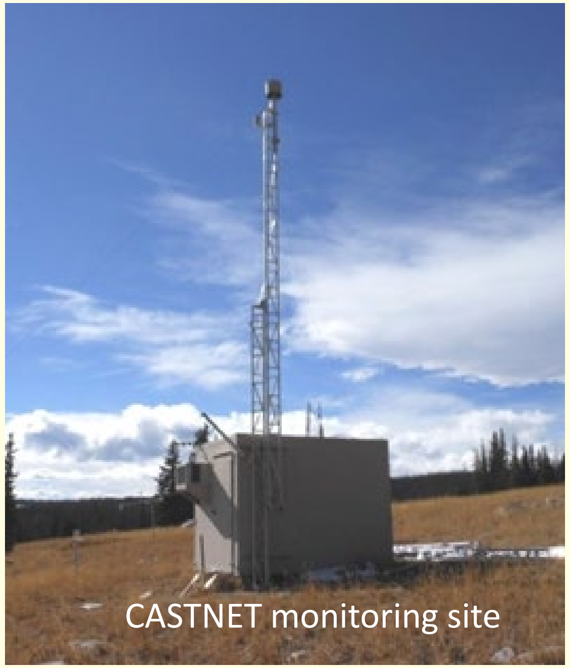
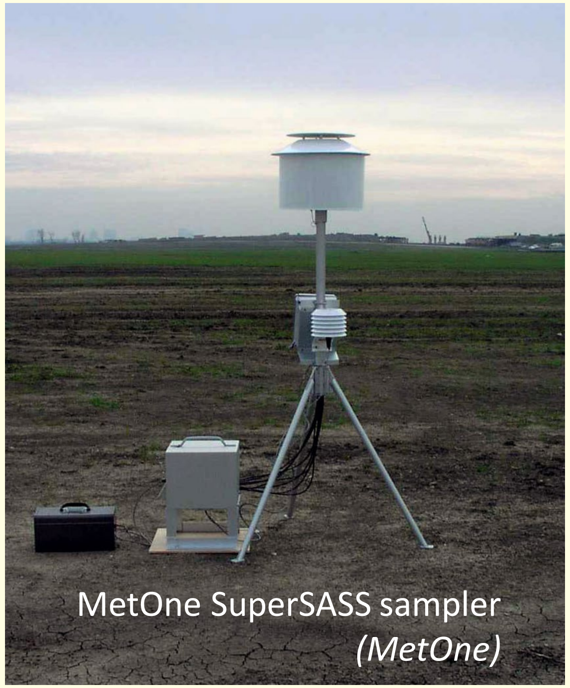
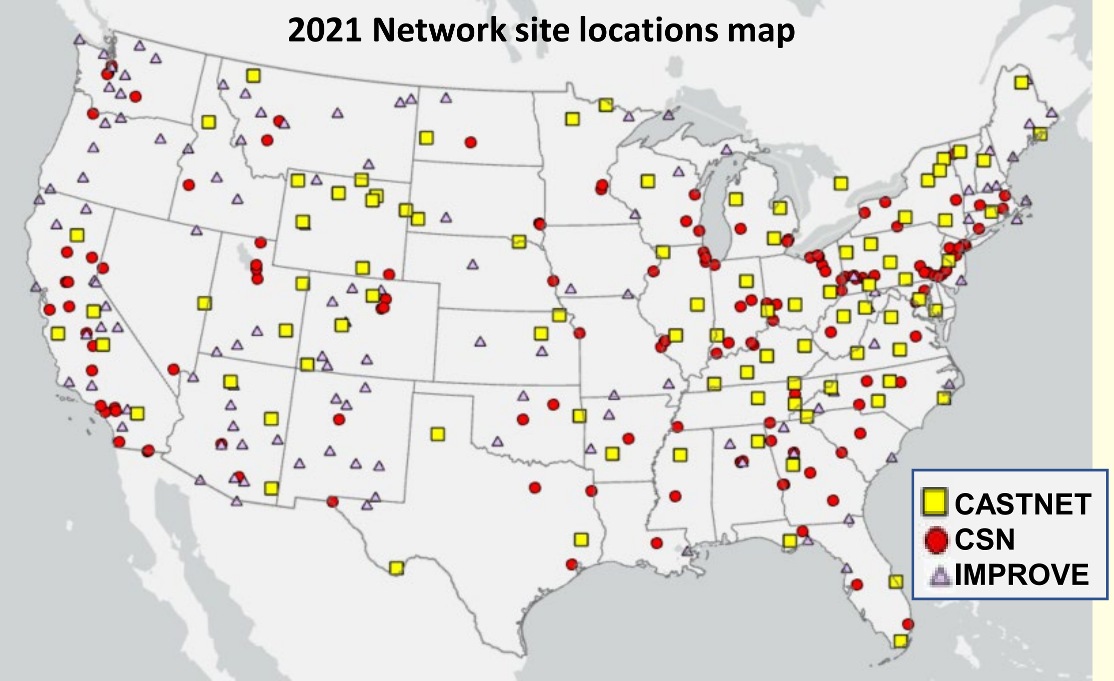


Exploration of Sulfate Concentrations in $PM_{2.5}$ across the United States using multiple monitoring networks and data fusion approaches

Beachley, G.M., Bowker, G.E., Puchalski, M.A., Sharac, T.

U.S. Environmental Protection Agency Office of Atmospheric Protection





U.S. ENVIRONMENTAL PROTECTION AGENCY

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Background and Motivation

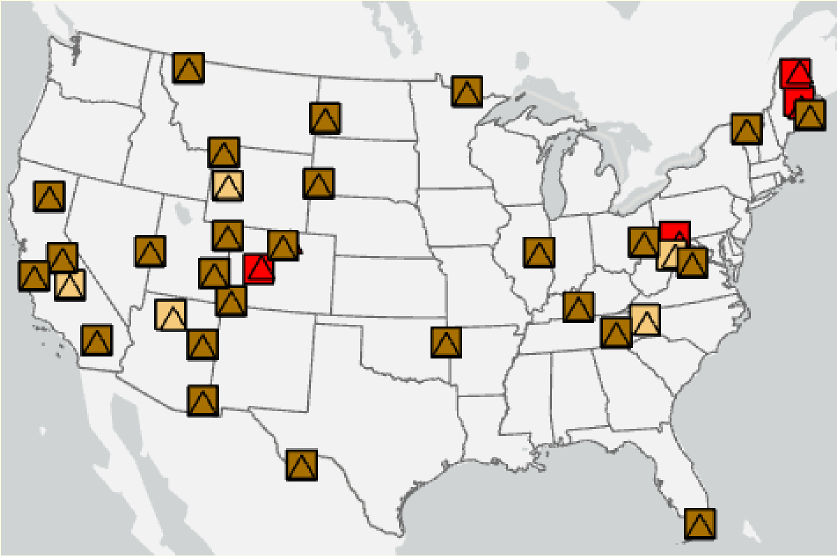
- National multipollutant monitoring networks in U.S. include the Chemical Speciation Network (***CSN***), Interagency Monitoring of Protected Visual Environments (***IMPROVE***), and Clean Air Status and Trends Network (***CASTNET***)
- These networks have similar atmospheric monitoring goals but include specific objectives that require differences in monitoring strategies:
  - IMPROVE and IMPROVE protocol sites are located in Class I areas or other sensitive ecosystems to monitor particulate species that impair visibility.
  - CSN is a component of the National $PM_{2.5}$ Monitoring Network focused on $PM_{2.5}$ precursors and particle formation
  - CASTNET is designed to assess trends in pollutant concentrations, atmospheric deposition, and resulting ecological impacts.
- Recent budget constraints to these monitoring networks threaten to reduce site density and efficacy of the networks
- Motivates a thorough comparison of the datasets to understand what applications and to what extent the datasets can be complementarily used
- Can the data from multiple networks be used in concert to expand spatial site density and to answer key scientific goals?
  - Establish better characterization of urban to rural concentration gradients
  - Evaluate trends and annual or seasonal variation of aerosols
  - Develop effective State Implementation Plans (SIPs) and assess regulatory compliance
- OAP with collaboration from National Atmospheric Deposition Program community has developed a robust data fusion product (TDep Measurement Model Fusion) used both nationally and internationally
  - Apply that framework to merge adjusted SO_4 concentrations from multiple networks to create a gridded SO_4 concentration surface

Approach

- Build on past studies investigating the comparability between SO_4, NO_3, and NH_4 measurements from the IMPROVE, CSN, and CASTNET networks (*e.g. Ames and Malm, 2001; Gego et al., 2005; Sickles and Shadwick, 2008*)
  - Extend analyses to recent datasets which will reflect recent shifts in concentrations of atmospheric pollutants
  - Identify sources of network sampling bias from instrumentation differences (e.g. size selection on inlet, volatilization, denuder impacts) and sampling frequency differences (e.g. weekly vs. 1 in 3 or 6 day)
    - Explore different aggregation intervals to homogenize variability from sampling frequency differences (*Gego et al., 2005)*
  - Assess any trends either from changes in potential sample methodology or atmospheric chemistry
- First step in the larger approach is to use Orthogonal Least Square (OLS) correlations of SO_4 at co-located sites to derive network-wide adjustment factors to scale datasets
  - ***Determine the correlation:*** Derivation of adjustment equations using optimal aggregation interval and site distance threshold to adjust SO_4 measurements
  - ***Adjust the datasets:*** We normalized with respect to CSN, but can normalize to CASTNET or IMPROVE depending on need
  - ***Utilize the dataset:*** Aggregated dataset to new interval period (e.g. annual or weekly maps) and used IDW data fusion to create concentration maps merging data from all networks

Co-located Site Locations from Network Comparisons from 1990 to 2022

IMPROVE to CASTNET

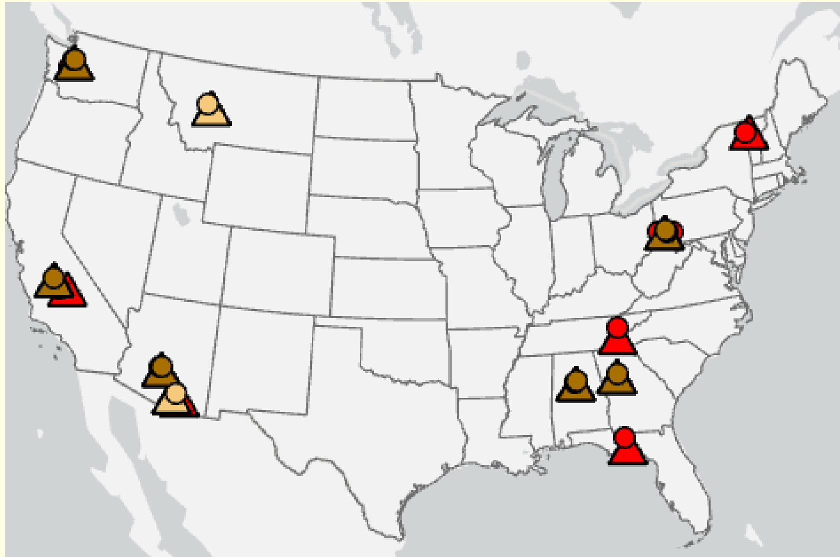


(1 km) N = 26 (25 unique)

(24 km) N = 34 (unique 33)

(50 km) N = 40 (unique IMPR(38), CAST(37))

IMPROVE to CSN

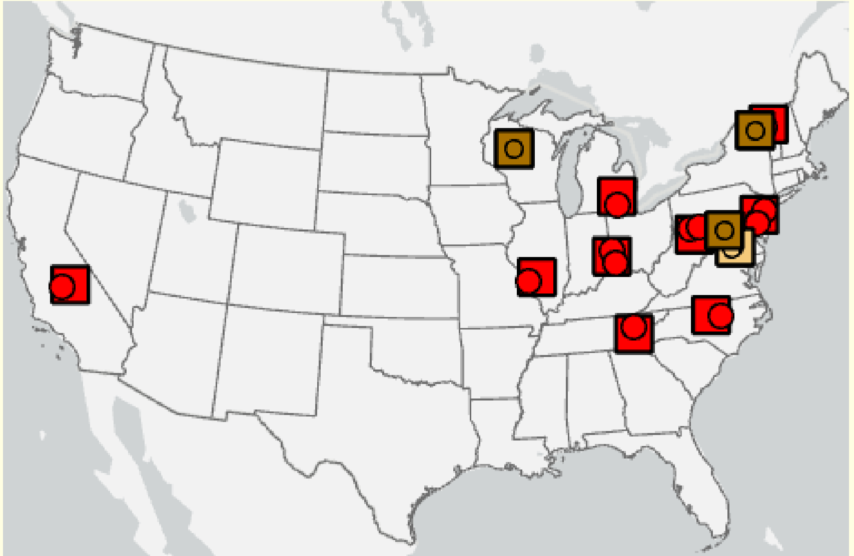


(1 km) N = 6

(24 km) N = 13 (unique IMPR (8), CSN(12))

(50 km) N = 22 (unique IMPR(13), CSN(21))

CASTNET to CSN



▲ - IMPROVE; ■ - CASTNET; ● - CSN

(1 km) N = 3

(24 km) N = 5 (unique CAST(4), CSN(5))

(50 km) N = 20 (unique CAST(13), CSN(20))

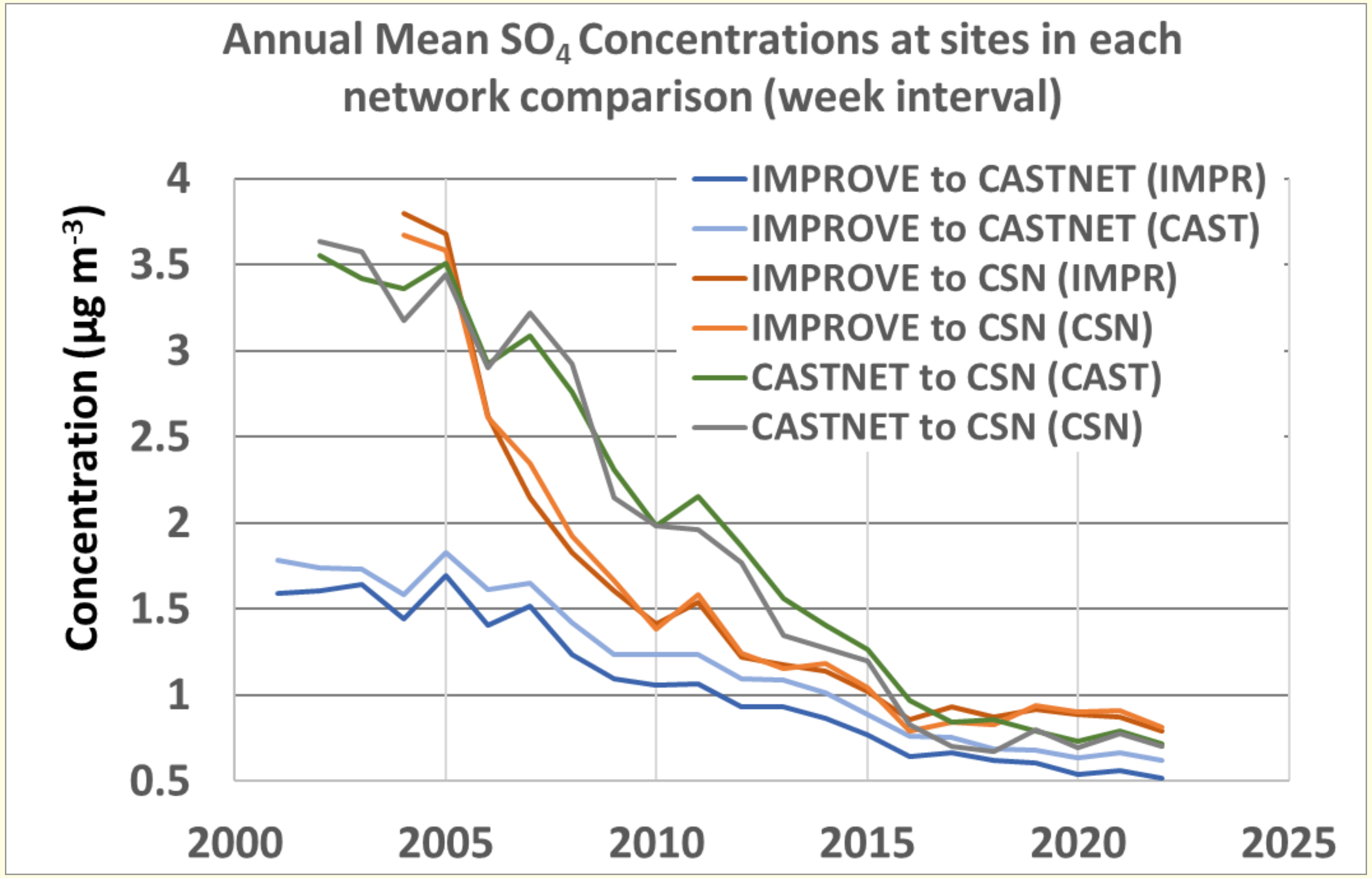
Looked at extending the distance threshold of "co-located" sites to increase number of site-pairs

- Could introduce spatial variability and potential issue with local source impacts.

Looked at different averaging intervals (weekly, biweekly, month, season, and year)

- Increasing the averaging intervals not expected to impact the OLS slope or intercepts
- Expected to increase the correlation by reducing the short-term variability and decreasing the magnitude of differences

Mean SO_4 Concentrations for Sites included in each Network Comparison

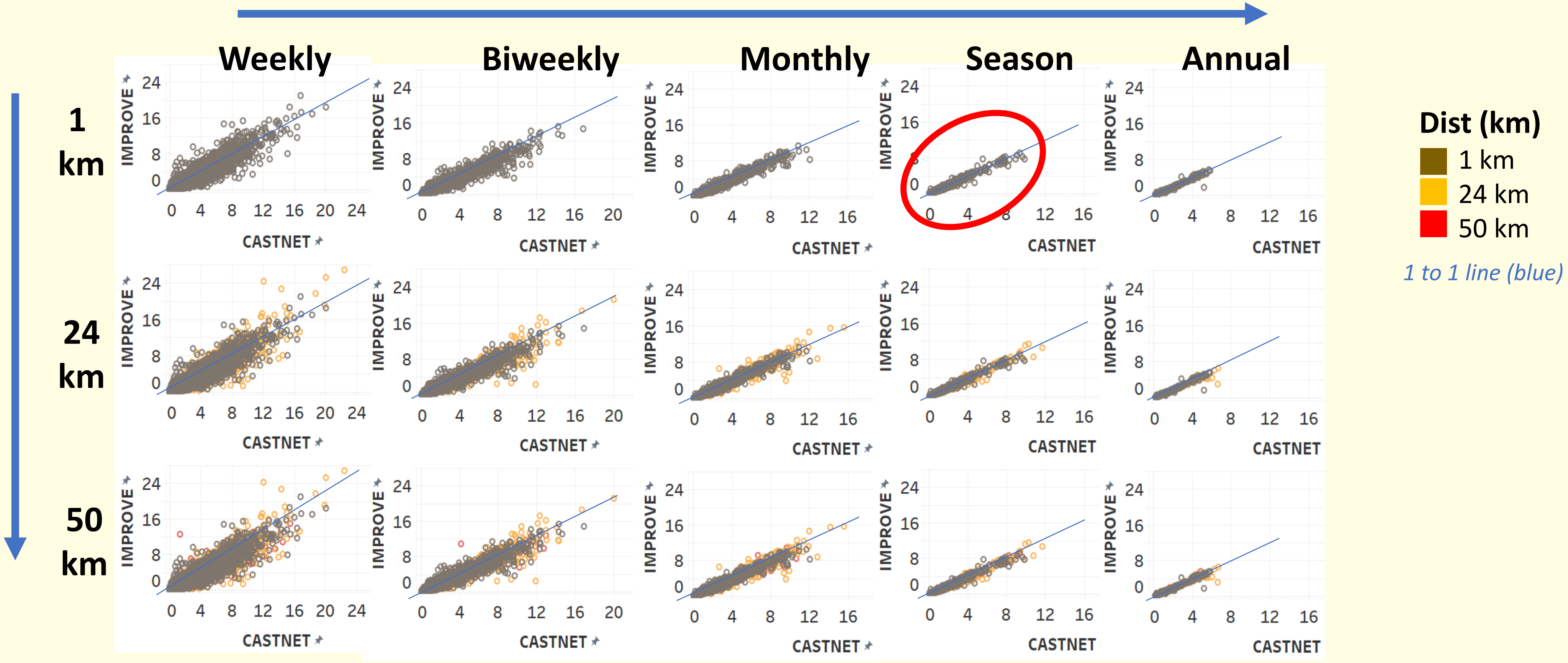


- Significant reductions in SO_4 concentrations at the co-located sites
  - Note lower concentrations in sites included in the IMPROVE to CASTNET comparison as they are mostly located in the Western U.S.
- Correlation statistics may reflect this reduction in SO_4

Comparisons for IMPROVE to CASTNET SO_4 (1990 to 2022)

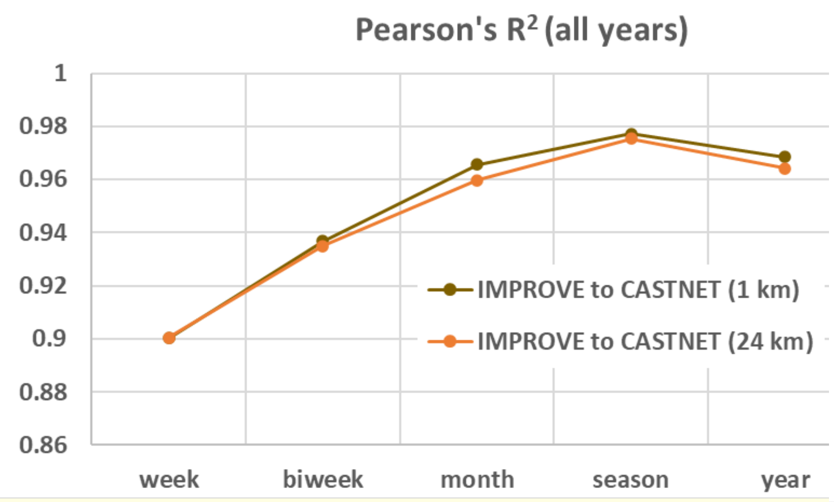
- Increasing aggregation interval length
- Decreasing outliers

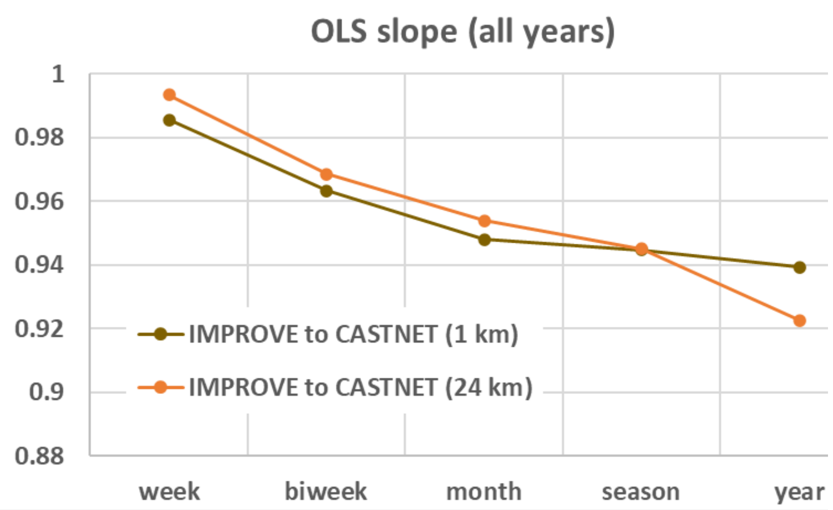
- Increasing co-location site distance
- Increasing N
- Increasing spatial variability and outliers

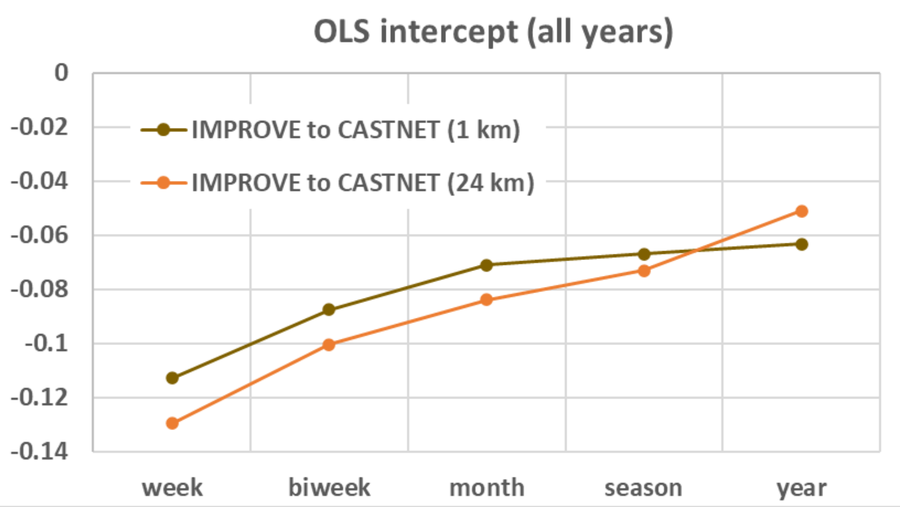


- IMPROVE to CASTNET comparison has adequate site-pairs, so will use only co-located sites within 1 km
- **Season** is a good aggregation interval
  - Need to add minimum threshold of observations per site-pair comparison (would improve correlations)

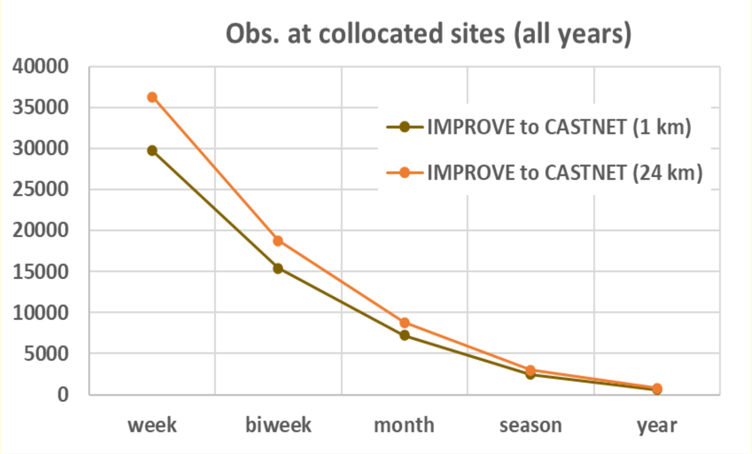
Statistical Measures for IMPROVE to CASTNET SO_4 (1990 to 2022)







- Increasing aggregation interval improves the correlation up to season
- Also impacts the OLS slope and intercept parameters, due to high outliers being averaged out
  - Note the change of those parameters slows after monthly interval

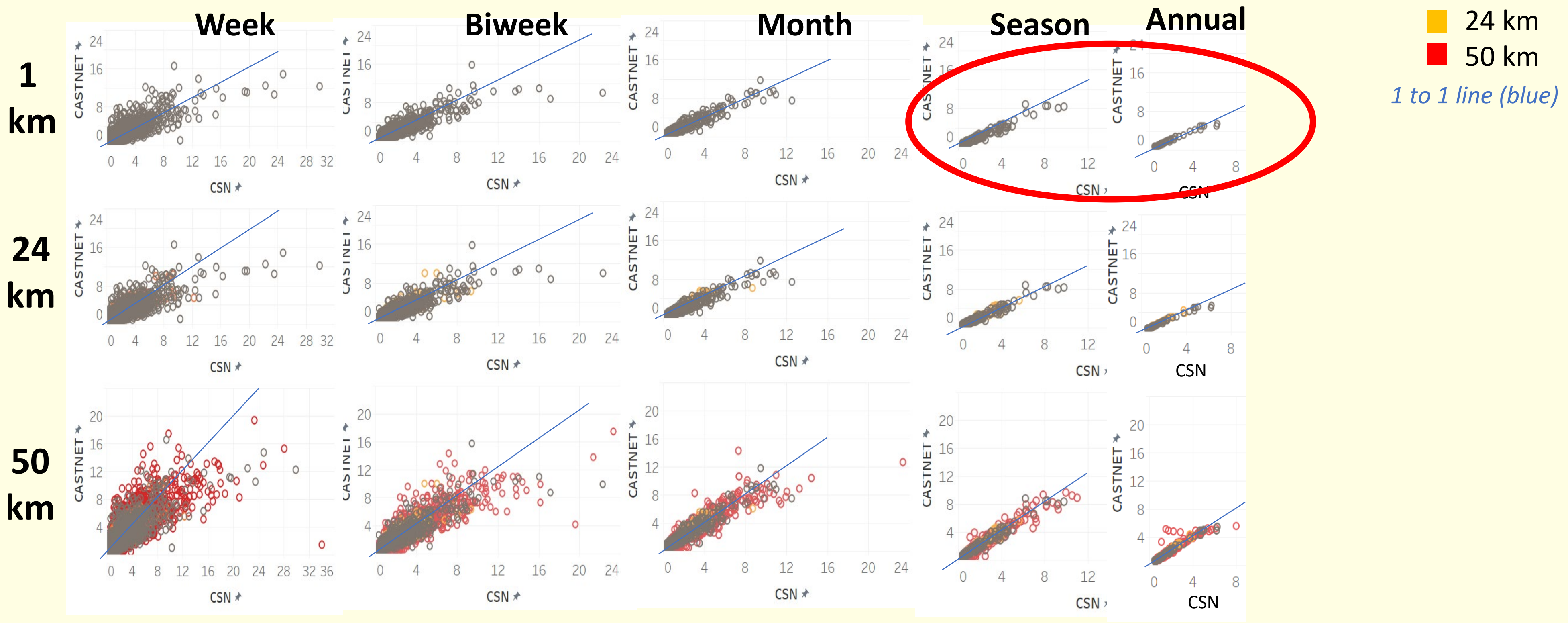


- Site-pair observations diminish with longer averaging intervals
- **Seasonal is optimal averaging interval** (highest correlation, stable OLS parameters, adequate site-pair observations)

Comparisons for CASTNET to CSN SO_4 (2002 to 2022)

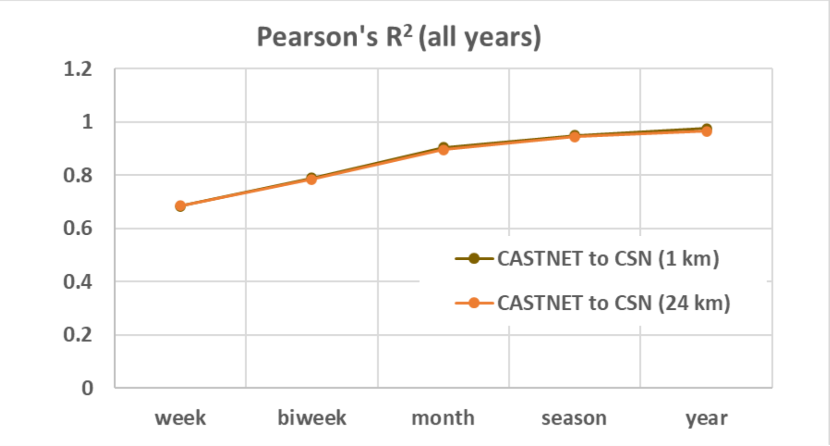
- Increasing aggregation interval length
- Decreasing outliers

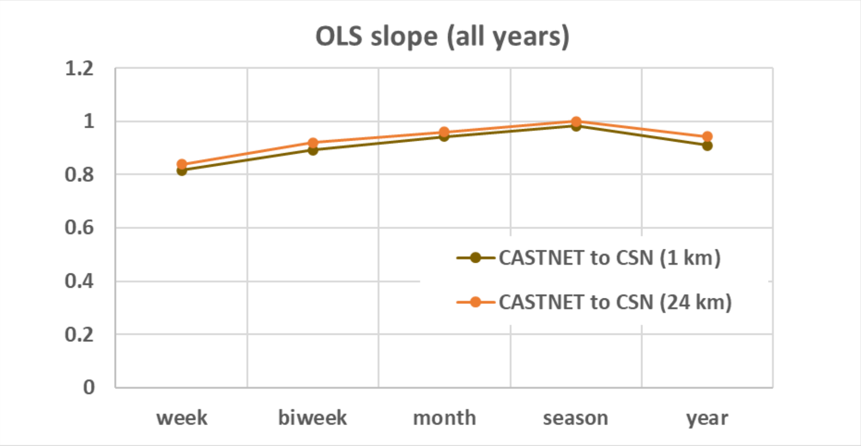
- Increasing co-location site distance
- Increasing N
- Increasing spatial variability and outliers

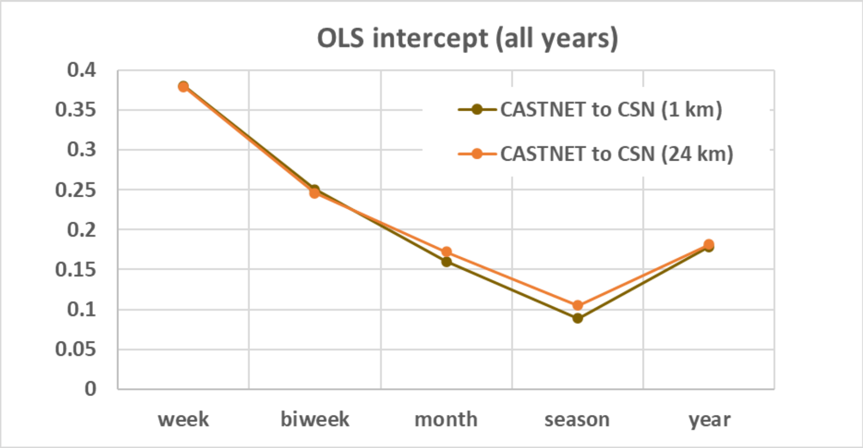


- Fewer co-located site-pairs (*N = 3*) with poorer correlations and more outliers
    - All co-located CSN sites have 1 in 6 day sampling
- Extending co-located site radius to 24 km adds 2 more sites (*N= 5*) without noticeable degradation
    - Extending to 50 km adds 15 more site-pairs, but with a large degree of spatial variance (possibly influenced by urban/rural site locations)
- **Seasonal or annual are the optimal aggregation intervals**

Statistical Measures for CASTNET to CSN SO_4 (2002 to 2022)







Seasonal or Annual is best interval for CASTNET to CSN comparison

- Annual shows some instability in OLS parameters (low N?)

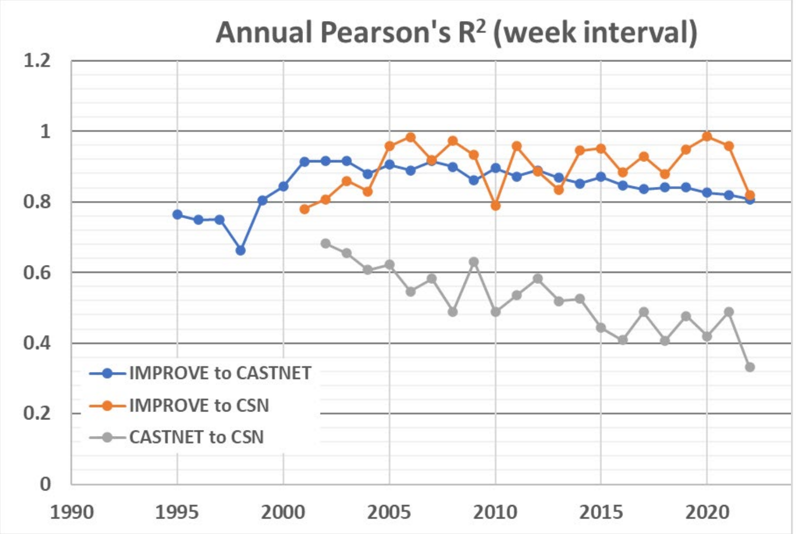
Extended 24 km co-located site radius shows similar statistics, but no improvement

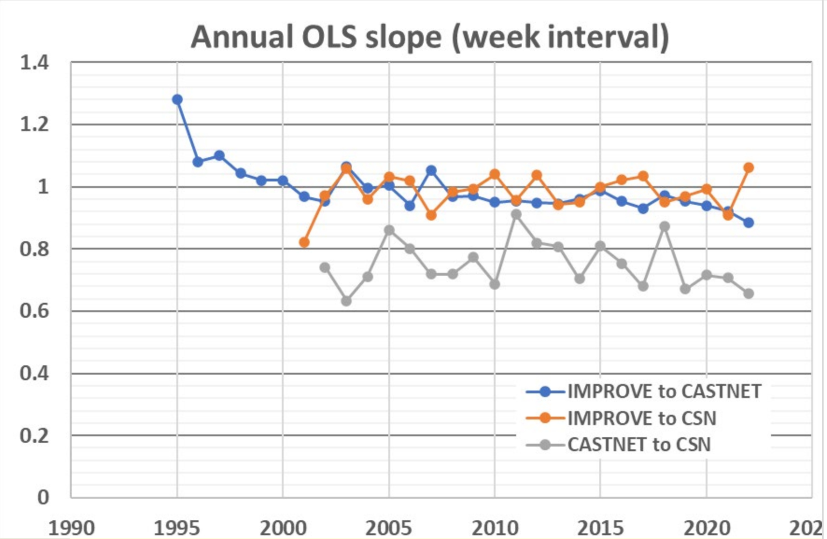
Used **Seasonal interval at 1 km** for consistency with other comparisons

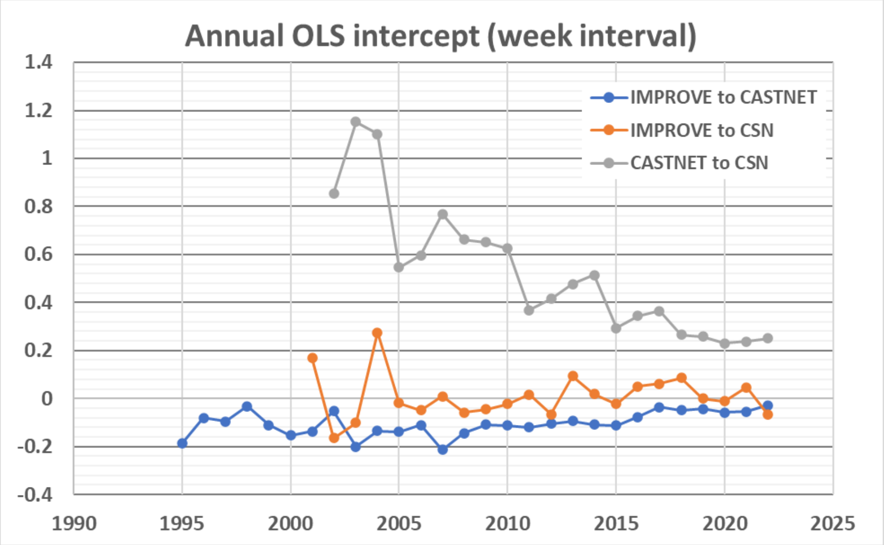
Comparison for IMPROVE to CSN SO_4 (2001 to 2022) also run

- Observed tighter correlations at smaller intervals (due to averaging daily measurements to weekly)
- Increasing the co-location site distance threshold added too much spatial variance
- Statistics indicated that ***Month or Season*** is optimal aggregation interval –***chose Seasonal for consistency***

Annual Stability of OLS Statistics for Co-located Sites (within 1 km) with Weekly Interval







Tests the sensitivity of the correlation statistics over time especially given the reductions in SO_4

- Note that analyzing correlations for each year reduces the Nobs, an issue for the CASTNET to CSN comparison

Do trends in OLS statistics change over time?

- IMPROVE to CASTNET (from 2007): decrease in R^2 and slope, increase in intercept
- IMPROVE to CSN: no trend
- CASTNET to CSN: decrease in R^2 and intercept

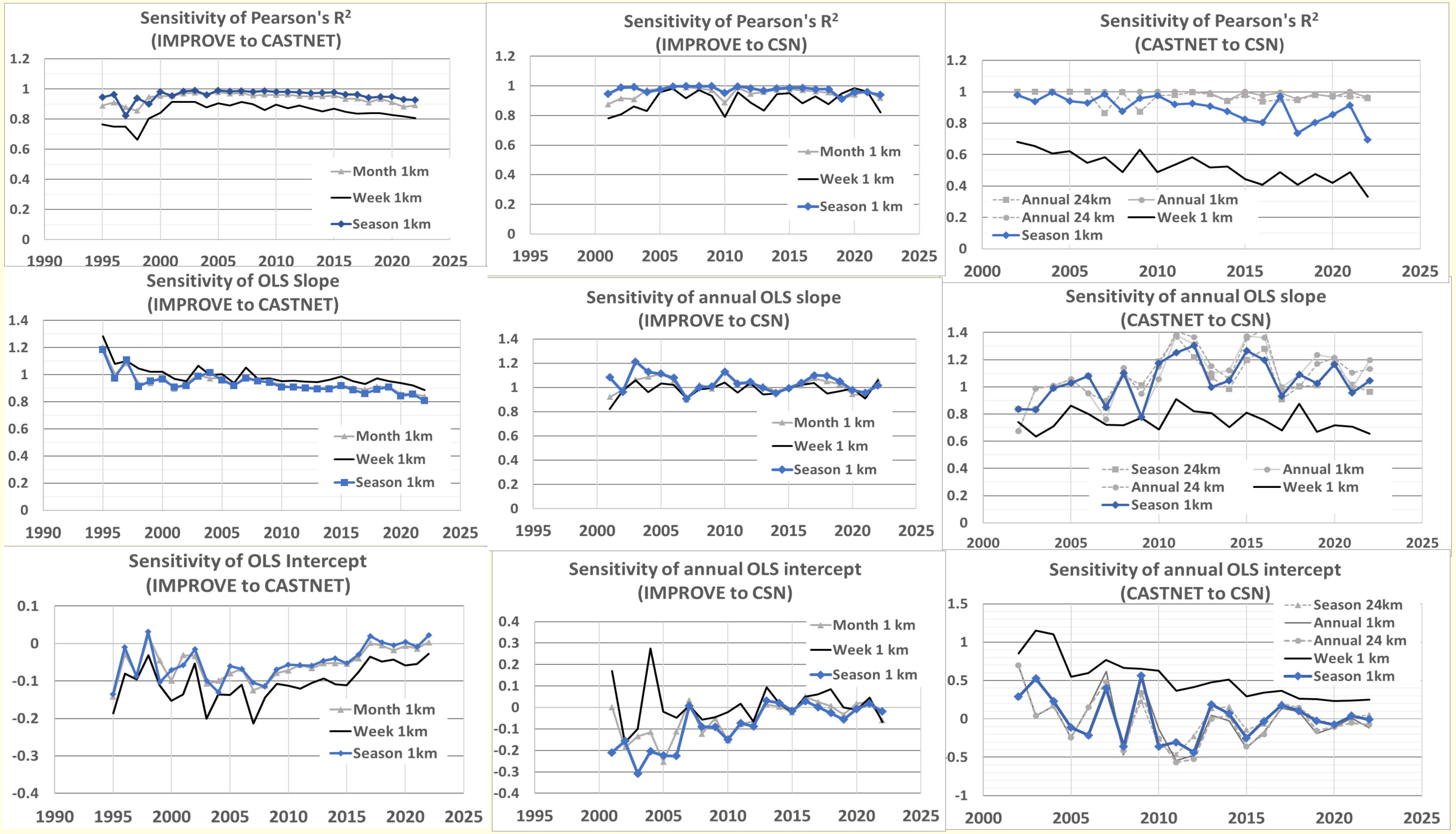
Are these trends real or driven by outliers?

Sensitivity of Annual Trends in Correlation and OLS Parameters to Aggregation Interval

R^2

OLS slope

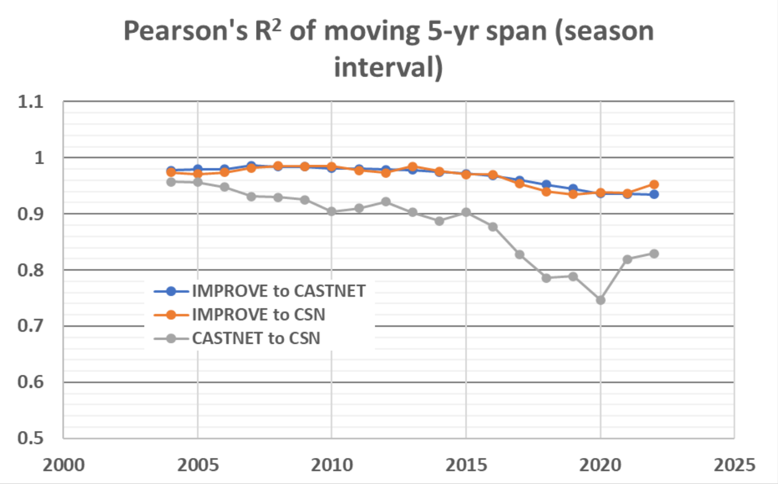
OLS intercept

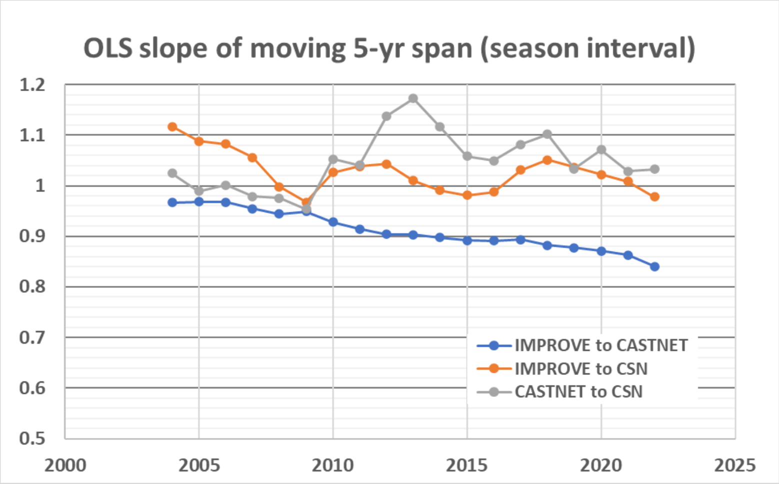


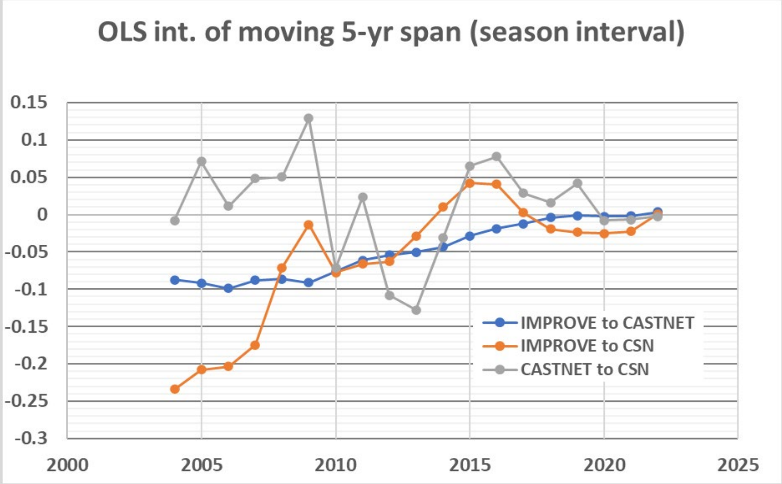
- Trends from weekly interval are still present in seasonal interval
  - May be due to lower concentrations and MDL issues
- Significant differences from weekly, but similar trends for longer aggregates (month/season/annual)
- Improvement in correlation and increased slope (step-change ~2010?) with longer interval for CASTNET to CSN

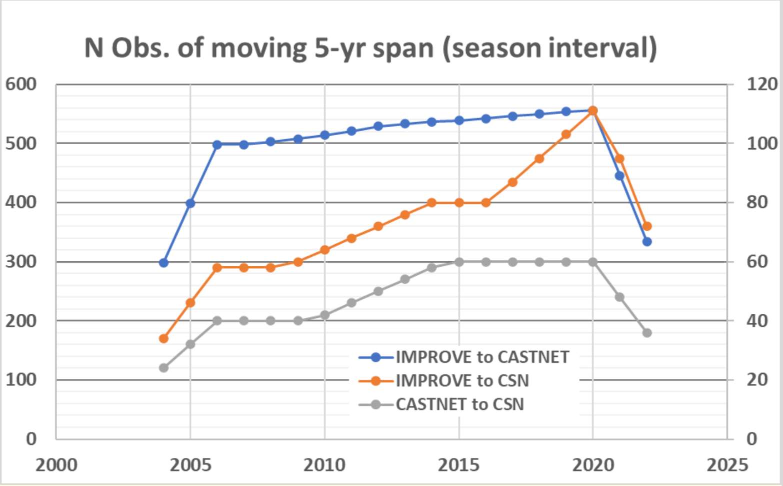
Derivation of Adjustment Equation using Seasonal Interval: Annual trends in moving 5-year window

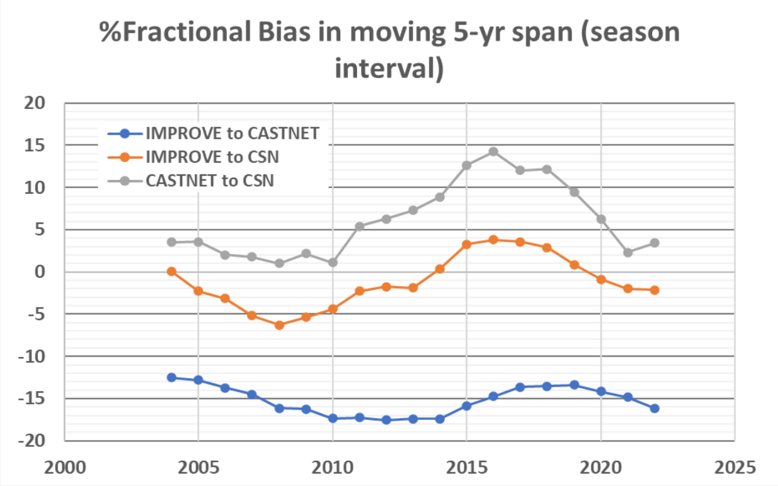
- Statistics were calculated for each year using a moving 5-year data window (e.g. 2007 OLS statistics calculated using 2005 to 2009)
  - Preserves trends, increases the number of site-pairs, smooths fluctuations





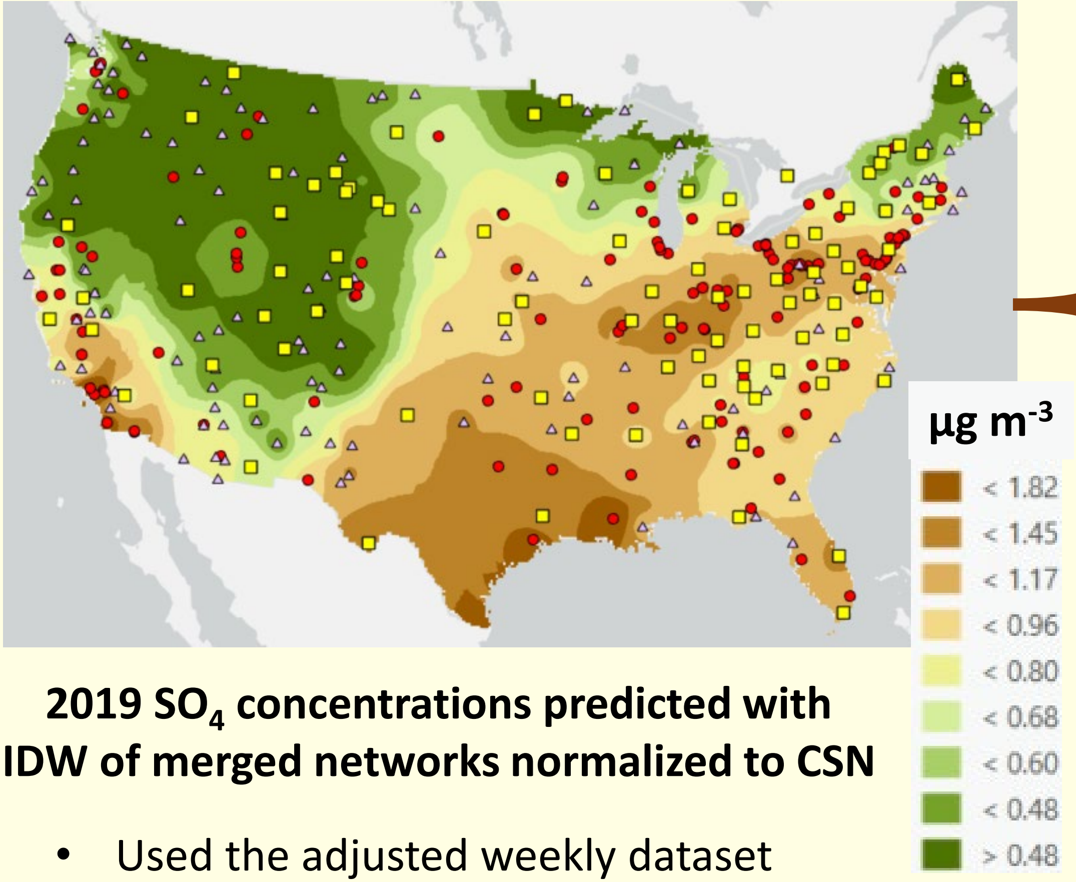






- Multiply the annual moving 5-year OLS slope and intercept to weekly data based on each year to create a new adjusted dataset
  - Normalized to CSN here, but can base off of CASTNET or IMPROVE depending on need

Utilize adjusted datasets to Create SO_4 Concentration Maps: e.g. Summer 2019

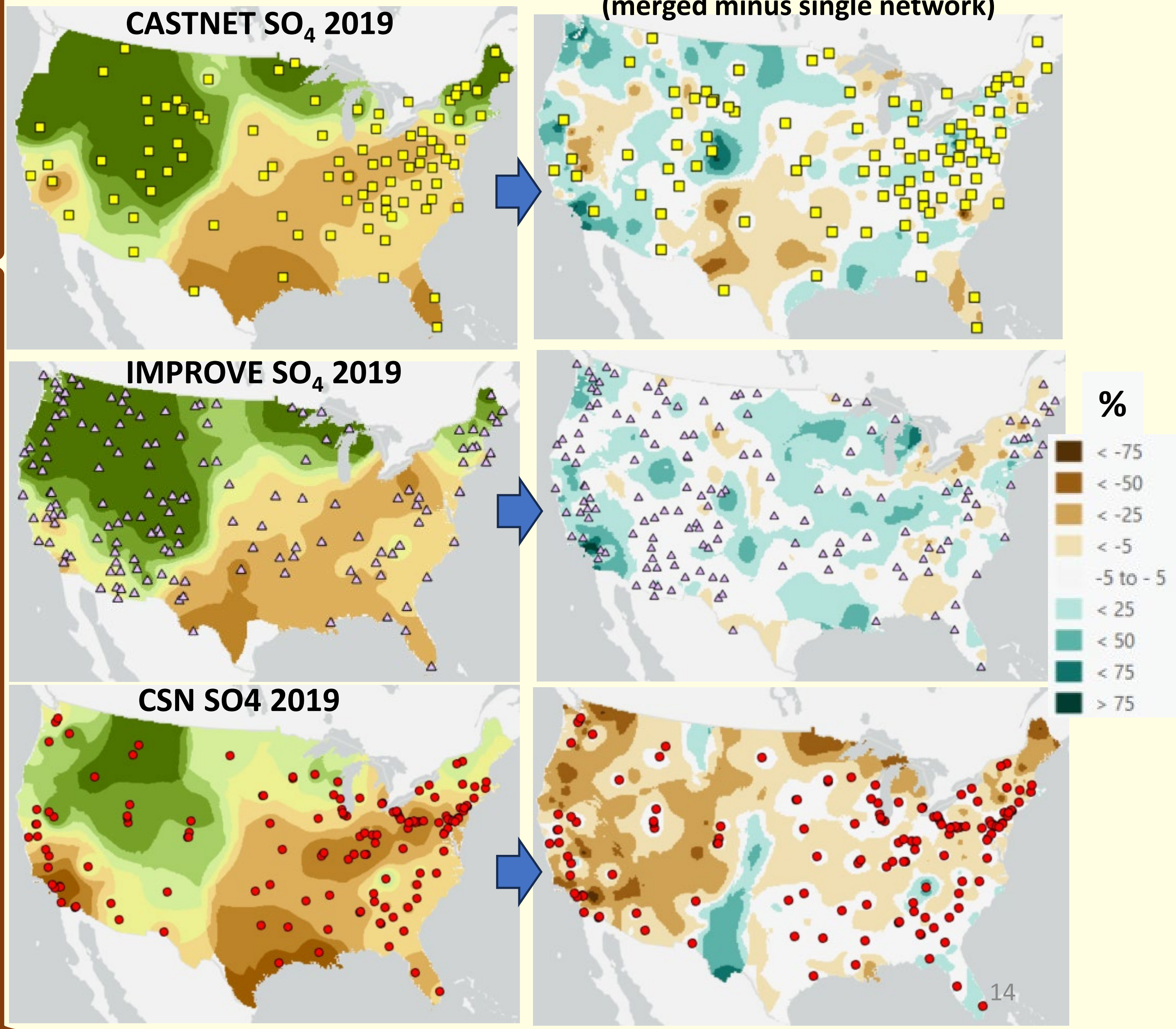


2019 SO_4 concentrations predicted with IDW of merged networks normalized to CSN

- Used the adjusted weekly dataset normalized to CSN
- Aggregated to seasonal level (as example)
  - Could aggregate to any interval
- Created maps with IDW data fusion using all adjusted networks

Single network maps:

Percent Difference Maps: (merged minus single network)



NOTE: Seasonal IDW distance from Variogram of EQUATES seasonal concentrations

Conclusions

- Our results are consistent with previous comparisons
  - We found the aggregation interval decreases the variance from the differences in sampling frequency, allowing us to isolate a trend.
    - Statistical correlations were sensitive to aggregation at the weekly levels, but stable at higher intervals
    - Seasonal aggregation interval (N=13) was optimal for all the inter-network comparisons
    - Adding completeness criteria might improve statistics and stability at higher aggregation intervals
  - We found that comparisons between different networks had different statistical correlations:
    - *IMPROVE to CASTNET* comparison had a large number of site-pairs, strong correlations, and stable parameters
      - A decreasing trend was observed in correlation and slope since 2007 and persisted in all aggregation intervals
    - *IMPROVE to CSN* comparison had the highest degree of correlation with no trends observed
    - *CASTNET to CSN* comparison had the fewest number of site-pairs and the poorest correlation. This is likely due to 1 in 6 day sampling schedule of all the co-located CSN sites.
      - Decreasing trend in correlation and intercept was observed. The slope showed an apparent step-change around 2010 and possible decreasing trend afterwards.
- Decreasing annual trends in statistical correlations were observed in both comparisons that included CASTNET sites
  - lower concentrations making network sampling bias from instrumentation differences more evident?
- Extending the distance threshold of ‘co-located’ sites was not effective in isolating correlation (too much spatial variance)
- Merged adjusted datasets used for IDW SO_4 concentration map showed substantial increases in spatial resolution and significant changes in regional areas when compared with those for the lone networks

Future Work

- Extend this process for NO_3 and NH_4 which will be more challenging (volatilization, larger impacts of particle size cut differences, etc.)
  - Utilize tools (geographical analysis, EQUATES estimates, and MARGA hourly data at two co-located sites) to help identify and understand biases
- Promote more collaboration between the networks:
  - Utilize data to fill in sampling gaps and for managers to generate network optimization strategies with this complementarity in mind
    - Demonstrated benefit of collaboration with substantial increase of the spatial information on SO_4 concentrations
    - The addition of CSN and IMPROVE sites for SO_4 in TDep MMF will help to resolve its rural concentration bias
    - Determine degree with which NH_4 measurements from CASTNET and IMPROVE can be used to complement derivation estimates used by CSN
    - Brainstorm additional uses for combining data from the multiple networks
  - Tool for managers to generate network optimization strategies with this complementarity in mind
    - Identify areas where networks are complimentary and where they might be redundant

Sources

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Appendix

- ***CASTNET:*** Weekly integrated (Tues to Tues) concentrations collected at 10 m on filter (flow ranges from 1.5 to 3 L min^{-1}) and analyzed with IC. No sampling cut-point device, but particle size limited by sampling flow mechanics of sampling apparatus.
- ***CSN:*** Concentrations collected on 'MetOne SASS' nylon filter at 6.7 L min^{-1} with cyclone $PM_{2.5}$ size cut and determined with IC. Integrated exposure for 24 hr period every 3 or 6 days (~52% and 44% at all sites since 2002).
- ***IMPROVE:*** Concentrations collected on 'IMPROVE Module B' on nylon filter at 3 m at 22.8 L min^{-1} with cyclone $PM_{2.5}$ size cut and determined with IC. Integrated exposure for 24 hr period every 3 days (97% since 2001).

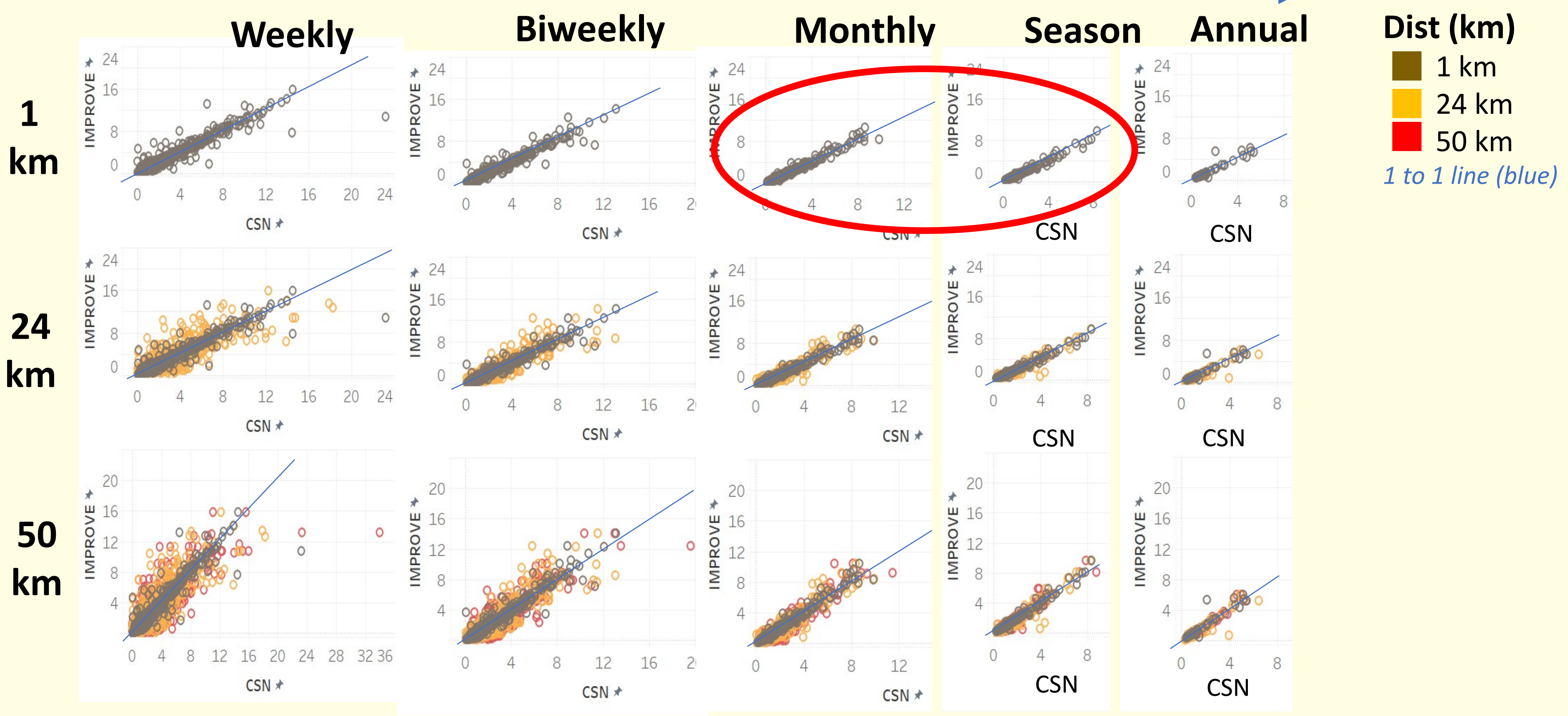
Sampling differences have been observed to be minor for sulfate (particle size) be less impactful

Nitrate has been shown to have higher and tends to suffer from more sampling artifacts including denuder inefficiency and volatilization from collection filters.

Determine the correlations: Comparisons for IMPROVE to CSN

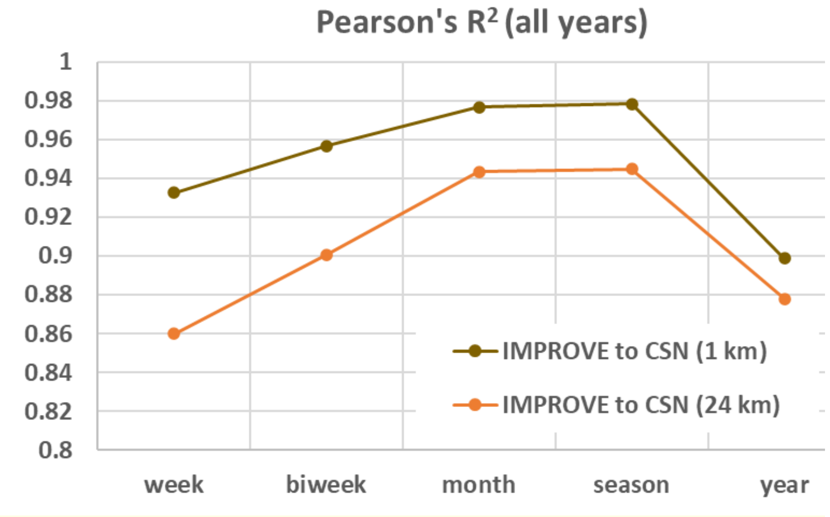
- Increasing aggregation interval length
- Decreasing outliers

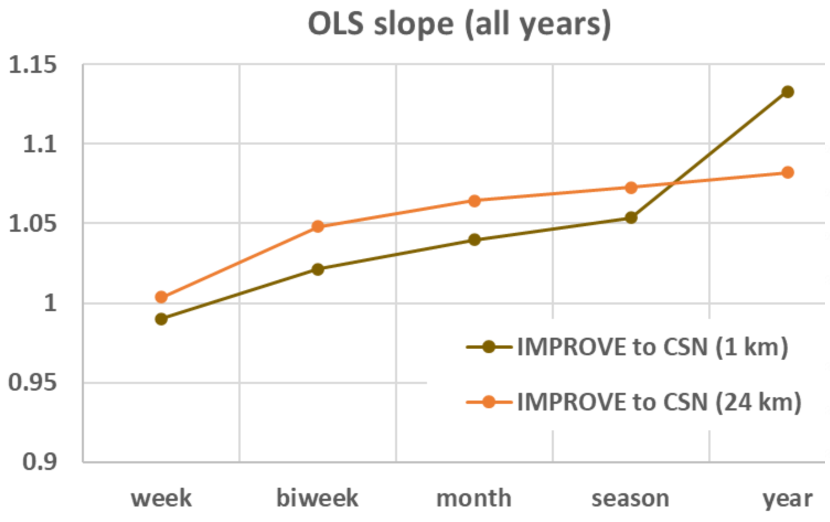
- Increasing co-location site distance
- Increasing N
- Increasing spatial variability and outliers

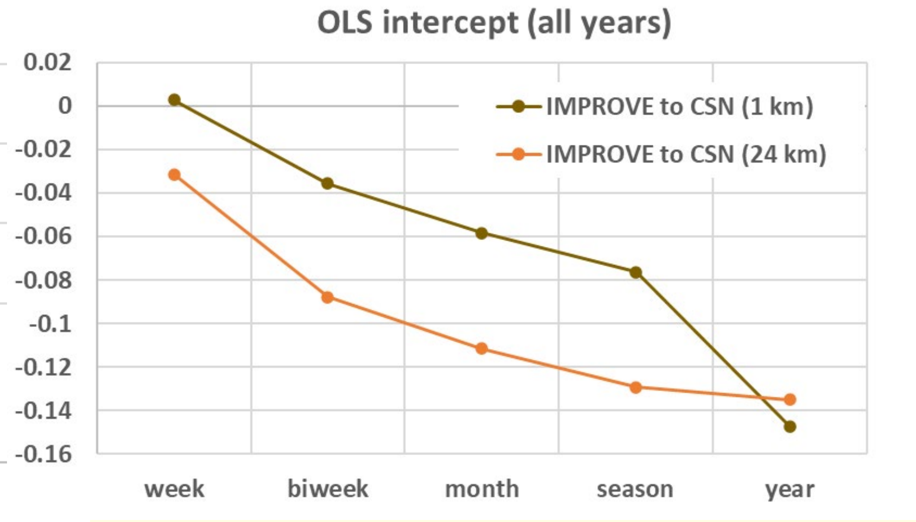


- Noticeably tighter correlations at smaller intervals (weekly interval will be averaged daily values)
- Increasing the co-location site distance threshold adds noticeable spatial variability and should not be done.
- Visually appears that ***Month or Season*** is a good aggregation interval for correlation

Comparisons for IMPROVE to CSN: Statistical measures



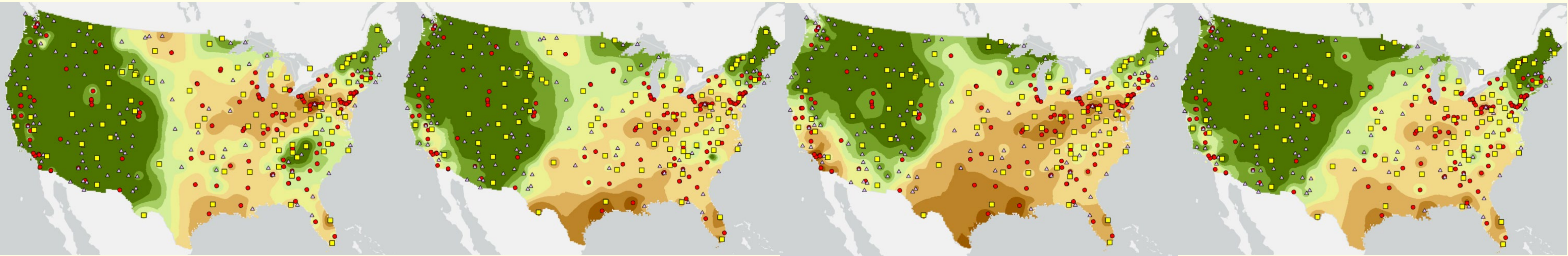


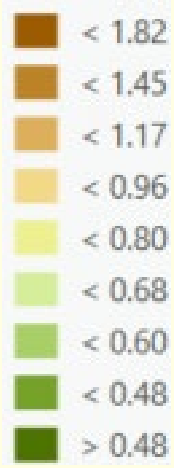


- Correlation highest for monthly and seasonal intervals. OLS parameters also stable at these intervals
- Statistics affirm that **monthly or seasonal intervals are optimal aggregation period**, will choose seasonal for consistency

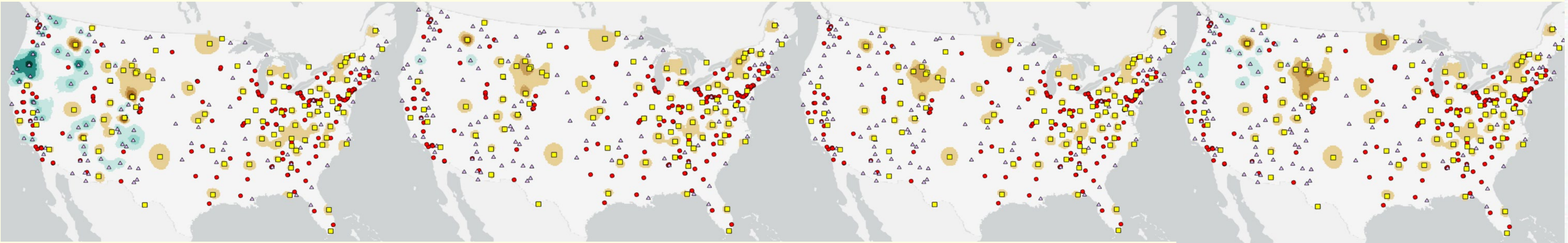
Maps: Impact of the adjustment

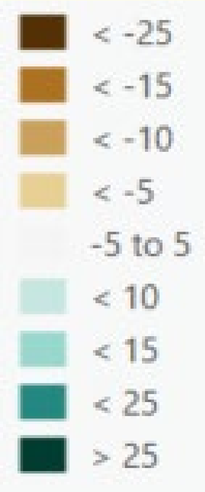
Winter | Spring | Summer | Fall





Seasonal merged adjusted SO_4 surfaces for 2019





Percent Difference (merged adjusted SO_4 minus merged observed SO_4)

Impact of the adjustment to the dataset is smaller (more localized around sites and within a ±25% scale) than the impact of the adjustment plus the merged sites shown in previous slide (regional changes and within a ±75% scale)