DECADAL-SCALE CHANGES IN RADIUM CONCENTRATIONS IN GROUNDWATER AND RELATION TO ROAD SALT APPLICATION

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U.S. GEOLOGICAL SURVEY

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OVERVIEW

- National Groundwater Quality Monitoring: Rotational Surveillance network
- Highlights of national sodium, chloride, and radium results
- Relations between road-salt, sodium, chloride, and radium in New Jersey
- Changes in radium concentrations in other aquifers

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Article

Relation between Road-Salt Application and Increasing Radium Concentrations in a Low-pH Aquifer, Southern New Jersey

Bruce D. Lindsey,* Charles A. Cravotta III, Zoltan Szabo, Kenneth Belitz, and Paul Stackelberg

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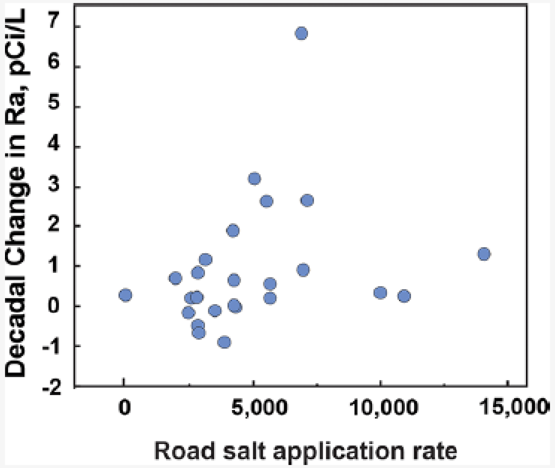
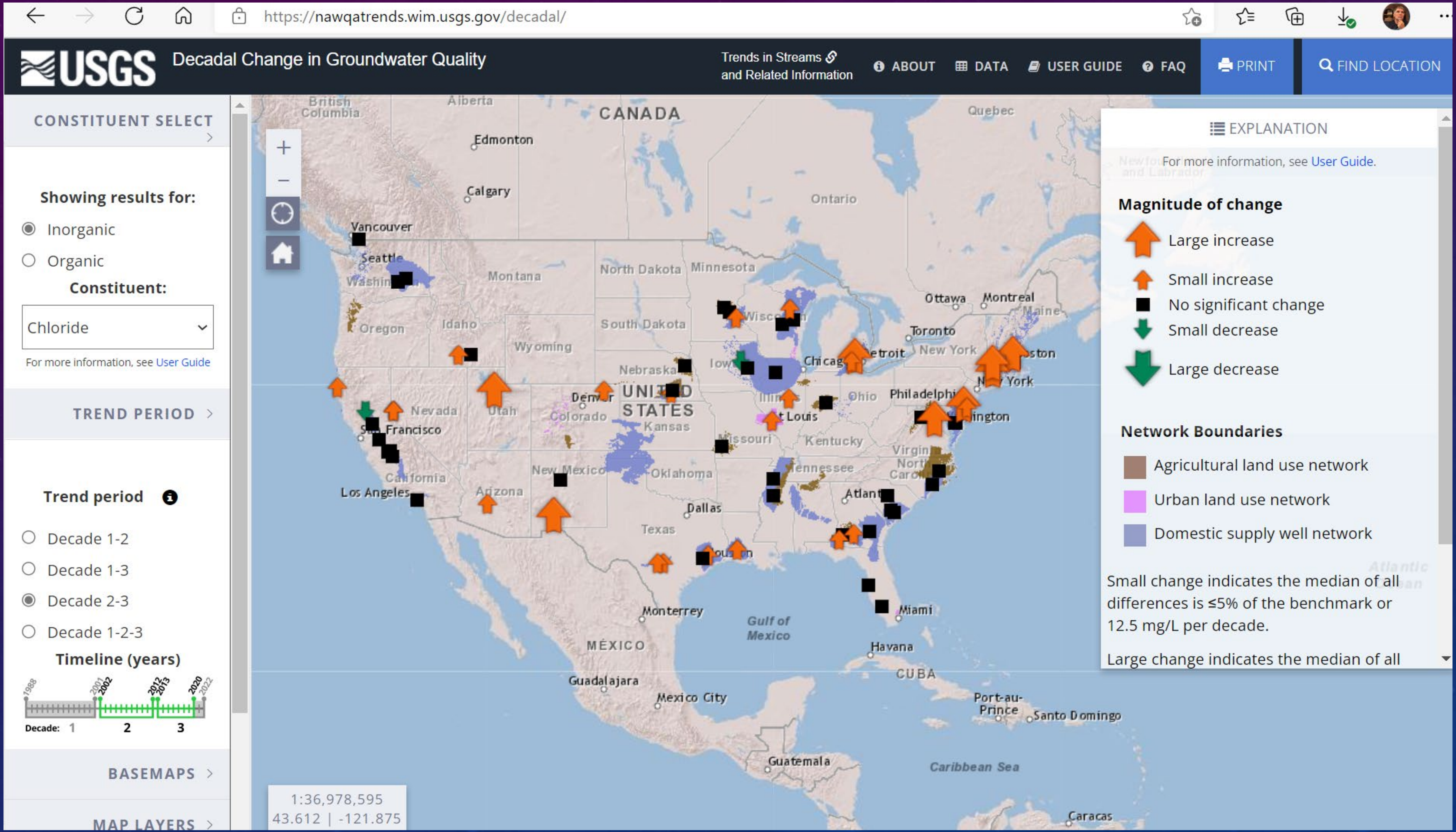
ABSTRACT: The Kirkwood–Cohansey aquifer in southern New Jersey is an important source of drinking-water supplies, but the availability of the resource is limited in some areas by high concentrations of radium, a potential carcinogen at elevated concentrations. Radium (^{226}Ra plus ^{228}Ra) concentrations from a network of 25 drinking-water wells showed a statistically significant increase over a decadal time scale (p < 0.05), with a median increase of 0.35 picocuries per liter. Increases in Ra are correlated with road-salt application rates, and we hypothesize that the correlation is causal. Geochemical processes associated with road-salt applications that can mobilize Ra into solution include competition by excess sodium for sorption sites and formation of chloride complexes ($RaCl^+$ and $RaCl_2$). The largest increases in Ra were in groundwater with low pH (≤5), which is an indirect surrogate for low cation-sorption capacity. Correlations with other potential anthropogenic causes for the increase in Ra were not observed, further suggesting a road-salt effect. Given the significant increase in Ra concentrations in this drinking-water source, the known carcinogenic risks from Ra, the direct link to road-salt application, and the likelihood for continued increases, additional monitoring is necessary in areas with similar hydrogeologic and geochemical settings.



KEYWORDS: *radium, road salt, deicing, groundwater trends, sodium, chloride, salinization*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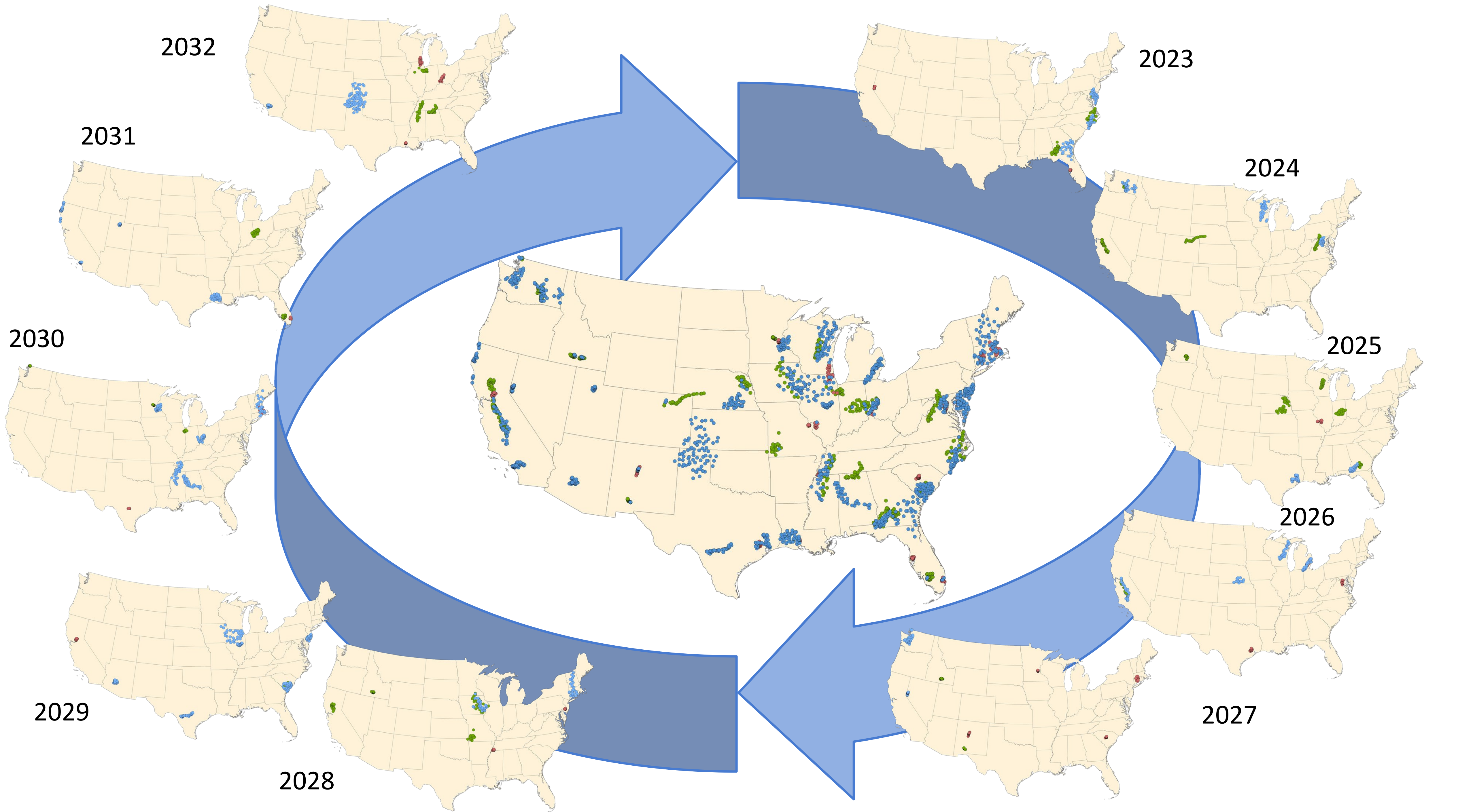
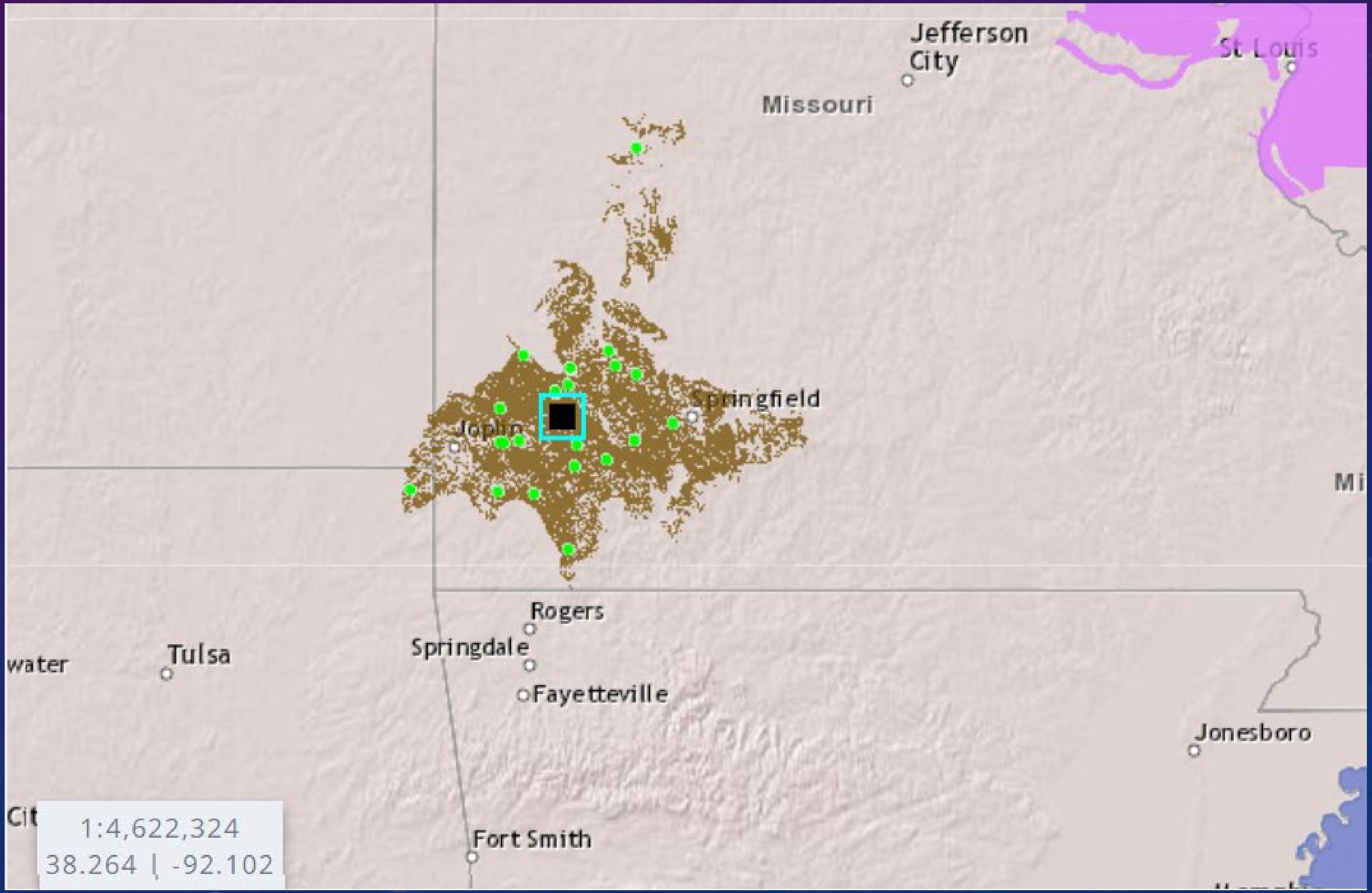
USGS
Decadal Change in Groundwater Quality
Trends in Streams and Related Information
ABOUT
DATA
USER GUIDE
FAQ
PRINT
FIND LOCATION
CONSTITUENT SELECT
Showing results for:
Inorganic
Organic
Constituent:
Chloride
For more information, see User Guide
TREND PERIOD
Trend period
Decade 1-2
Decade 1-3
Decade 2-3
Decade 1-2-3
Timeline (years)
1988
2001
2002
2012
2013
2020
2022
Decade: 1 2 3
BASEMAPS
MAP LAYERS
1:36,978,595
43.612 | -121.875
EXPLANATION
For more information, see User Guide.
Magnitude of change
Large increase
Small increase
No significant change
Small decrease
Large decrease
Network Boundaries
Agricultural land use network
Urban land use network
Domestic supply well network
Small change indicates the median of all differences is ≤5% of the benchmark or 12.5 mg/L per decade.
Large change indicates the median of all
CANADA
UNITED STATES
MEXICO
CUBA
Gulf of Mexico
Caribbean Sea
Vancouver
Seattle
Edmonton
Calgary
San Francisco
Los Angeles
Denver
Dallas
Houston
Chicago
Detroit
St Louis
Toronto
Ottawa
Montreal
New York
Philadelphia
Washington
Boston
Atlanta
Miami
Havana
Monterrey
Guadalajara
Mexico City
Guatemala
Port-au-Prince
Santo Domingo
Caracas

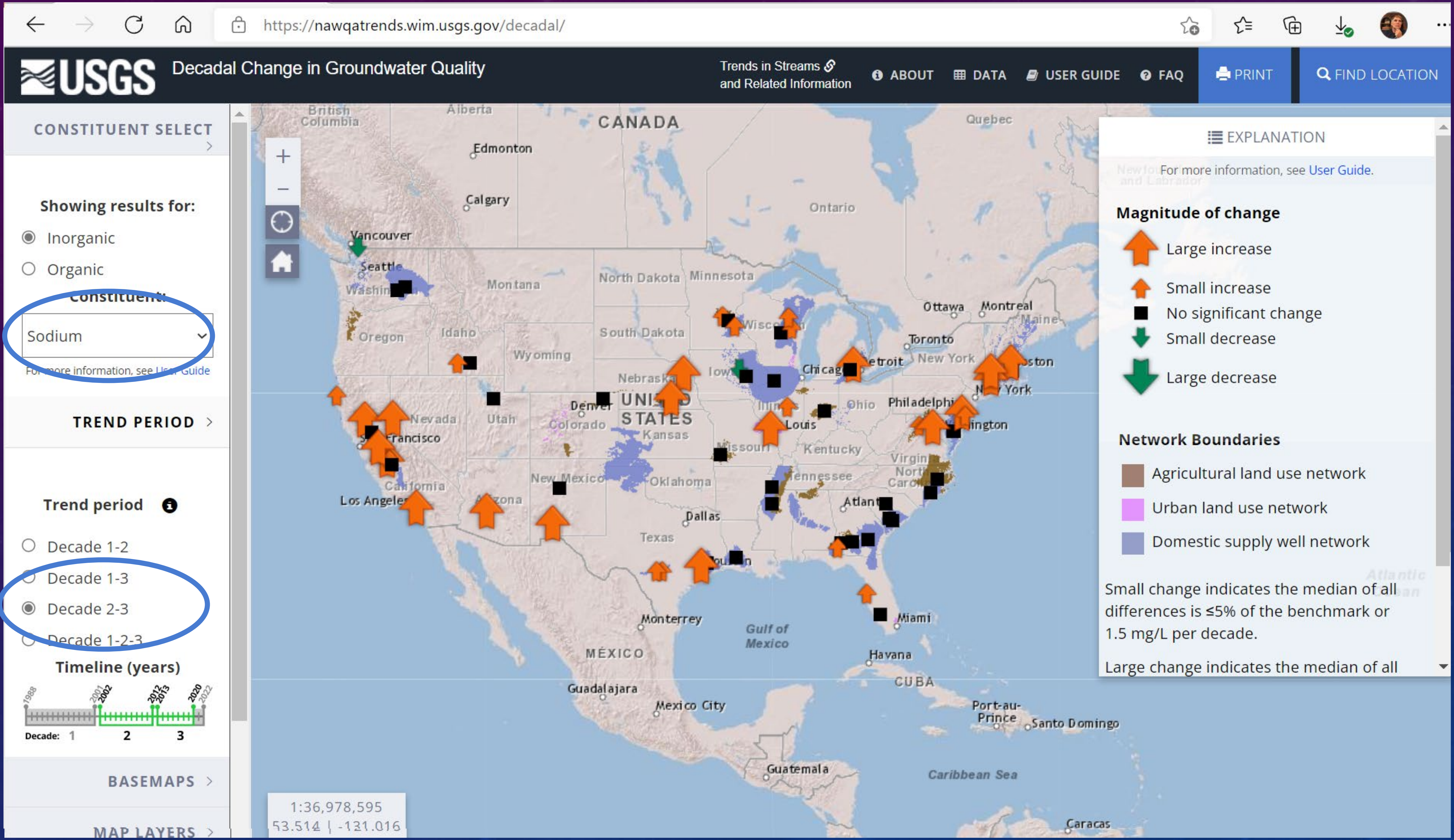
DECADAL NETWORKS

- National network
- Decadal change mapper: how are concentrations of contaminants in groundwater changing during decadal periods across the Nation.
- Each network of 25-30 wells represents a specific aquifer, region and sometimes land use.
- Nutrients, metals, radionuclides, and organic contaminants in groundwater are evaluated in a total of 88 networks.
- Decadal intervals: 1988-2001; 2002-2012; 2013-2020.


2032
2023
2031
2024
2030
2025
2026
2029
2027
2028

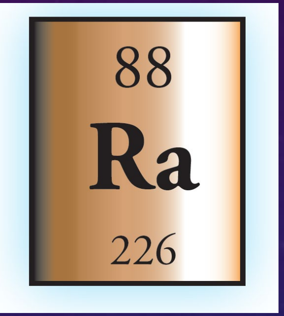
- Each network of 25-30 wells represents aquifer, region and sometimes land use.
- Icons on the national map represent a group of wells with similar characteristics.



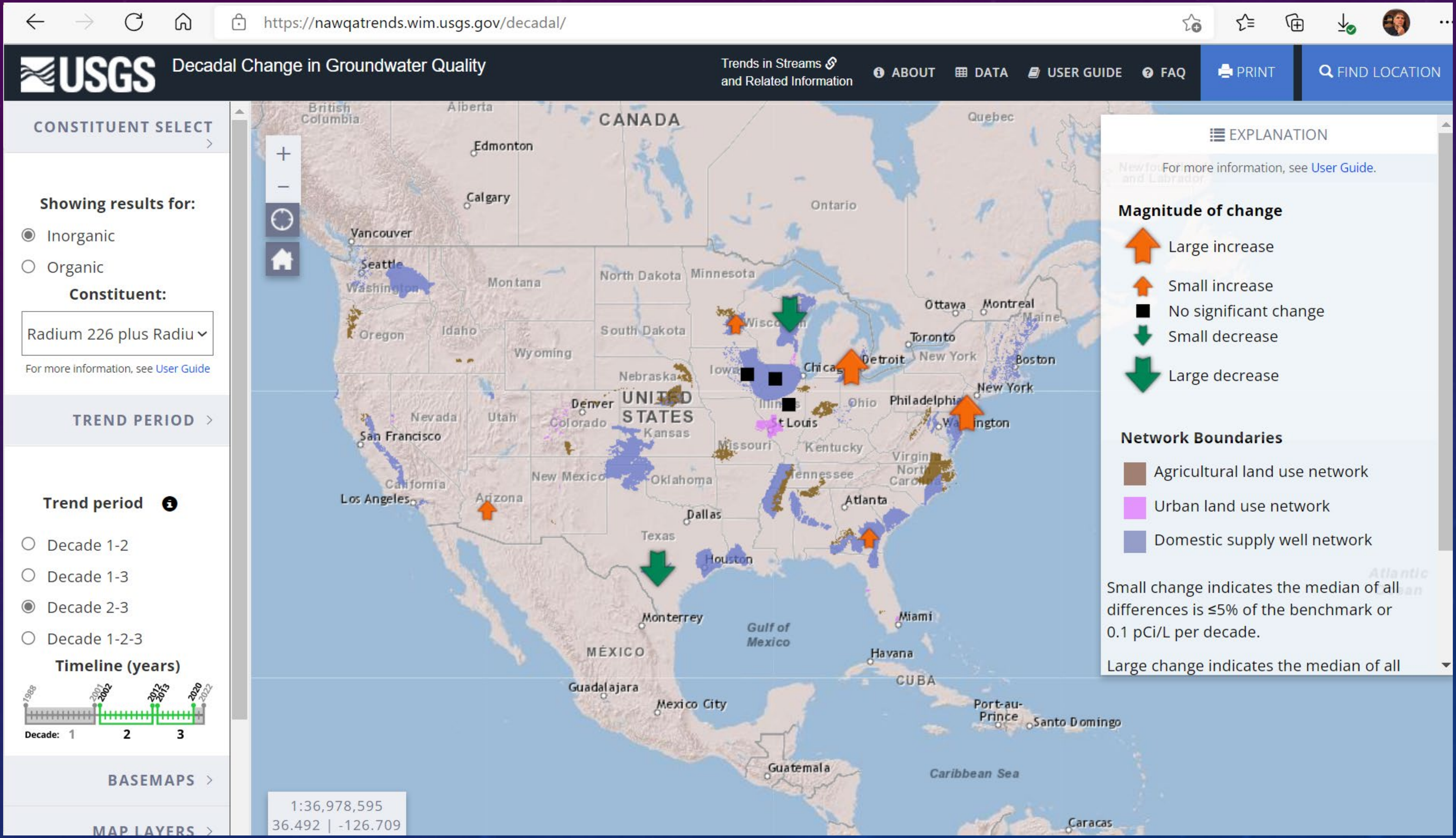


https://nawqatrends.wim.usgs.gov/decadal/
USGS
Decadal Change in Groundwater Quality
Trends in Streams and Related Information
ABOUT
DATA
USER GUIDE
FAQ
PRINT
FIND LOCATION
CONSTITUENT SELECT
Showing results for:
Inorganic
Organic
Constituent:
Sodium
For more information, see User Guide
TREND PERIOD
Trend period
Decade 1-2
Decade 1-3
Decade 2-3
Decade 1-2-3
Timeline (years)
1988
2001
2002
2012
2013
2020
2022
Decade: 1 2 3
BASEMAPS
MAP LAYERS
1:36,978,595
53.514 | -131.016
EXPLANATION
For more information, see User Guide.
Magnitude of change
Large increase
Small increase
No significant change
Small decrease
Large decrease
Network Boundaries
Agricultural land use network
Urban land use network
Domestic supply well network
Small change indicates the median of all differences is ≤5% of the benchmark or 1.5 mg/L per decade.
Large change indicates the median of all
CANADA
UNITED STATES
MÉXICO
Gulf of Mexico
CUBA
Caribbean Sea

RADIUM RESULTS FROM DECADAL NETWORK



- Radium was not sampled during 1st and 2nd decades at all networks
- By 2020, 10 networks had sufficient data for statistical analysis
  - That is – a minimum of 18 sites sampled over a near-decadal time period
- As used herein radium is sum of ^{226}Ra and ^{228}Ra
- The EPA Maximum Contaminant Level (MCL) for radium is 5 picocuries per liter (pCi/L)
- 5 networks increasing, 2 decreasing, 3 no change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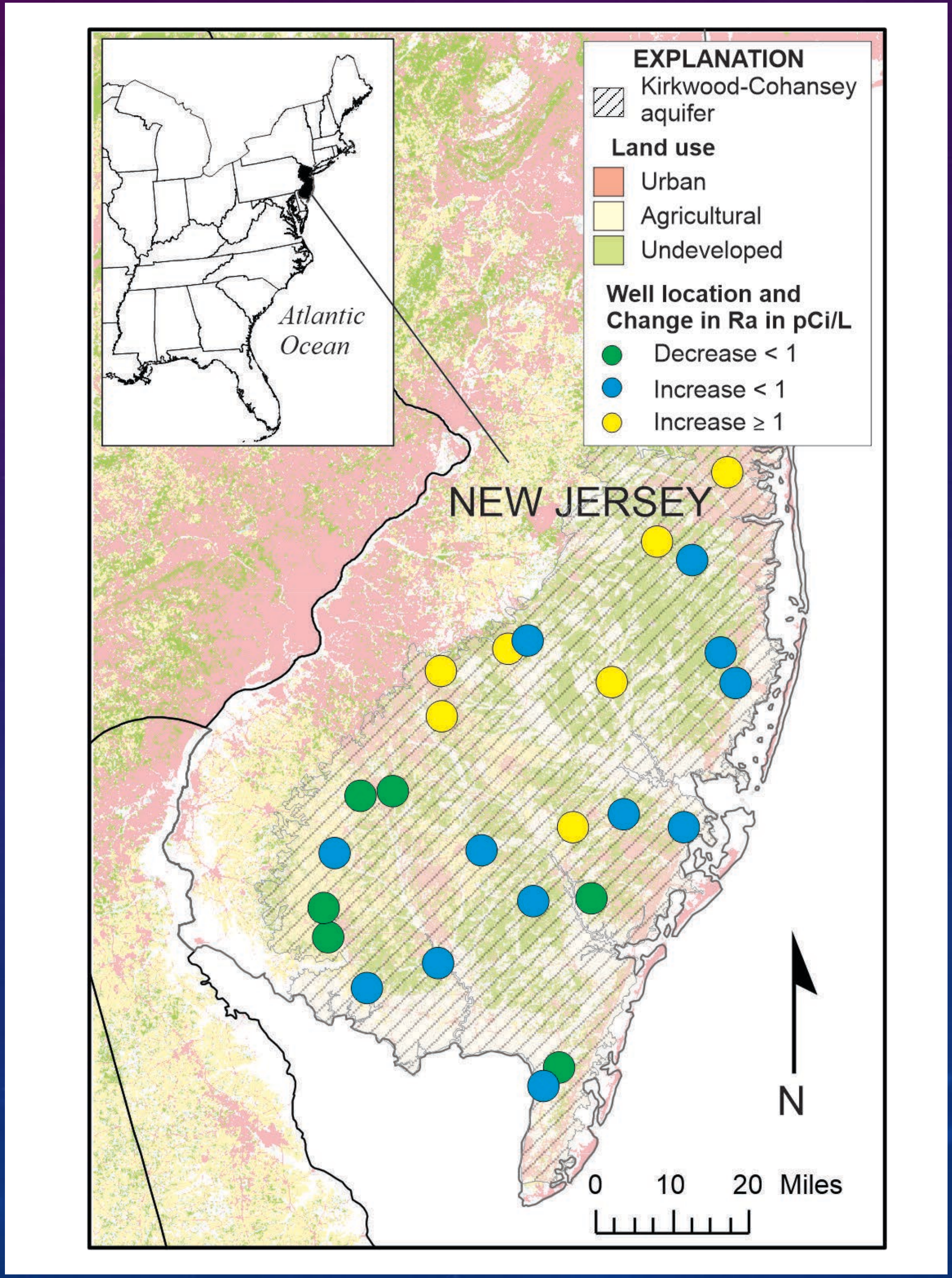
USGS
Decadal Change in Groundwater Quality
Trends in Streams and Related Information
ABOUT
DATA
USER GUIDE
FAQ
PRINT
FIND LOCATION
CONSTITUENT SELECT
Showing results for:
Inorganic
Organic
Constituent:
Radium 226 plus Radiu
For more information, see User Guide
TREND PERIOD
Trend period
Decade 1-2
Decade 1-3
Decade 2-3
Decade 1-2-3
Timeline (years)
1988
2001
2002
2012
2013
2020
2022
Decade: 1 2 3
BASEMAPS
MAP LAYERS
1:36,978,595
36.492 | -126.709
EXPLANATION
For more information, see User Guide.
Magnitude of change
Large increase
Small increase
No significant change
Small decrease
Large decrease
Network Boundaries
Agricultural land use network
Urban land use network
Domestic supply well network
Small change indicates the median of all differences is ≤5% of the benchmark or 0.1 pCi/L per decade.
Large change indicates the median of all

State	Aquifer	Probability	Median Change (in pCi/L)	Number of samples	Percent modern
Arizona	Basin and Range aquifers	0.03	0.01	29	21%
Iowa	Glacial aquifer system	0.20	0.12	19	53%
Georgia/Florida	Floridan aquifer system	0.01	0.10	25	8%
Michigan	Glacial aquifer system	0.01	0.33	23	38%
New Jersey	Kirkwood-Cohansey aquifer	0.001	0.35	25	76%
Illinois (lower)	Glacial aquifer system	0.22	-0.02	26	4%
Texas	Edwards-Trinity aquifer system	0.011	-0.28	21	81%
Illinois (upper)	Cambrian-Ordovician aquifer system	0.78	0.09	25	4%
Minnesota	Cambrian-Ordovician aquifer system	0.01	0.03	26	58%
Wisconsin	Glacial aquifer system	0.001	-0.34	18	72%

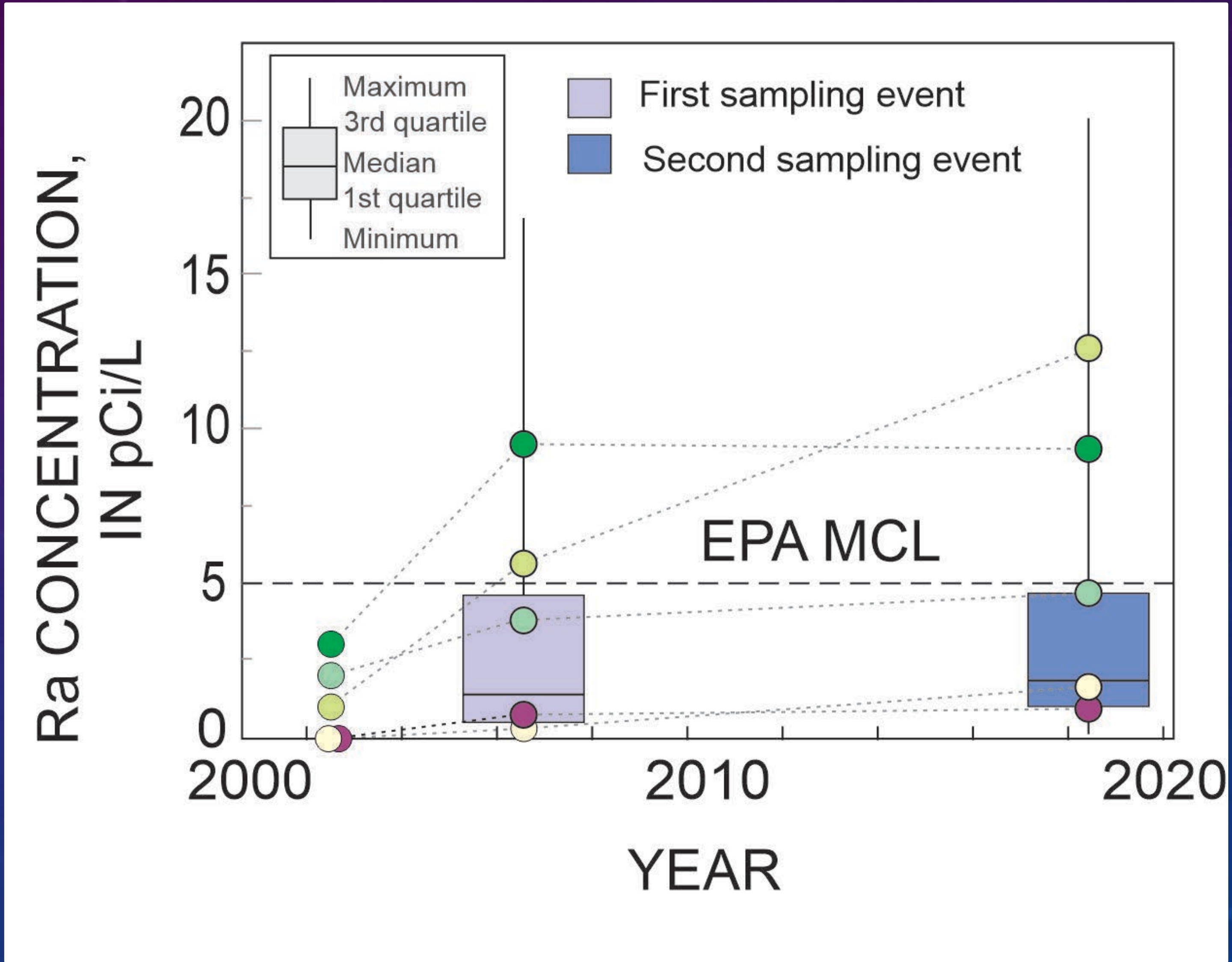
NEW JERSEY NETWORK

- Kirkwood-Cohansey aquifer
- Known for high radium concentrations
- Very low pH – median is less than 5 pH units
- Sandy aquifer with low sorption capacity

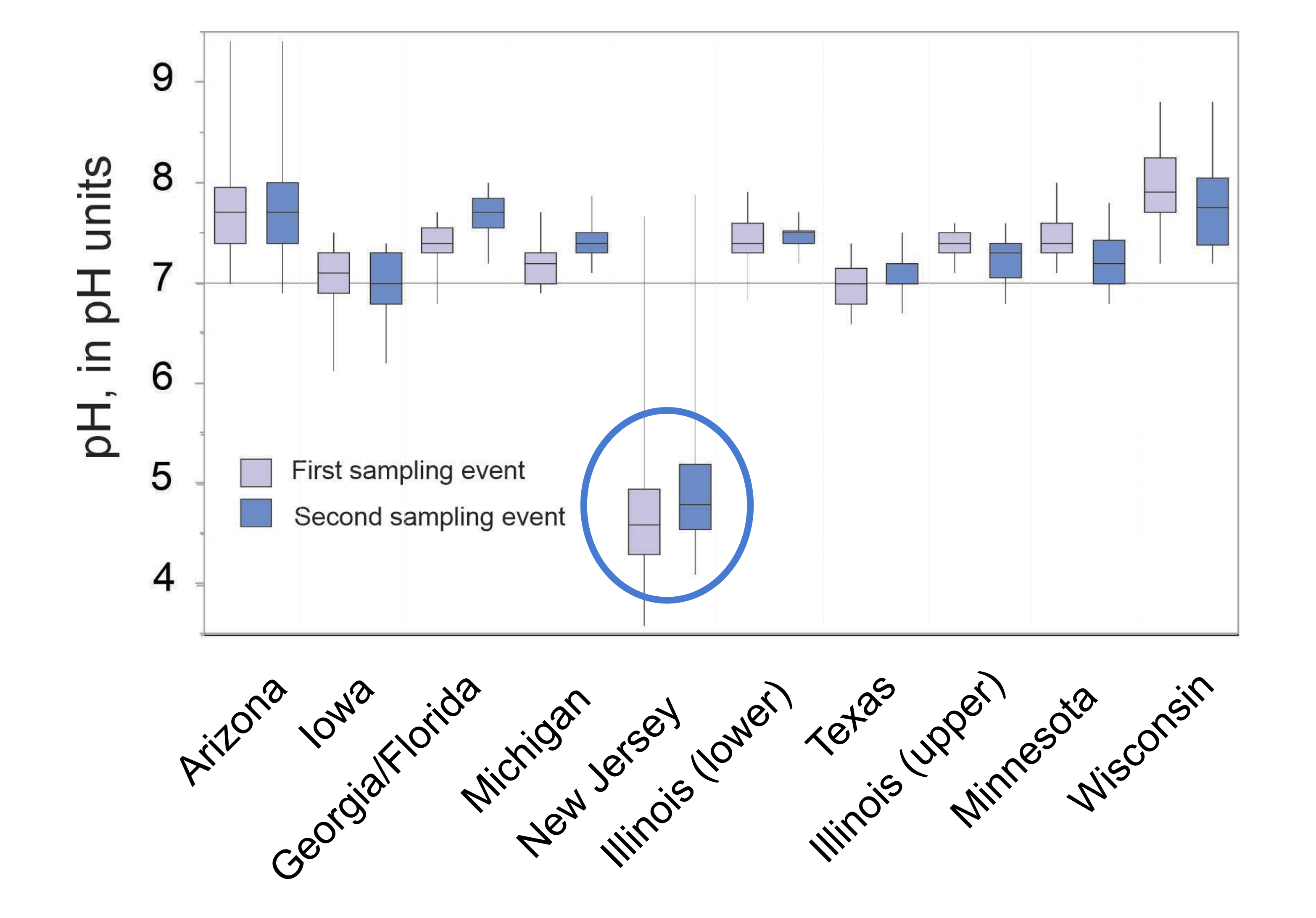


NEW JERSEY NETWORK

- Decadal change showed increase
- Subset of sites sampled 3 times

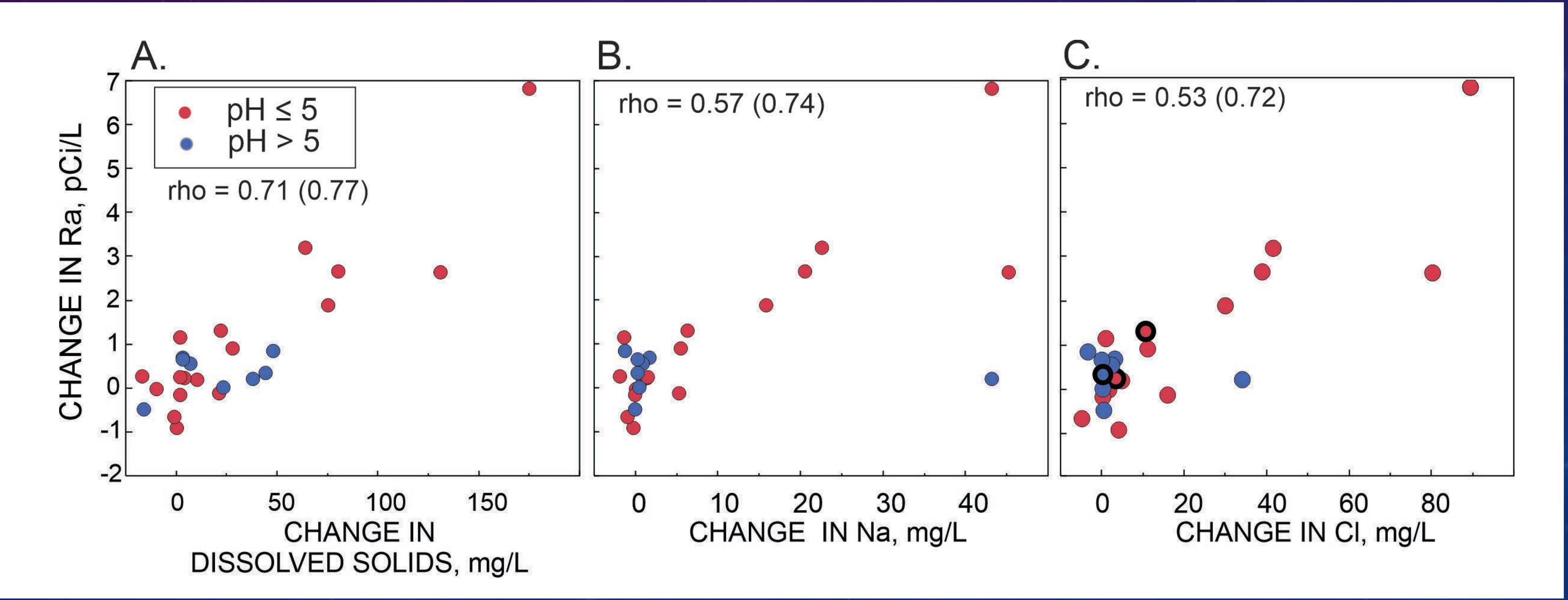


GEOCHEMICAL CONDITIONS IN NEW JERSEY NETWORK ARE UNIQUE



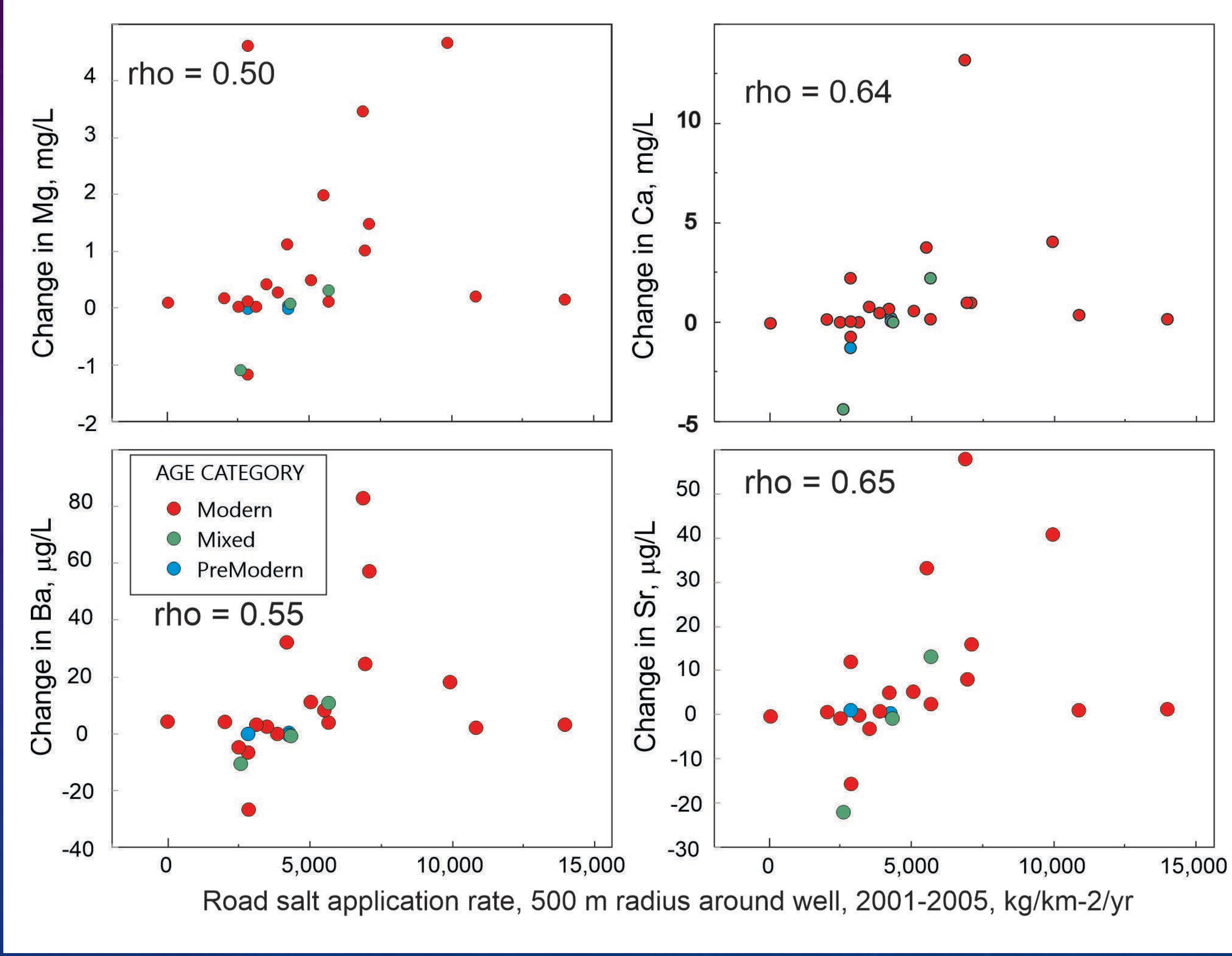
NEW JERSEY NETWORK

- Changes in concentration, 2006-2018
- Radium showed correlation with Cl, Na, and dissolved solids.
- Stronger in low pH samples
- What is the mechanism driving this?



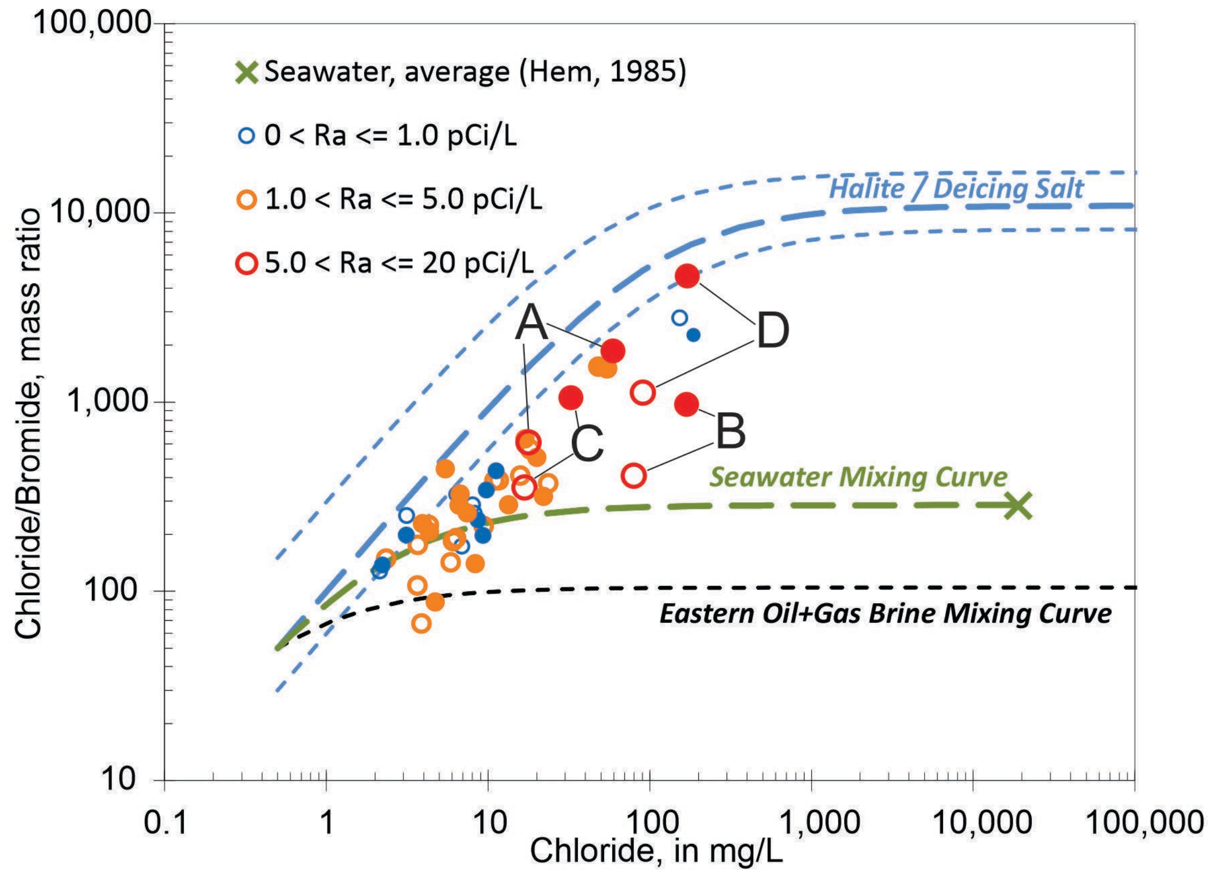
NEW JERSEY NETWORK

- Changes in concentration, 2006-2018
- Radium showed correlation with road salt application rate
- So did Cl, Na, and dissolved solids.
- So did Mg, Ca, Ba, and Sr



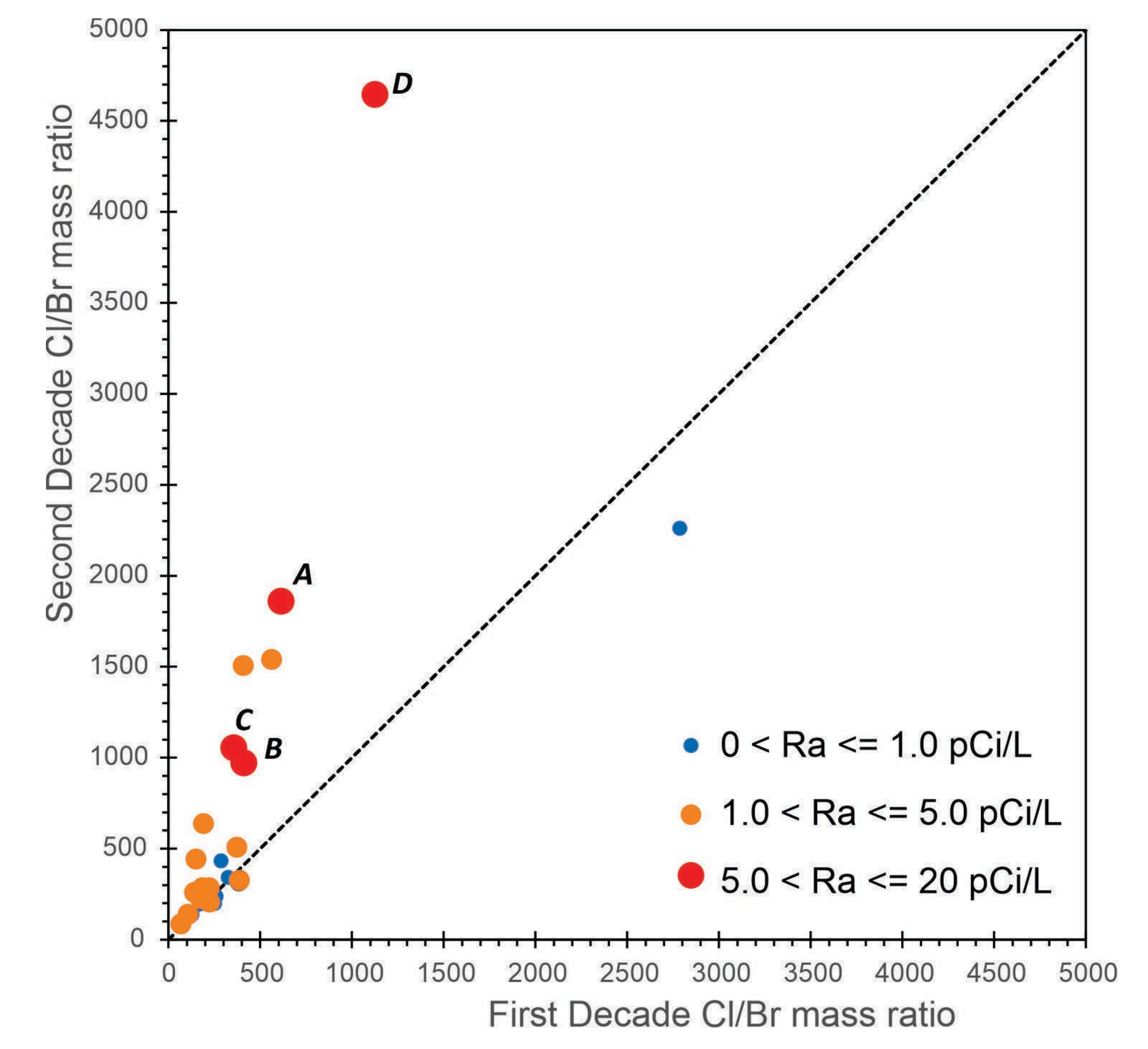
NEW JERSEY NETWORK

- Cl/Br mass ratios
- Open circles are 2006, filled circles are 2018
- Sites labeled A-D are largest increases in Ra
- Sites with highest increases in Ra moving toward halite/deicing salt mixing curve
- Brines not used in this area



NEW JERSEY NETWORK

- Cl/Br mass ratios
- Sites labeled A-D are largest increases in Ra
- Sites with highest increases in Ra moving toward higher Cl/Br mass ratios (road salt indicator)

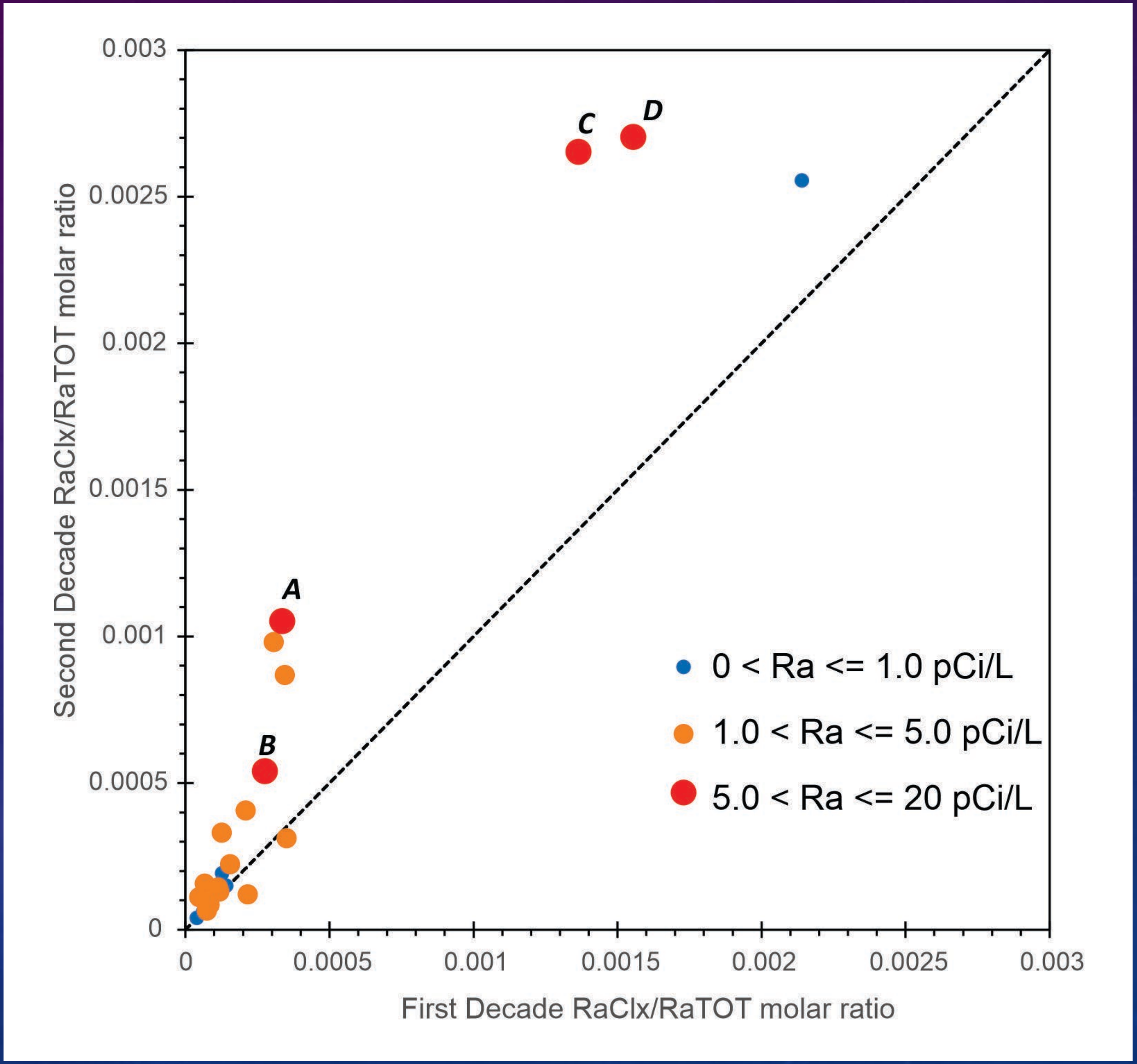


Displacement of Ra by Na

- Na is a weak competitor for sorption to displace Ra
- However large quantities of Na may replace Ca, Mg, Ba, Sr, and other divalent cations that are weakly bound
- Cascading effect – Na displaces Ca and Mg, which displace Ra
- Some Ca and Mg in salt may compete with Ra
- Increases in Na are less than increases in Cl supporting Na being retained on sorption sites.

Complexing of Ra with Cl

- RaCl+ and $RaCl_2$ complexes (RaClx) are more soluble
- RaClx/RaTOT ratios greater in second decade than in first decade
- Indicates increase in Cl complexation

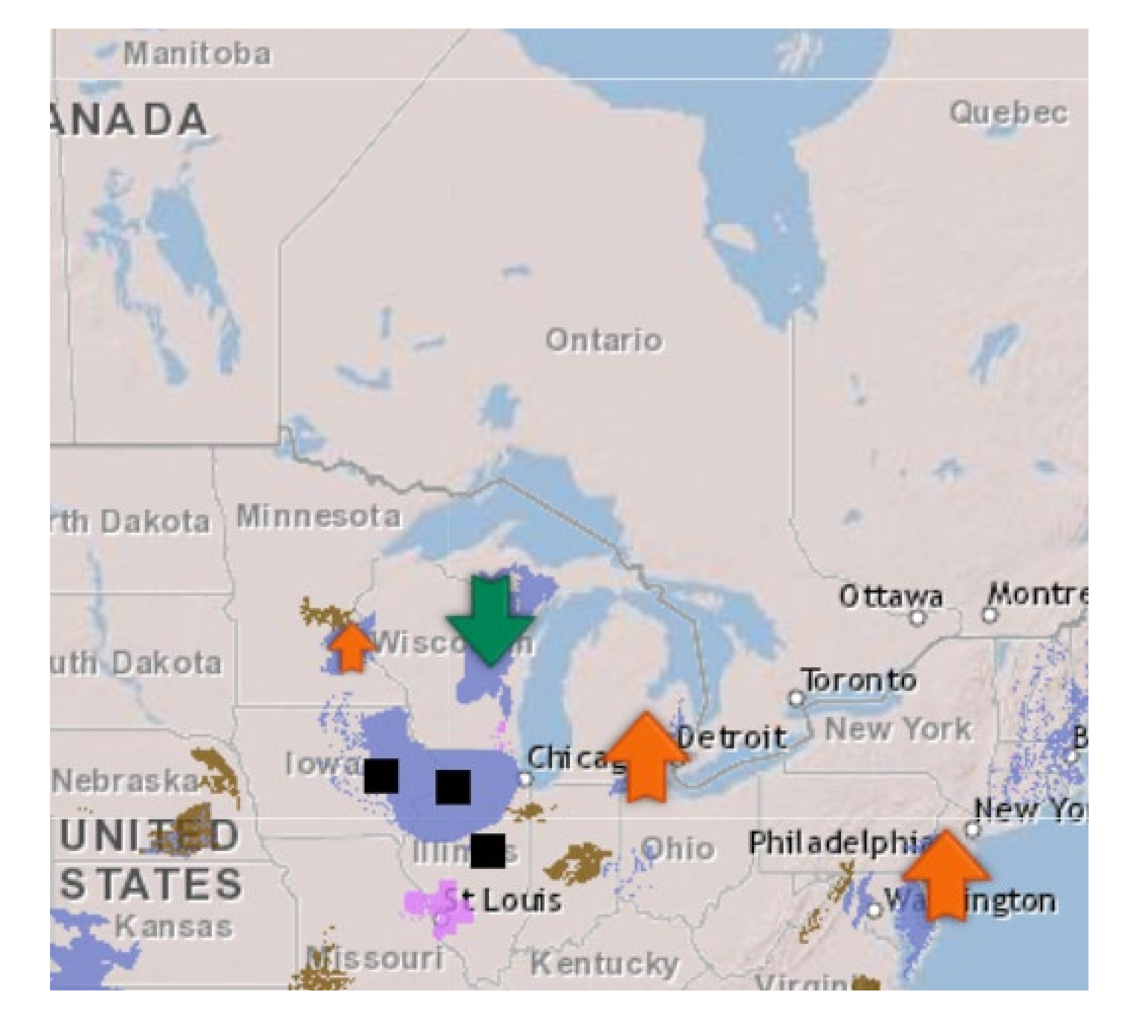


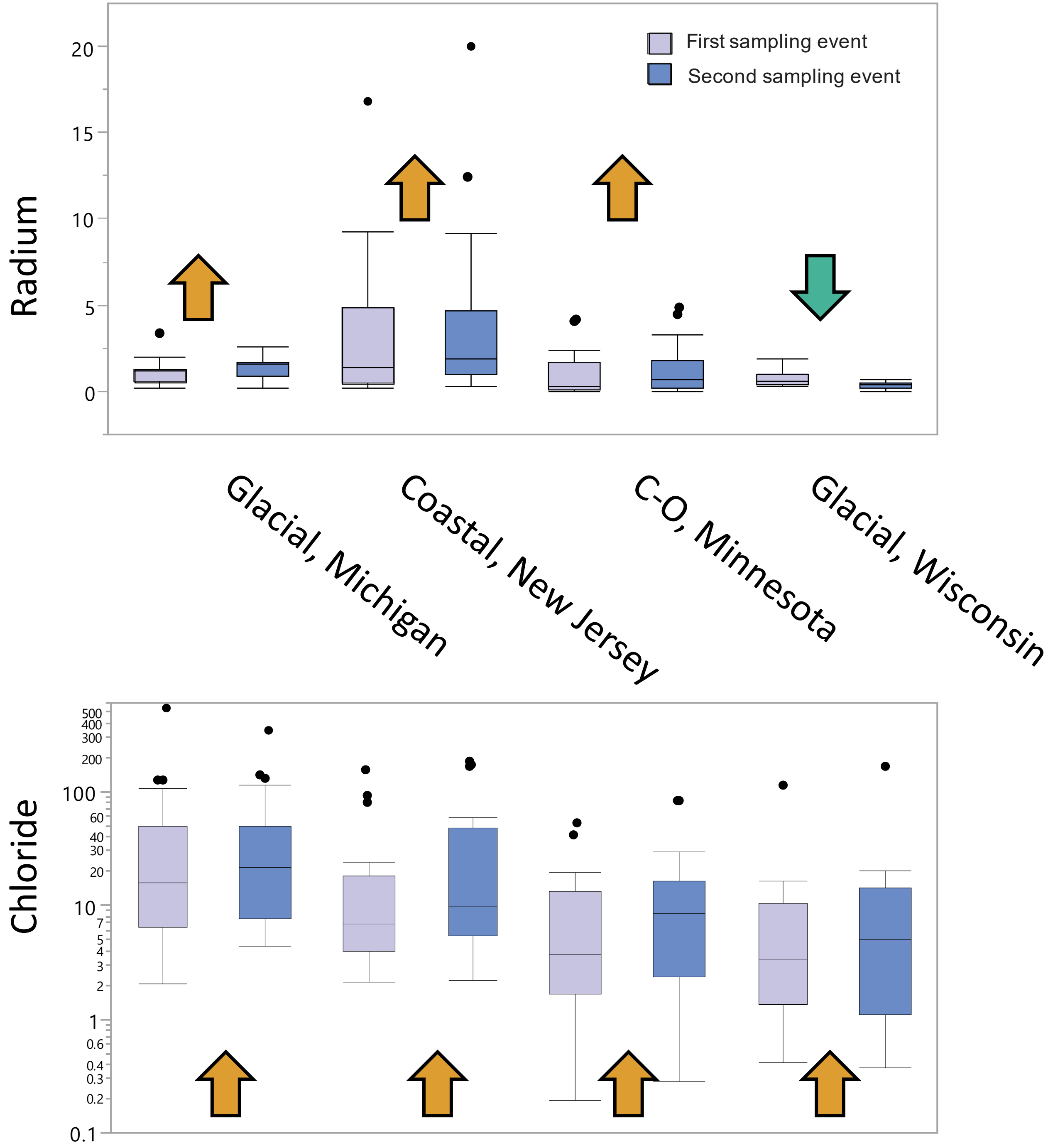
Other issues explored

- Ra in the road salt - not large enough to cause change in concentration
- Septic systems, population density, fertilizer use, and agricultural land use not correlated with changes in Ra.

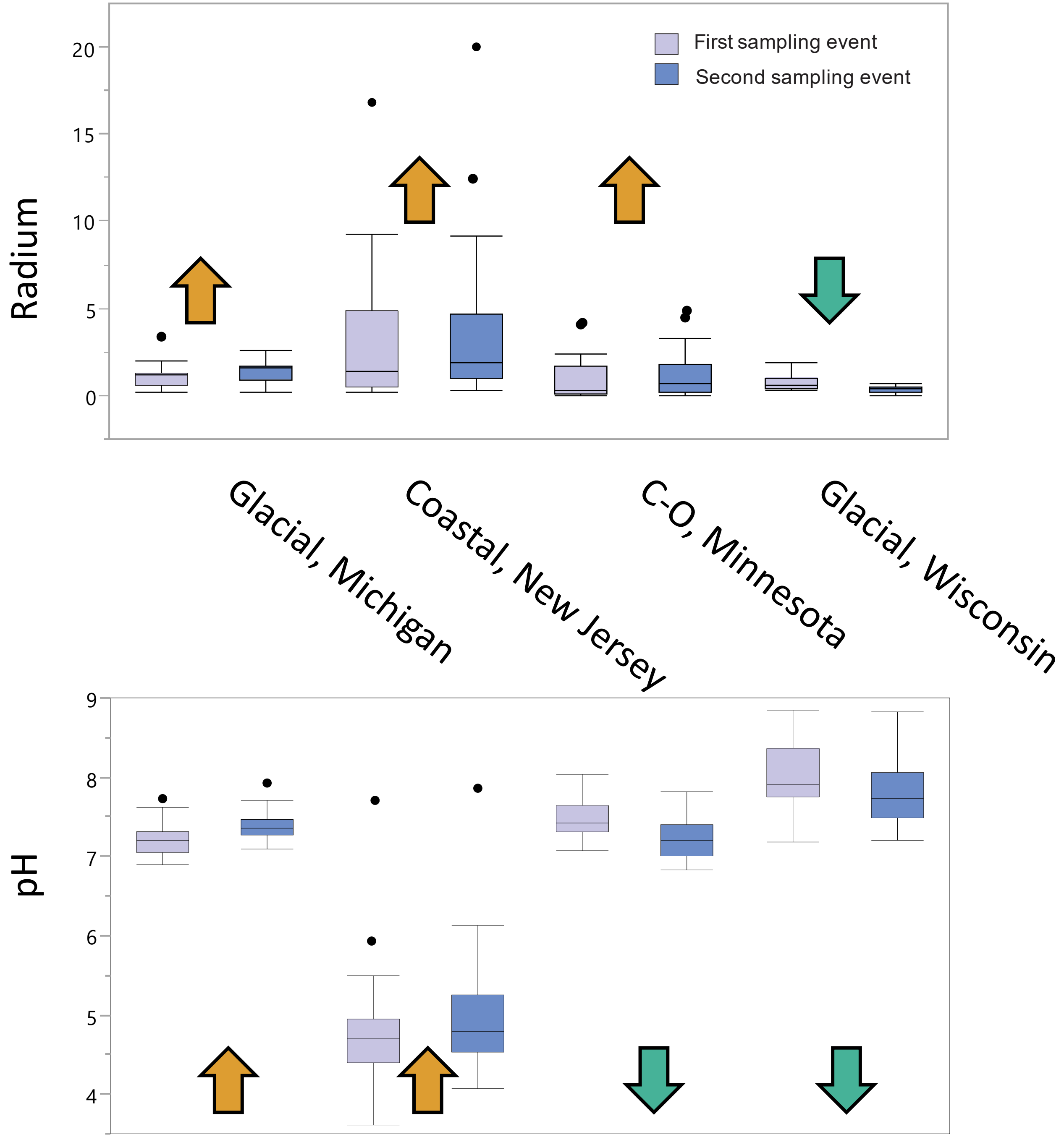
Other networks with changes in Ra

- Glacial aquifer, Michigan
- Glacial aquifer, Wisconsin
- Cambrian-Ordovician aquifer, Minnesota
- No correlation with road salt
- 2 of 3 have older groundwater
- Further study needed to understand changes in Ra

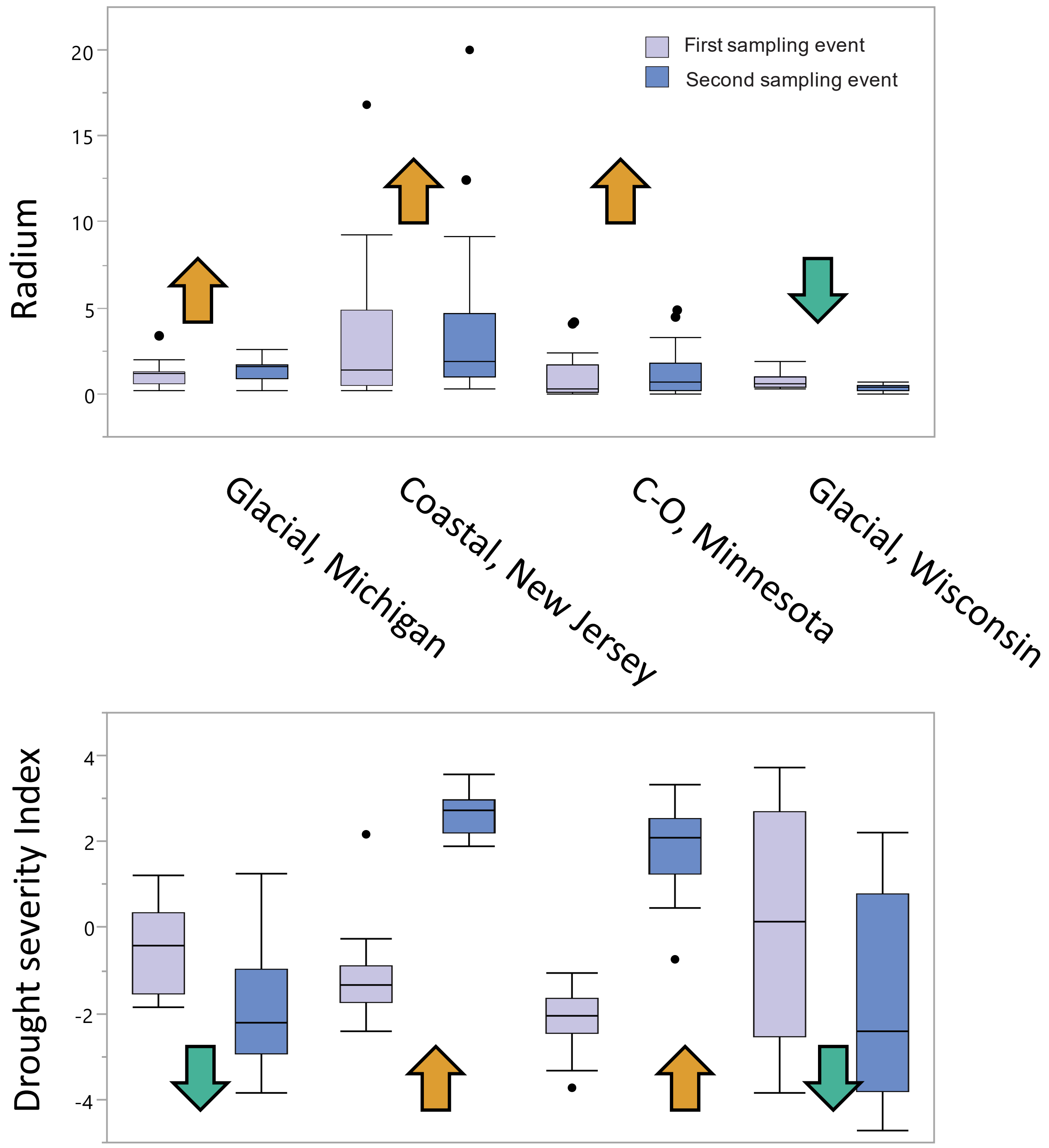




First sampling event
Second sampling event
Radium
20
15
10
5
0
Glacial, Michigan
Coastal, New Jersey
C-O, Minnesota
Glacial, Wisconsin
Chloride
500
400
300
200
100
60
40
30
20
10
7
5
4
3
2
1
0.6
0.4
0.3
0.2
0.1



First sampling event
Second sampling event
Radium
20
15
10
5
0
Glacial, Michigan
Coastal, New Jersey
C-O, Minnesota
Glacial, Wisconsin
pH
9
8
7
6
5
4



First sampling event
Second sampling event
Radium
20
15
10
5
0
Glacial, Michigan
Coastal, New Jersey
C-O, Minnesota
Glacial, Wisconsin
Drought severity Index
4
2
0
-2
-4

SUMMARY

- Na, Cl, and dissolved solids are increasing in a large percentage of networks across the country
- Road salt is a likely contributor to these increases in urban areas with a cold climate where road salt is used for deicing
- Increases in concentrations of Na and Cl are directly related to increases in Ra in a low pH aquifer
- Causes of changes in Ra in other aquifers have not been identified
- Effects of road salt on aquifers and other hydrologic systems cause issues beyond the increases in Na and Cl

QUESTIONS?

REFERENCES

Lindsey, Bruce D., Cravotta, Charles A. III, Szabo, Zoltan, Belitz, Kenneth, and Stackelberg, Paul, 2021, Relation between Road-Salt Application and Increasing Radium Concentrations in a Low-pH Aquifer, Southern New Jersey, ACS EST Water, v. 1. p. 2541-2547. https://doi.org/10.1021/acsestwater.1c00307

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