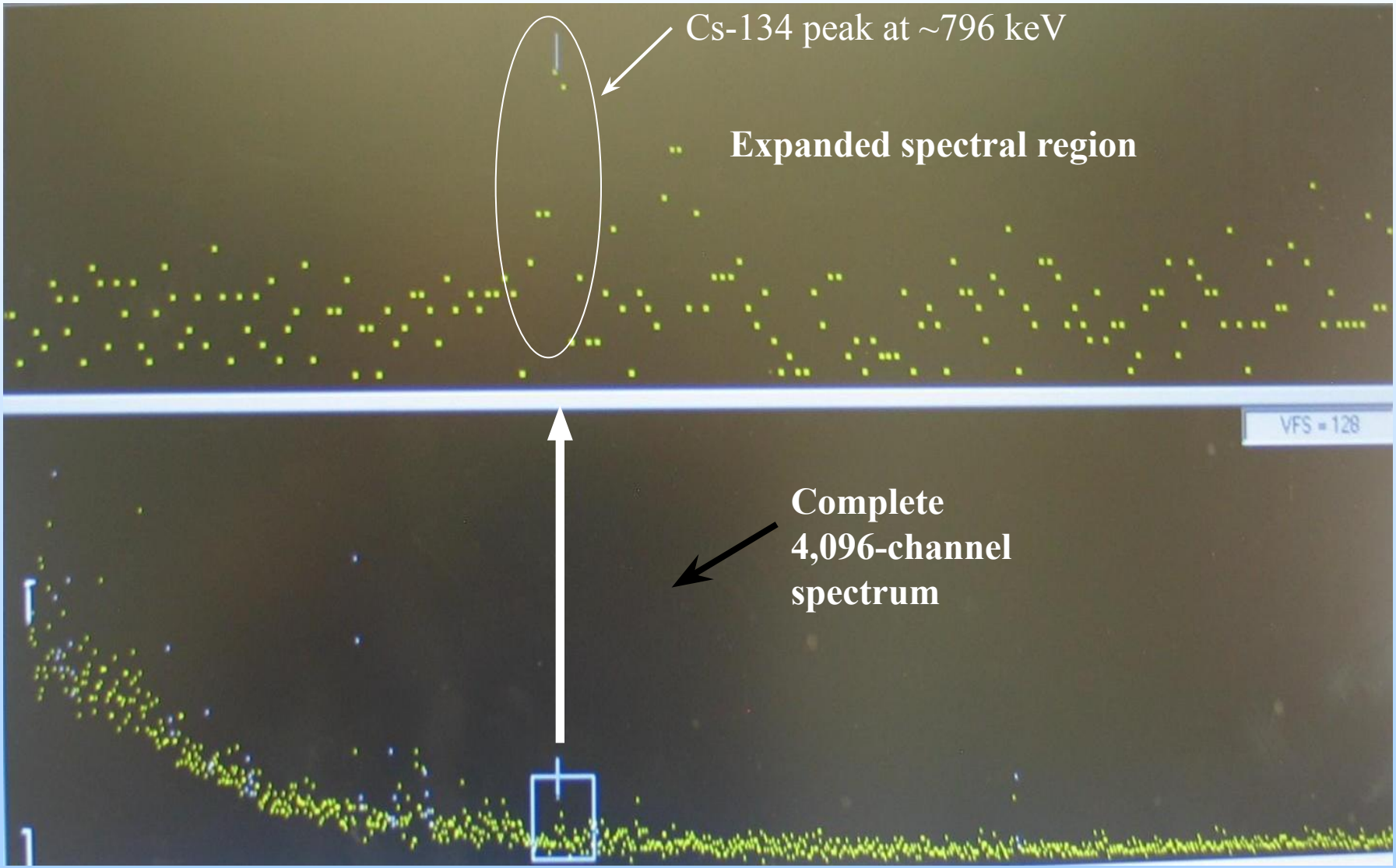
8. ^{131}I Activates, adjusted back to final precipitation during week

9. ^{134}Cs Activates manually estimated.


Gamma spectrometry detector mounted on liquid N dewer
LINDE


Calibration
Standard
In Merinelli
Beaker
LINDE


Sample
Pb
Shield
ROGEN NITROGEN



Counts

Gamma Ray Emission Energy (keV)

Activities vs. Deposition (Flux)

Gamma Spectrometry values in activity units
= picocuries per liter (pCi/L)

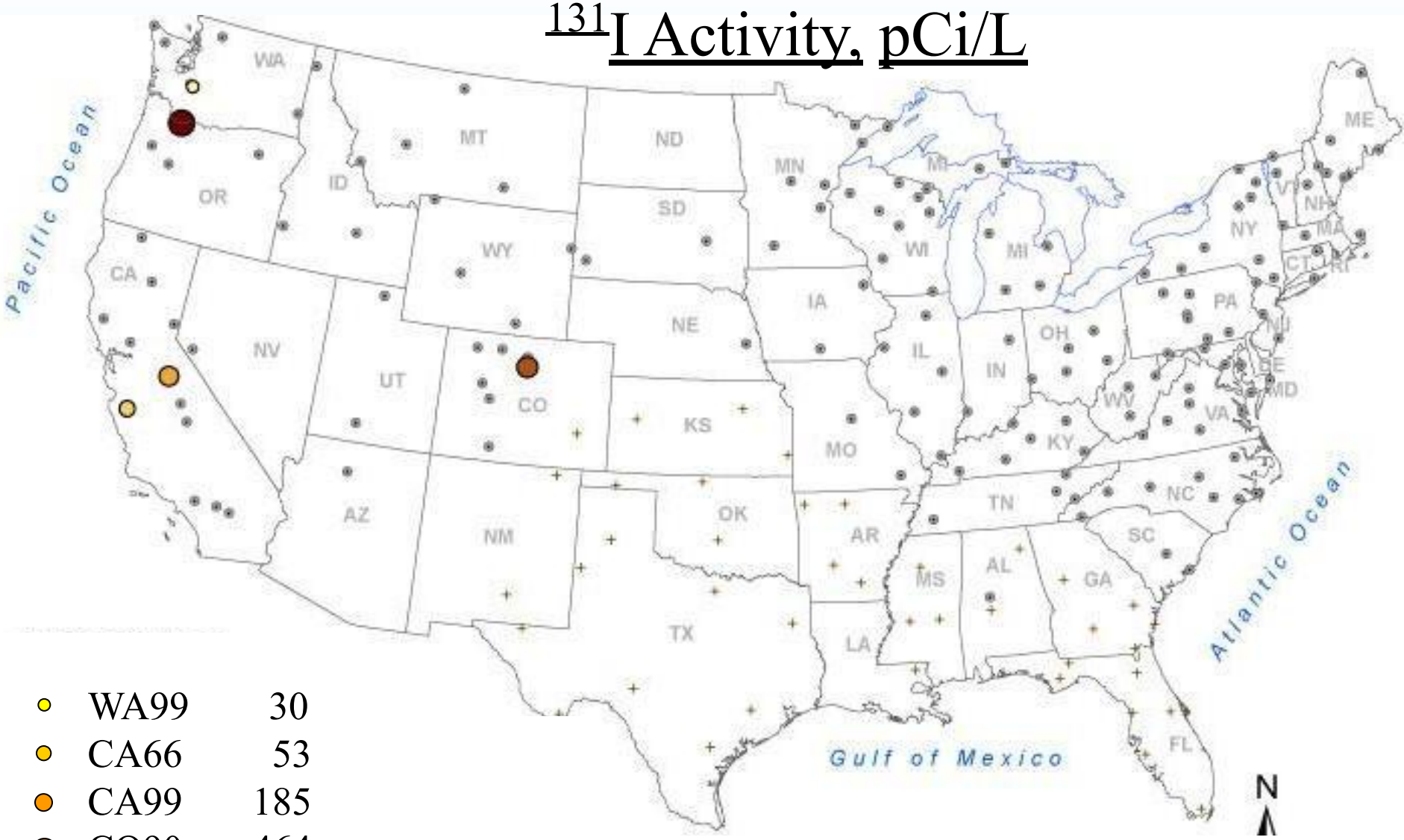
Deposition calculated using raingage depths in
= Becquerels per square meter (Bq/m^2)

Conversion Factors: 0.037 Bq / pCi
1 Liter = 1 mm depth / m^2

Deposition, (Bq/m^2)
= Activity (pCi/L) x Precip Depth (mm) x 0.037

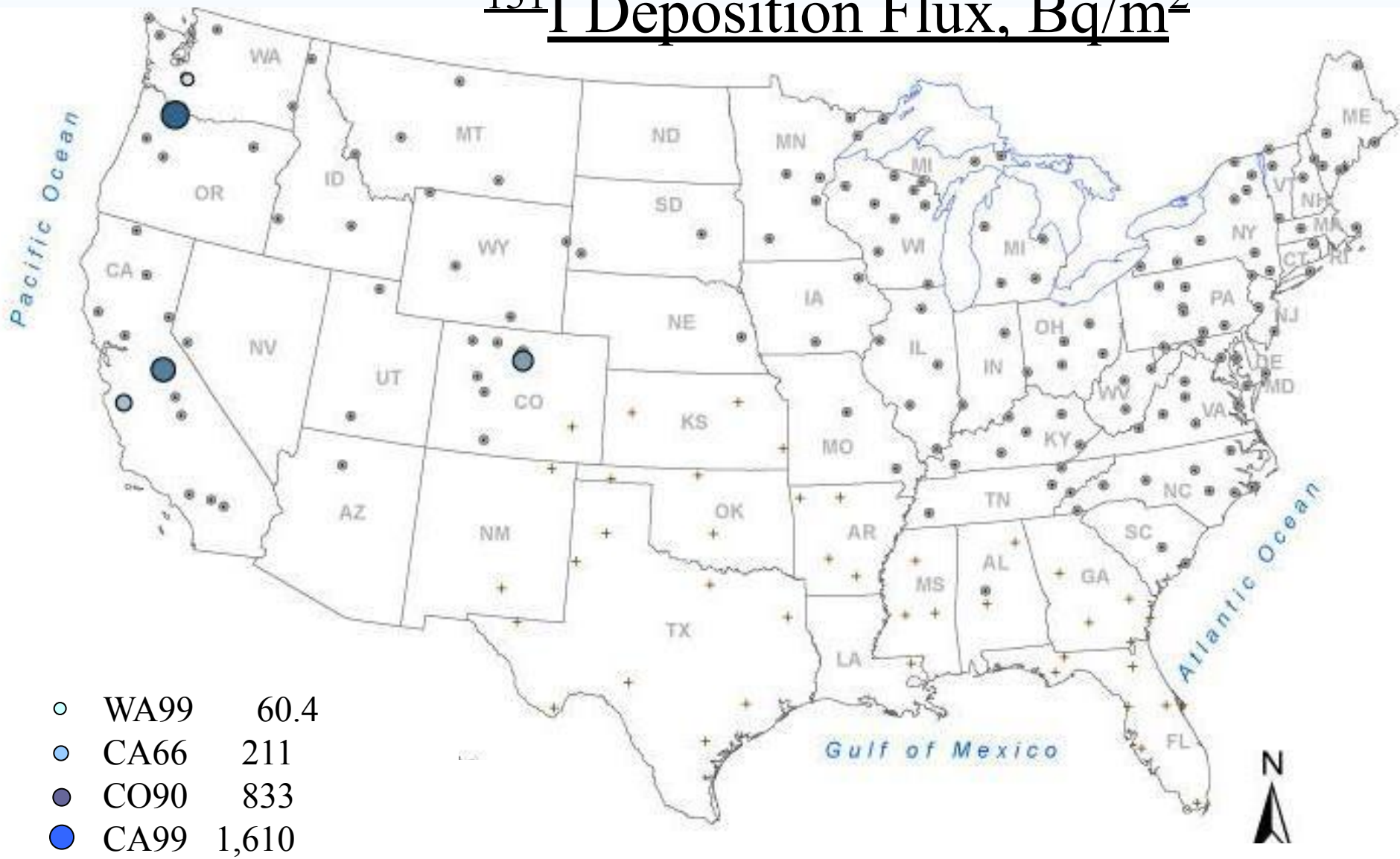
Results

^{131}I Activity, pCi/L



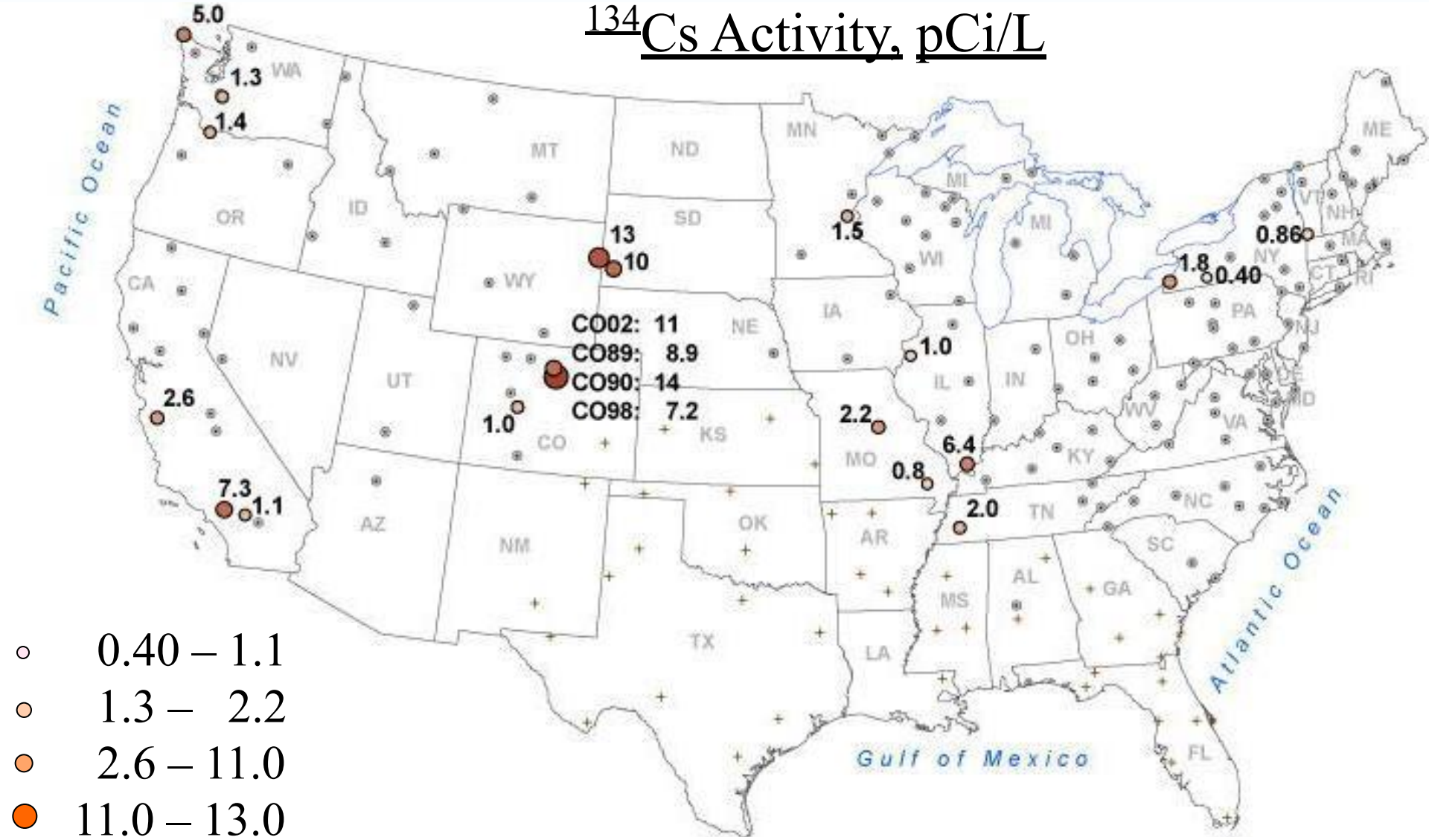
WA99	30	
CA66	53	
CA99	185	
CO90	464	
WA98	1,090	
+	No Sample	Not detected

^{131}I Deposition Flux, Bq/m^2



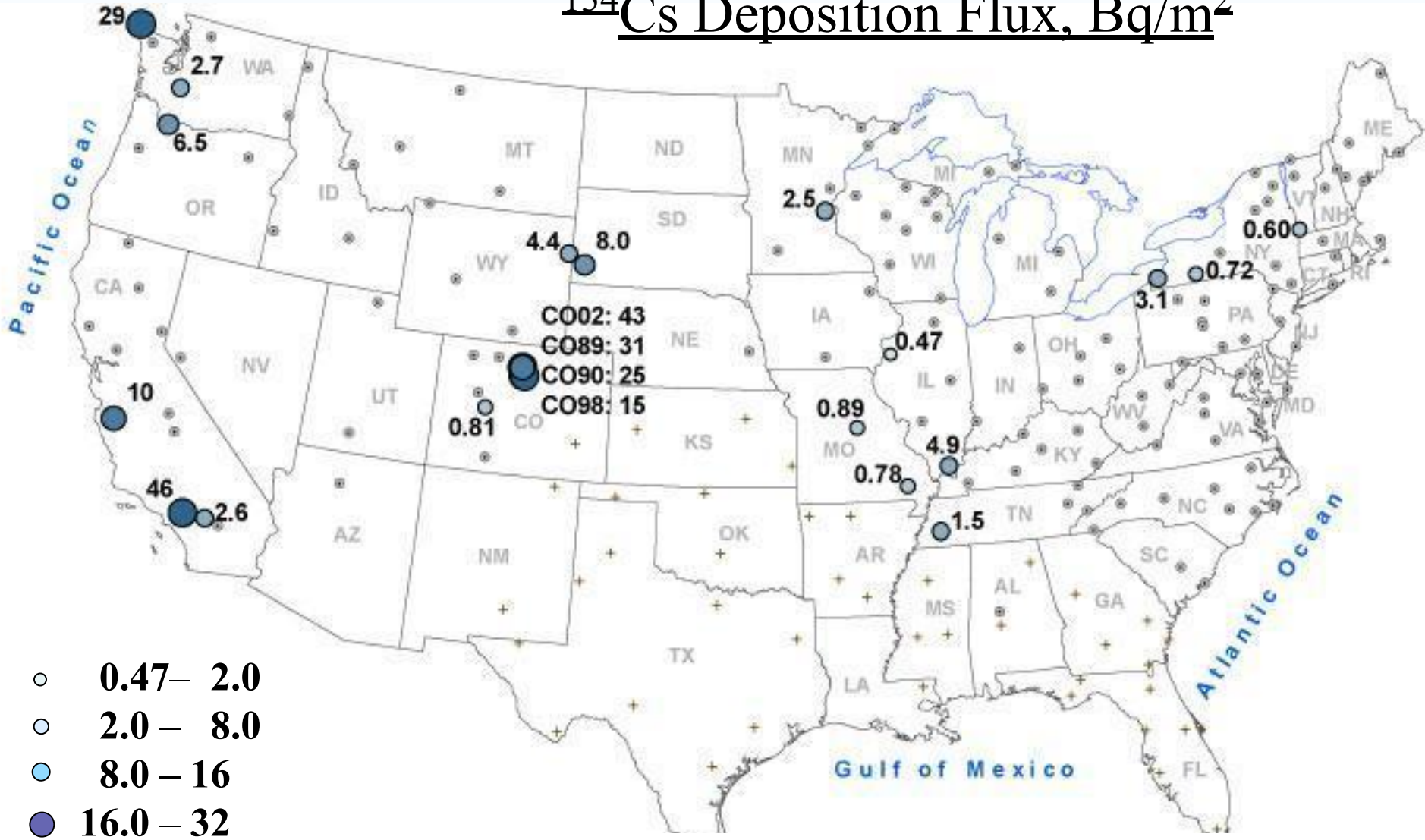
- WA99 60.4
- CA66 211
- CO90 833
- CA99 1,610
- WA98 5,100
- \+ No sample
- Not detected

^{134}Cs Activity, pCi/L



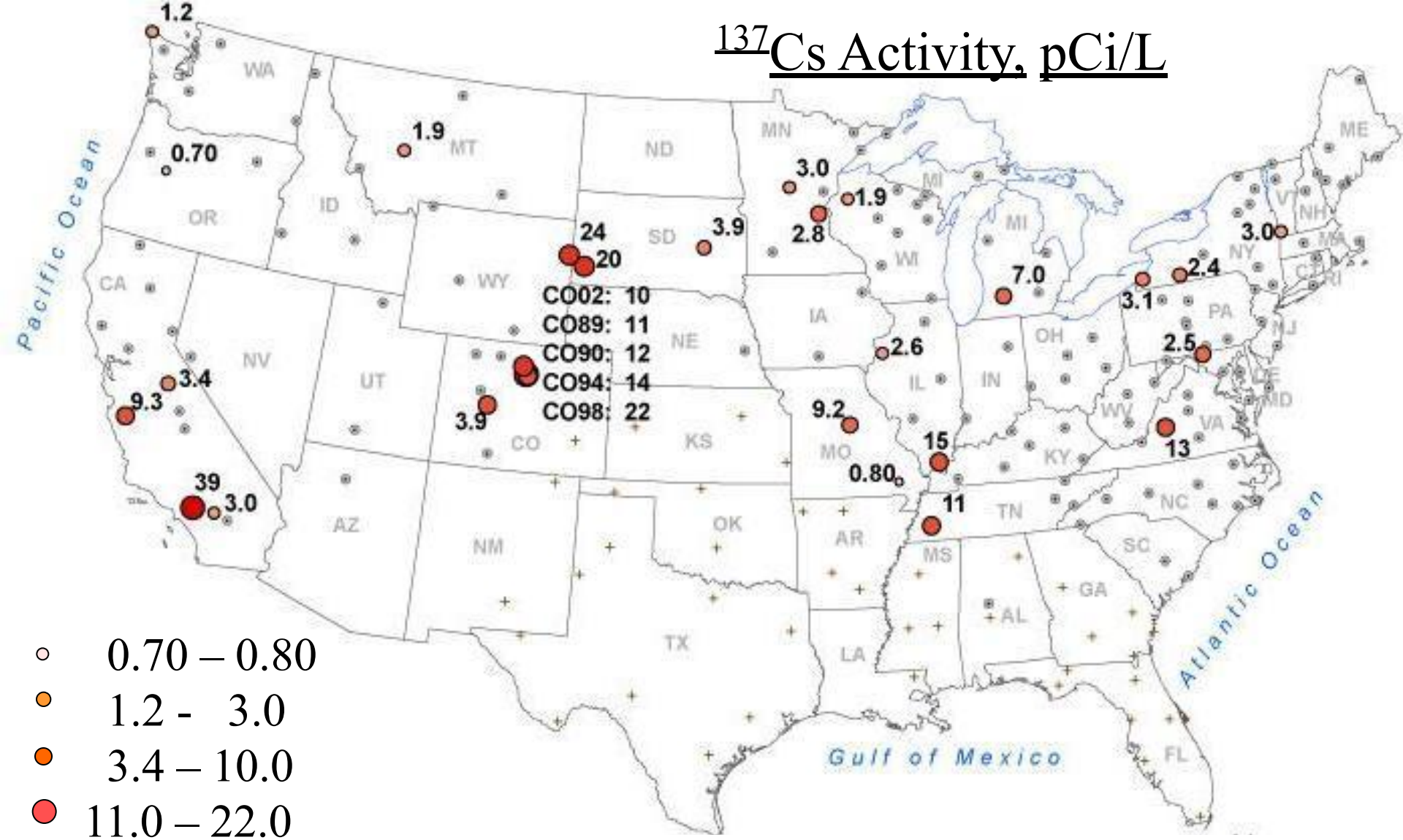
- ○ 0.40 – 1.1
- ○ 1.3 – 2.2
- ○ 2.6 – 11.0
- ● 11.0 – 13.0
- ● 14.0
- \+ No sample
- ○ Not detected

^{134}Cs Deposition Flux, Bq/m^2



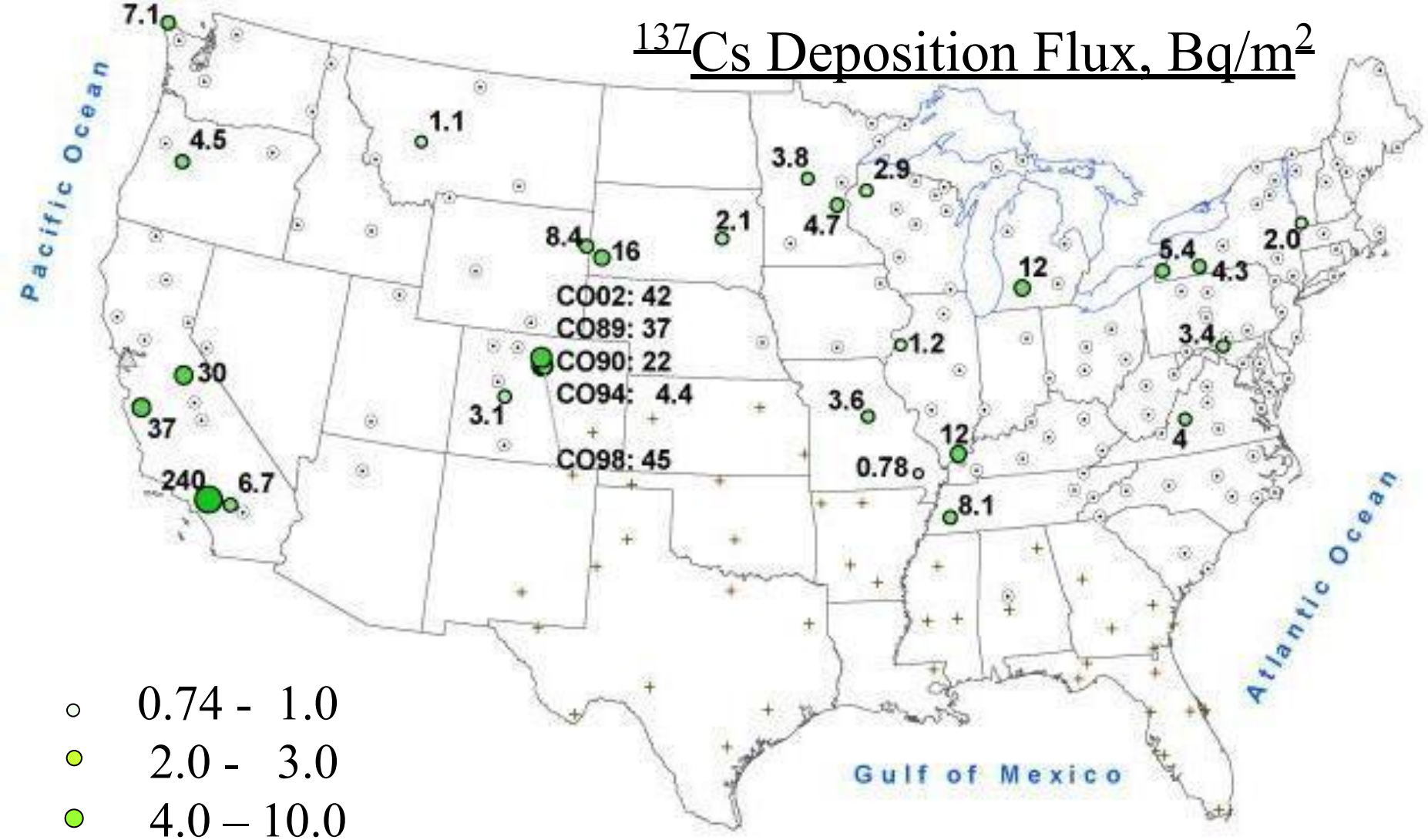
- ○ 0.47– 2.0
- ○ 2.0 – 8.0
- ○ 8.0 – 16
- ● 16.0 – 32
- ● 32 – 46
- \+ Plant Pathogen Study
- ○ Not detected

^{137}Cs Activity, pCi/L



- ◦ 0.70 – 0.80
- ● 1.2 - 3.0
- ● 3.4 – 10.0
- ● 11.0 – 22.0
- ● 24.0 – 39.0
- \+ No Sample
- ◦ Not detected

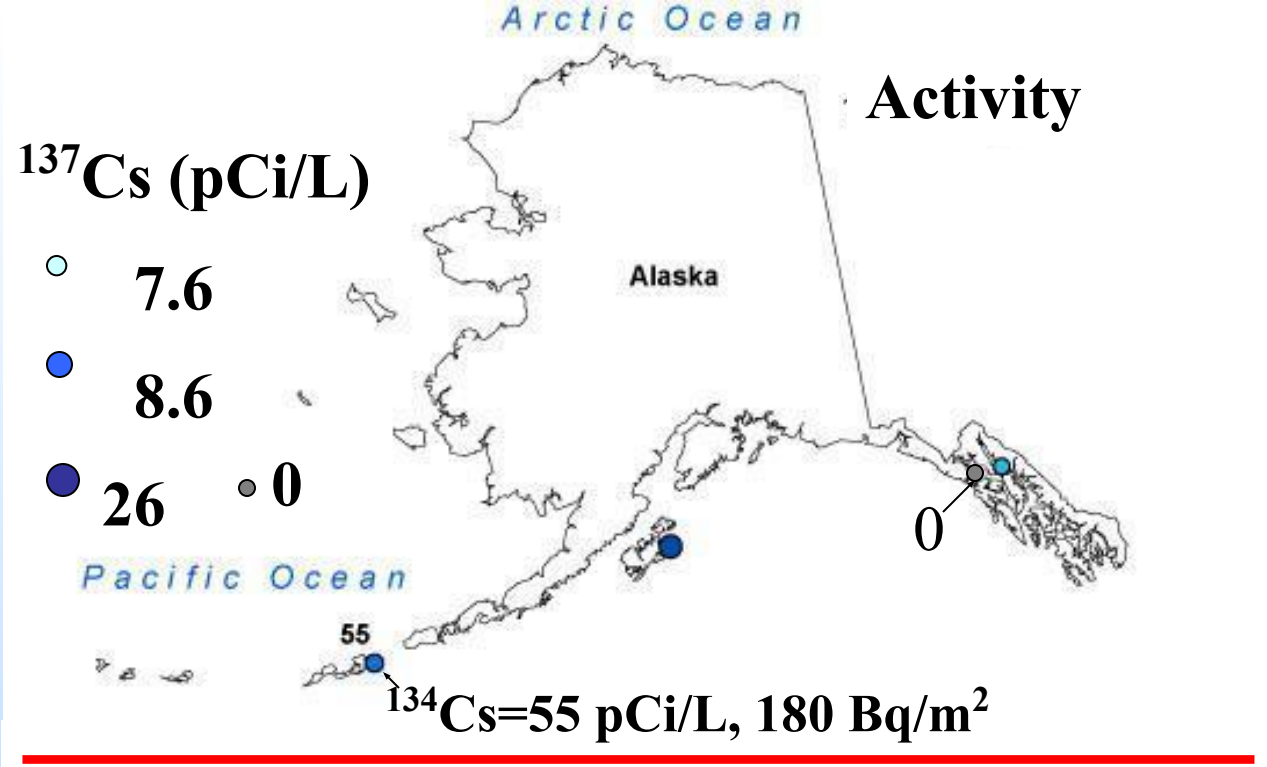
^{137}Cs Deposition Flux, Bq/m^2

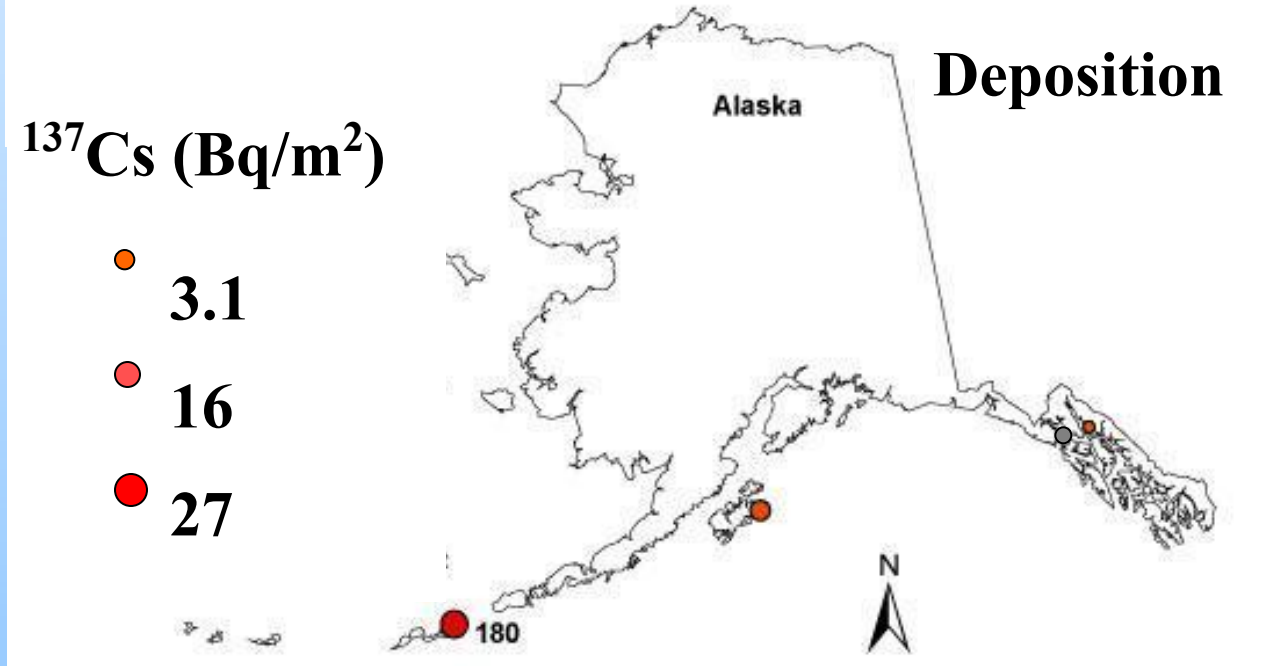


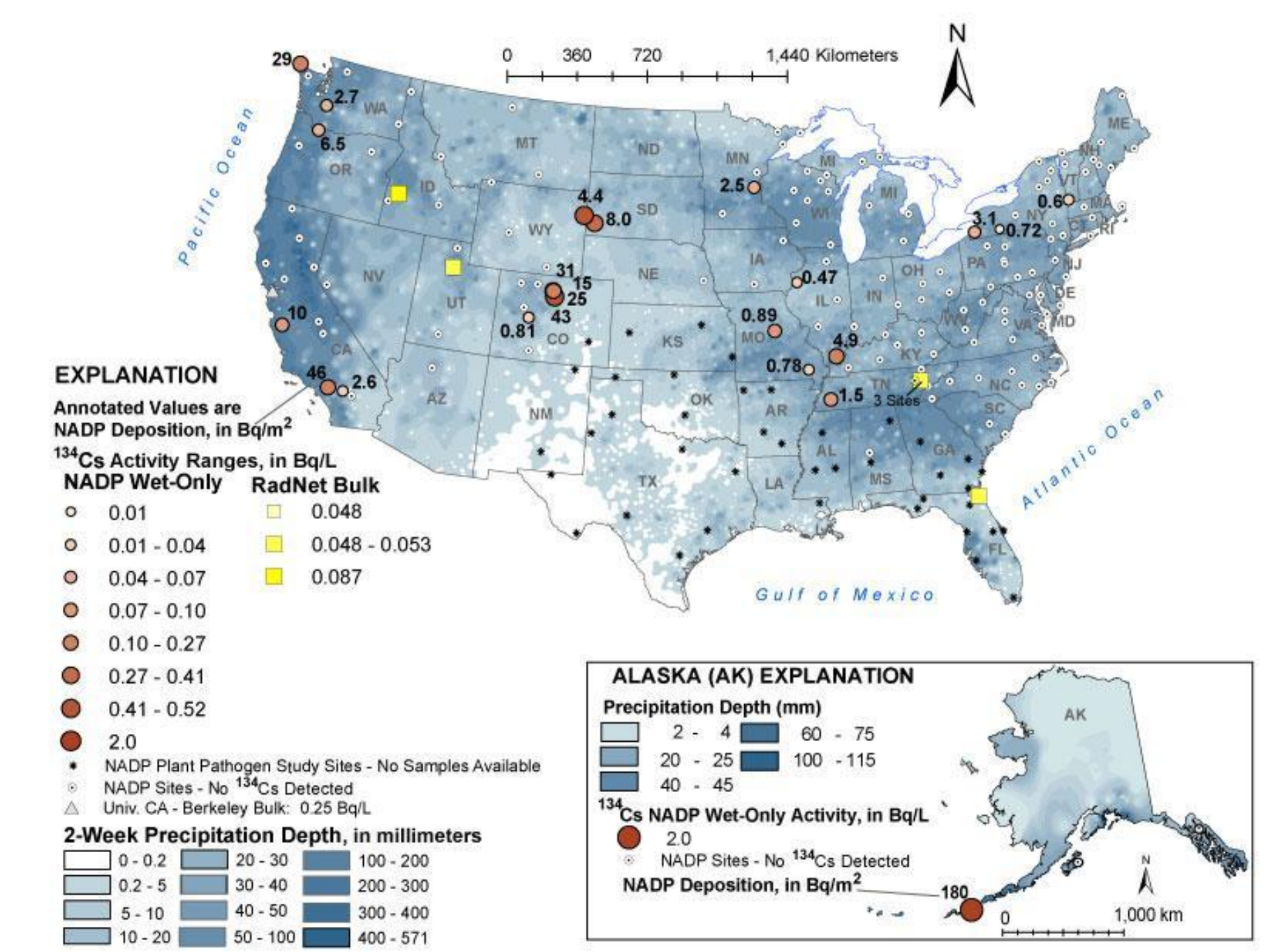
- ○ 0.74 - 1.0
- ○ 2.0 - 3.0
- ○ 4.0 – 10.0
- ● 20.0 – 40.0
- ● 74.0 - 240
- \+ Plant Pathogen Study
- ○ Not detected

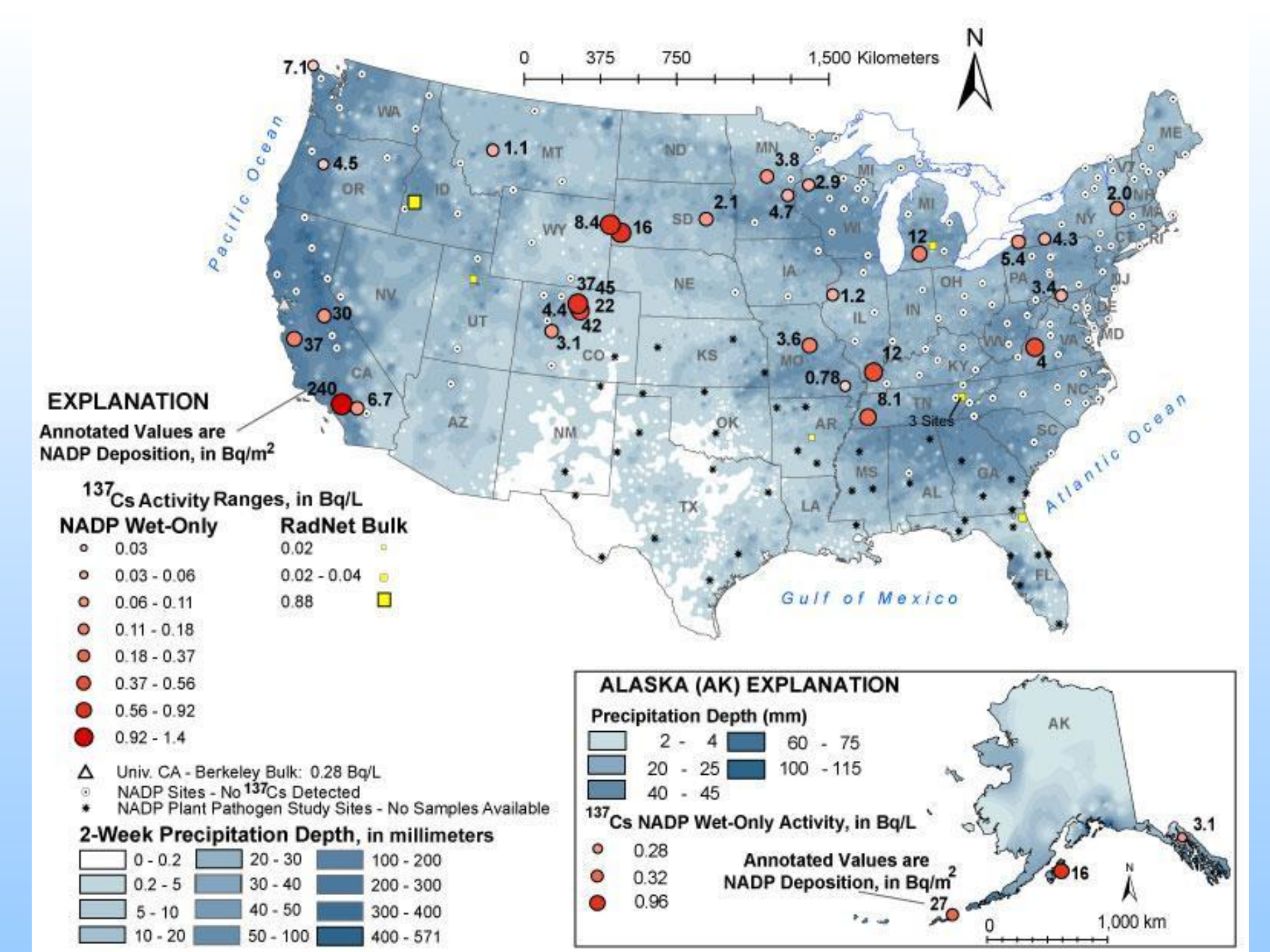
NADP IN ALASKA

^{134}Cs and ^{137}Cs






0 360 720 1,440 Kilometers
N
Pacific Ocean
Atlantic Ocean
Gulf of Mexico
WA
OR
CA
ID
NV
MT
WY
UT
AZ
CO
NM
ND
SD
NE
KS
OK
TX
MN
IA
MO
AR
LA
WI
IL
MS
MI
IN
KY
TN
AL
OH
WV
GA
FL
SC
NC
VA
PA
NY
MD
DE
NJ
CT
RI
MA
VT
NH
ME
29
2.7
6.5
10
46
2.6
4.4
8.0
31
15
25
43
0.81
2.5
0.47
0.89
4.9
0.78
1.5
3 Sites
3.1
0.72
0.6
EXPLANATION
Annotated Values are NADP Deposition, in Bq/m2
134Cs Activity Ranges, in Bq/L
NADP Wet-Only
0.01
0.01 - 0.04
0.04 - 0.07
0.07 - 0.10
0.10 - 0.27
0.27 - 0.41
0.41 - 0.52
2.0
RadNet Bulk
0.048
0.048 - 0.053
0.087
NADP Plant Pathogen Study Sites - No Samples Available
NADP Sites - No 134Cs Detected
Univ. CA - Berkeley Bulk: 0.25 Bq/L
2-Week Precipitation Depth, in millimeters
0 - 0.2
0.2 - 5
5 - 10
10 - 20
20 - 30
30 - 40
40 - 50
50 - 100
100 - 200
200 - 300
300 - 400
400 - 571
ALASKA (AK) EXPLANATION
Precipitation Depth (mm)
2 - 4
20 - 25
40 - 45
60 - 75
100 - 115
134Cs NADP Wet-Only Activity, in Bq/L
2.0
NADP Sites - No 134Cs Detected
NADP Deposition, in Bq/m2
180
AK
N
0
1,000 km


0 375 750 1,500 Kilometers
N
Pacific Ocean
Atlantic Ocean
Gulf of Mexico
WA OR CA ID NV MT WY UT AZ CO NM ND SD NE KS OK TX MN IA MO AR LA WI IL MS MI IN KY TN AL OH WV GA FL SC NC VA PA NY ME VT NH MA CT RI NJ DE MD
7.1
4.5
1.1
8.4
16
2.1
3.8
2.9
4.7
12
5.4
4.3
2.0
1.2
3.4
37
45
22
42
4.4
3.1
30
37
240
6.7
3.6
0.78
12
8.1
4
3 Sites
EXPLANATION
Annotated Values are
NADP Deposition, in Bq/m2
137Cs Activity Ranges, in Bq/L
NADP Wet-Only
0.03
0.03 - 0.06
0.06 - 0.11
0.11 - 0.18
0.18 - 0.37
0.37 - 0.56
0.56 - 0.92
0.92 - 1.4
RadNet Bulk
0.02
0.02 - 0.04
0.88
Univ. CA - Berkeley Bulk: 0.28 Bq/L
NADP Sites - No 137Cs Detected
NADP Plant Pathogen Study Sites - No Samples Available
2-Week Precipitation Depth, in millimeters
0 - 0.2
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ALASKA (AK) EXPLANATION
Precipitation Depth (mm)
2 - 4
20 - 25
40 - 45
60 - 75
100 - 115
137Cs NADP Wet-Only Activity, in Bq/L
0.28
0.32
0.96
Annotated Values are
NADP Deposition, in Bq/m2
27
16
3.1
AK
N
0
1,000 km

...s Emitted

Fig. 2. Spee
wind avera
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2007). Fuk
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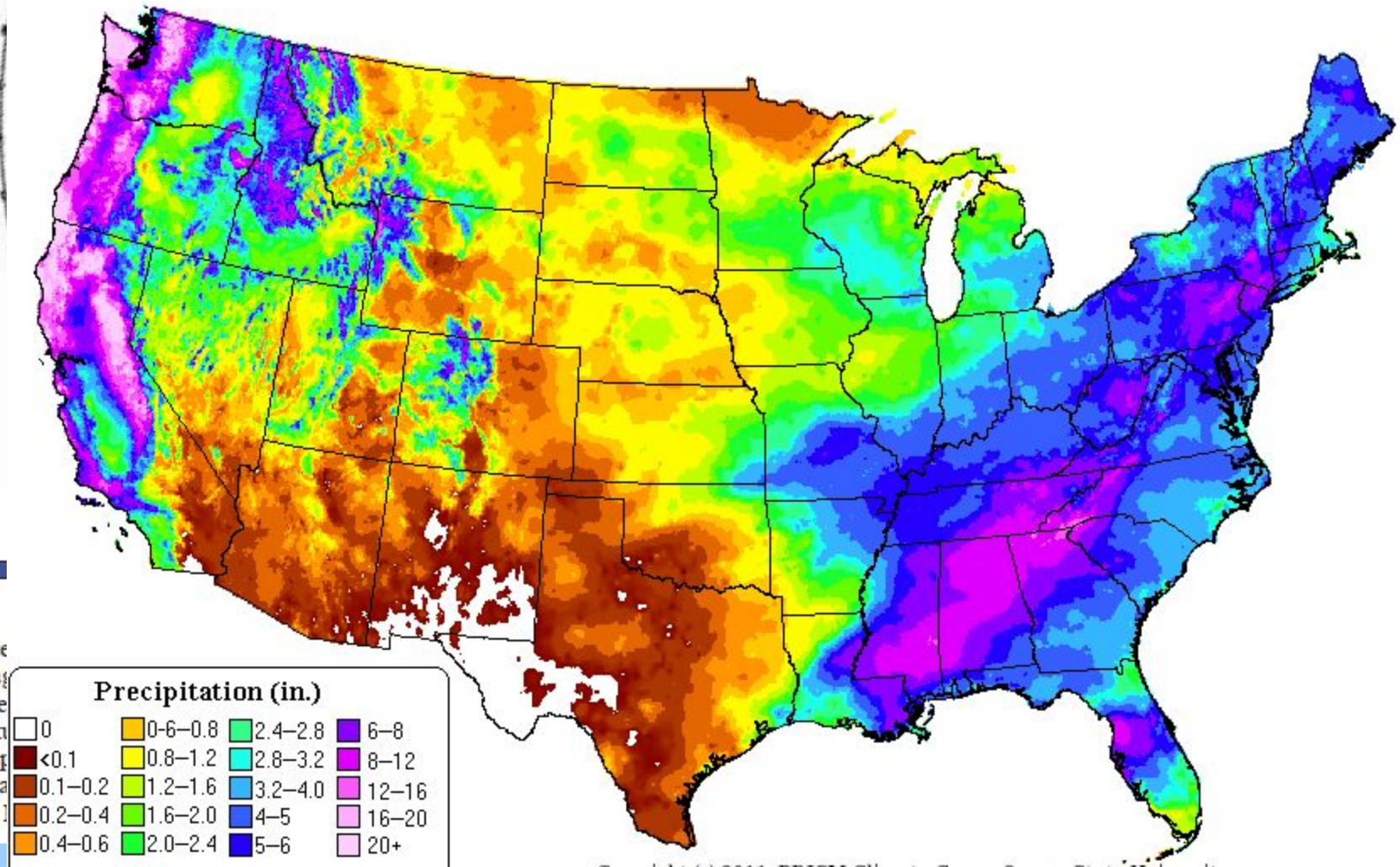
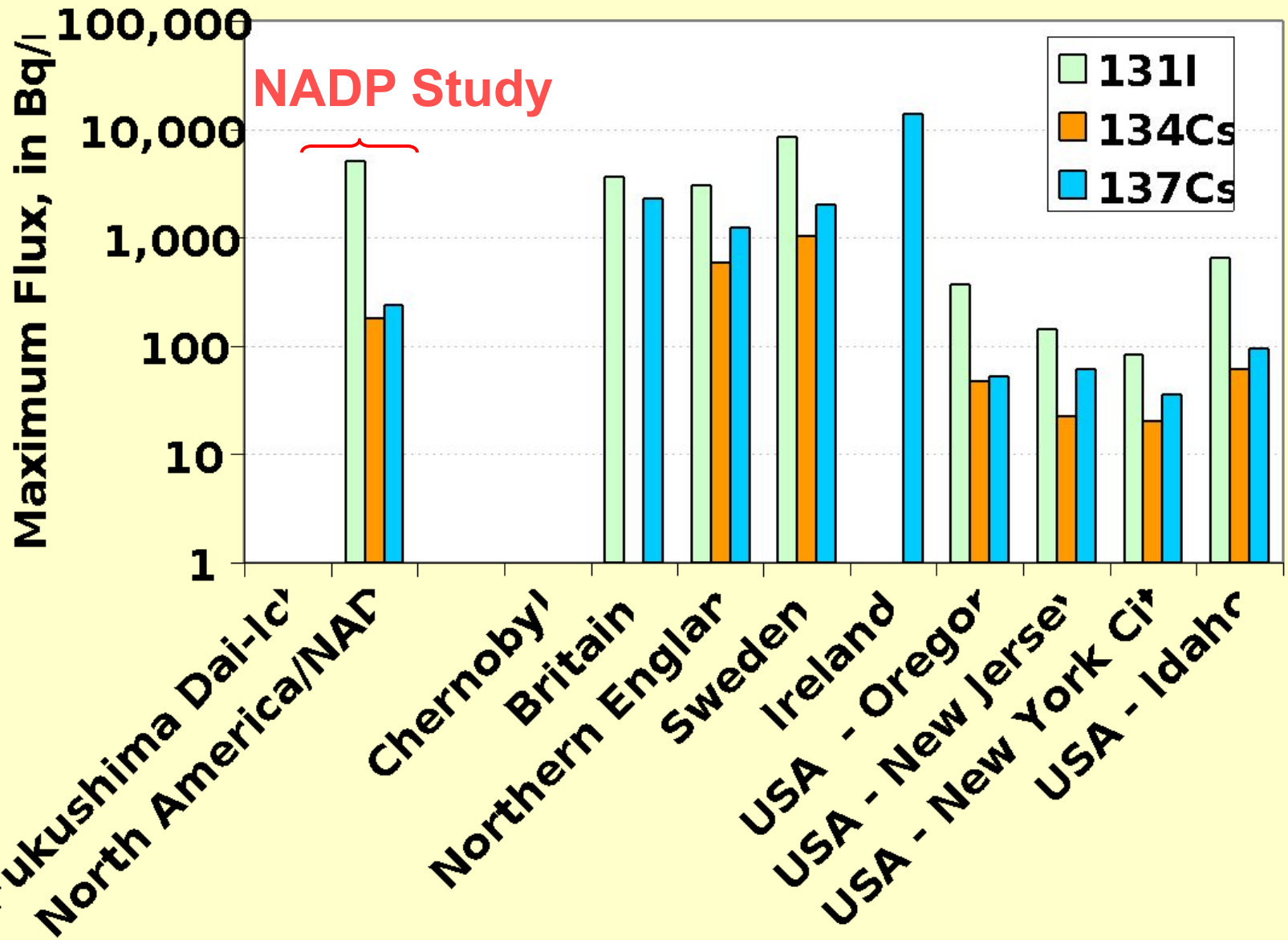
stronger tha
1200UTC,



Fukushima vs Chernobyl



Wetherbee, G.A., Gay, D.A., Debey, T.M., Lehmann, C.M.B., and Nilles, M.A., 2012, Wet deposition of fission-product isotopes to North America from the Fukushima Dai-ichi Incident, March 2011, Table 1, Environmental Science and Technology, 46 (5), pp 2574–2582.

Results in Context

- Maximum ^{137}Cs deposition:

  ~ 3%-10% additional radioactivity to that present in a common m^2 of soil (5 cm deep).

- Maximum NADP-measured 137Cs (240 Bq/m^2)

  ~ 17% of the NYC and Birmingham deposition (1,400 Bq/m2) during atmospheric nuclear testing in 1963.

Summary & Conclusions

1. Detectable ^{131}I, ^{134}Cs, & ^{137}Cs
   20% of sampled locations.

2. Estimated Deposition (FLUX) Ranges:

   ^{131}I: 60. – 5,100 Bq/m^2 @ 5 sites
   ^{134}Cs: 0.47 – 46 Bq/m^2 @ 25 site
   ^{137}Cs: 0.74 – 240 Bq/m^2 @ 33 sites

3. Spatial distribution of deposition and source region consistent with back trajectory analysis.

Summary & Conclusions

4. Fission products associated with particles < 0.45 mm, *OR* dissolved species.

5. NADP demonstrated a national capability to monitor unexpected releases of radionuclides to the environment.

Lessons Learned

1. Run precipitation samples first
   - archive filters for possible later analysis.

2. If we did this routinely, we would:
   - faster sampling to capture short-lived isotopes (^{131}I)
   - Need a dedicated collector
   - Some acidification/mixing changes

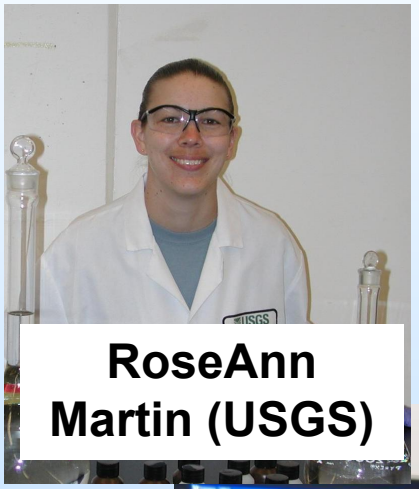
Future Work

1. Reanalysis of samples for ^{90}Sr.

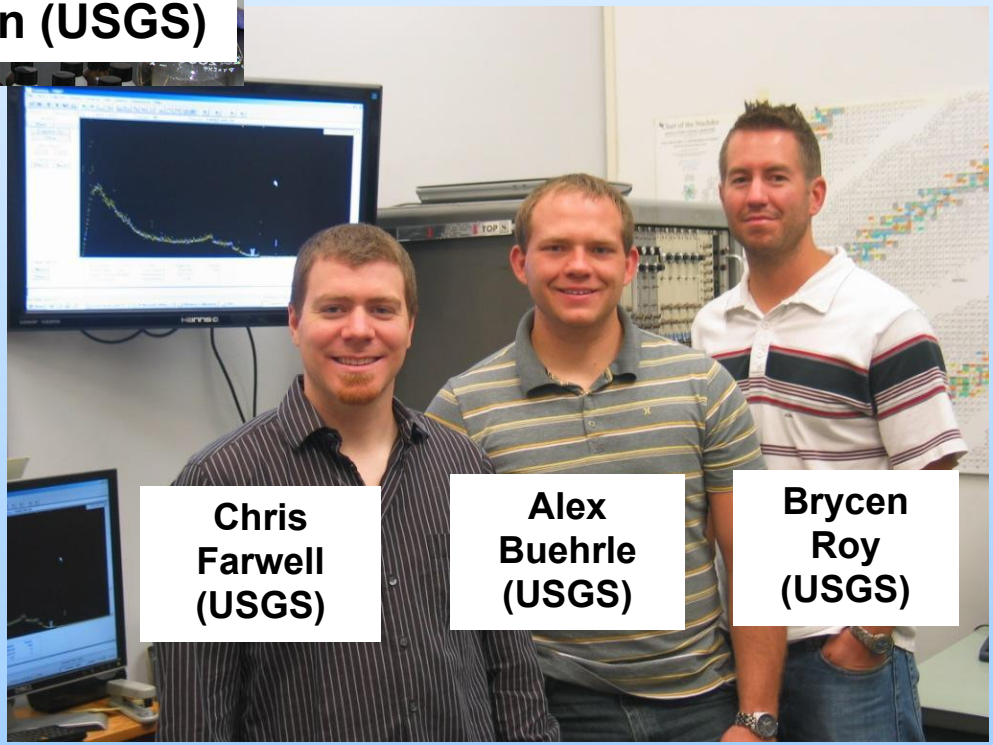
   We need a lab to do radiochemistry!

2. Proposed ^{3}H network for southeastern USA.

ACKNOWLEDGMENTS



RoseAnn Martin (USGS)

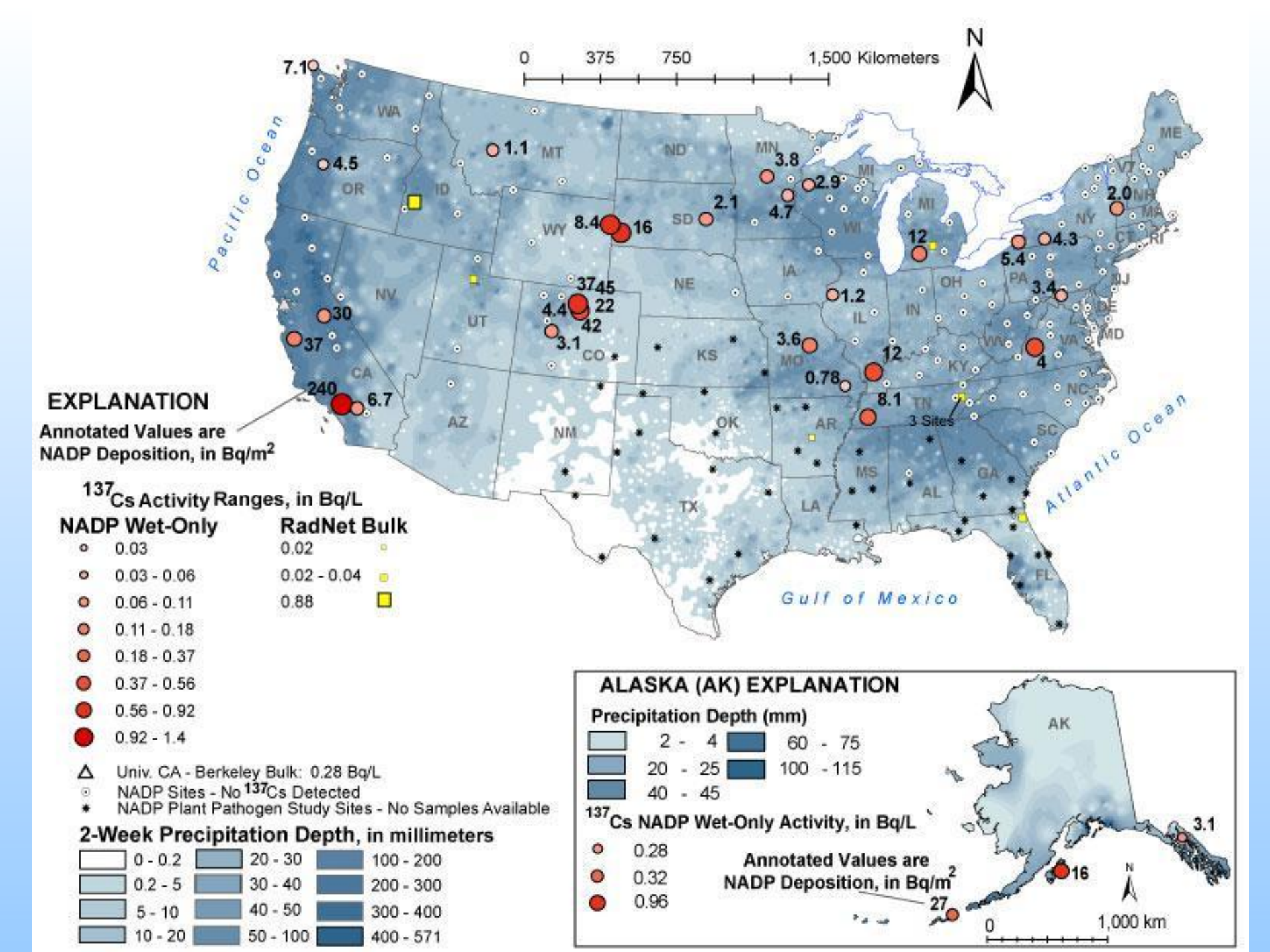


Chris Farwell (USGS)

Alex Buehrle (USGS)

Brycen Roy (USGS)

Brenda Riney (NADP)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3.1
AK
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0
1,000 km