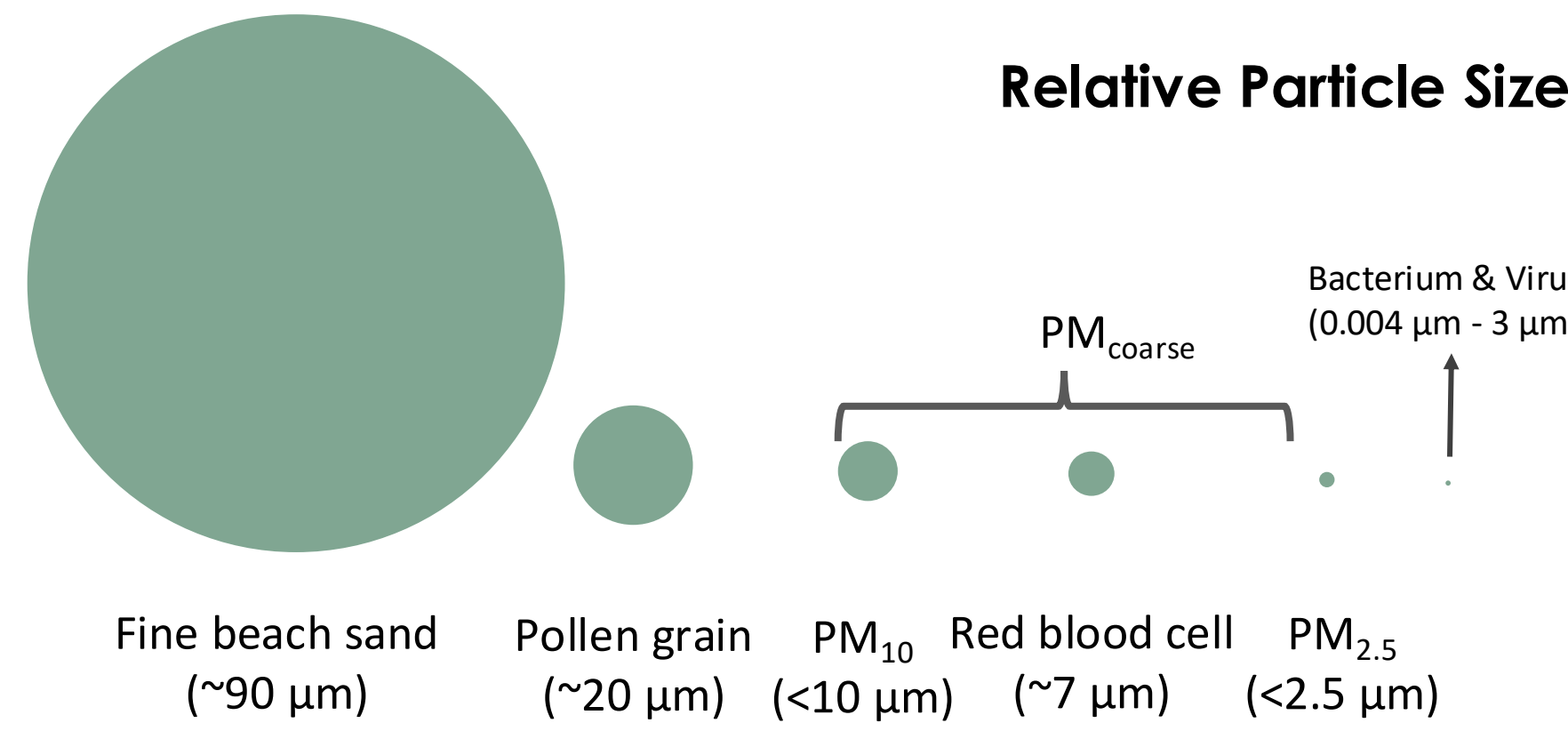
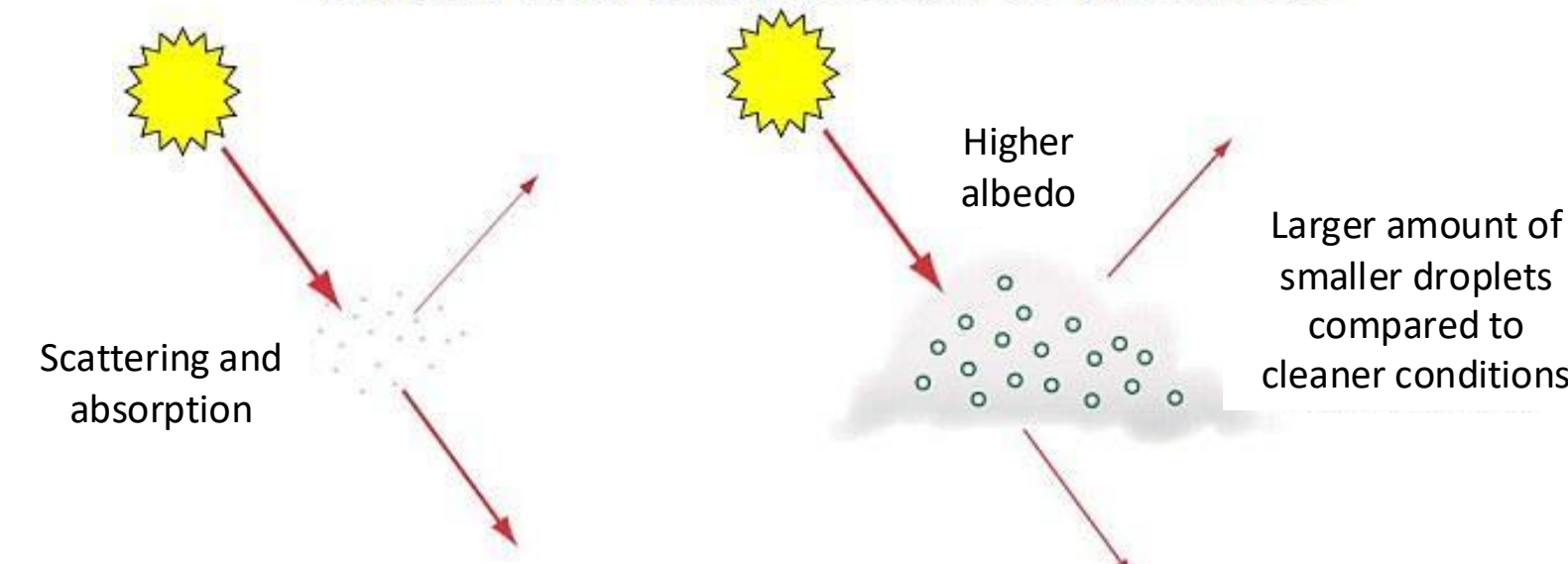


## 1) Anthropogenic activities emit dust that affects air quality and climate



### Direct and indirect aerosol effects



- Anthropogenic dust can contribute to PM<sub>2.5</sub> and PM<sub>10</sub>, both are regulated air pollutants in the U.S. that can cause respiratory diseases. It can change the climate through direct and indirect radiative effects.
- PM<sub>coarse</sub> represents the particles larger than 2.5 μm but smaller than 10 μm, which is mostly dust and has been understudied.

## 2) The GEOS-Chem model significantly underestimates PM<sub>coarse</sub> in the contiguous U.S.

- We use the GEOS-Chem chemical transport model (v14.4.1) to simulate ambient PM<sub>coarse</sub> with a spatial resolution of 0.25°. We use the anthropogenic dust emission from National Emission Inventory (NEI) provided by U.S. EPA. The natural dust emissions are simulated using a process-based scheme from Leung et al. (2023). The study period is 2019.
- Filter based PM<sub>2.5</sub> and PM<sub>10</sub> concentrations are used as the ground truth.
- We found PM<sub>coarse</sub> (PM<sub>10</sub> - PM<sub>2.5</sub>) to be greatly underestimated by GEOS-Chem across the contiguous U.S. (Figure 1), despite a good agreement in PM<sub>2.5</sub> (slope = 1.15, R<sup>2</sup> = 0.45, mean bias = +1.4 μg m<sup>-3</sup>).
- We found the low bias is persistent throughout the year (Figure 2)

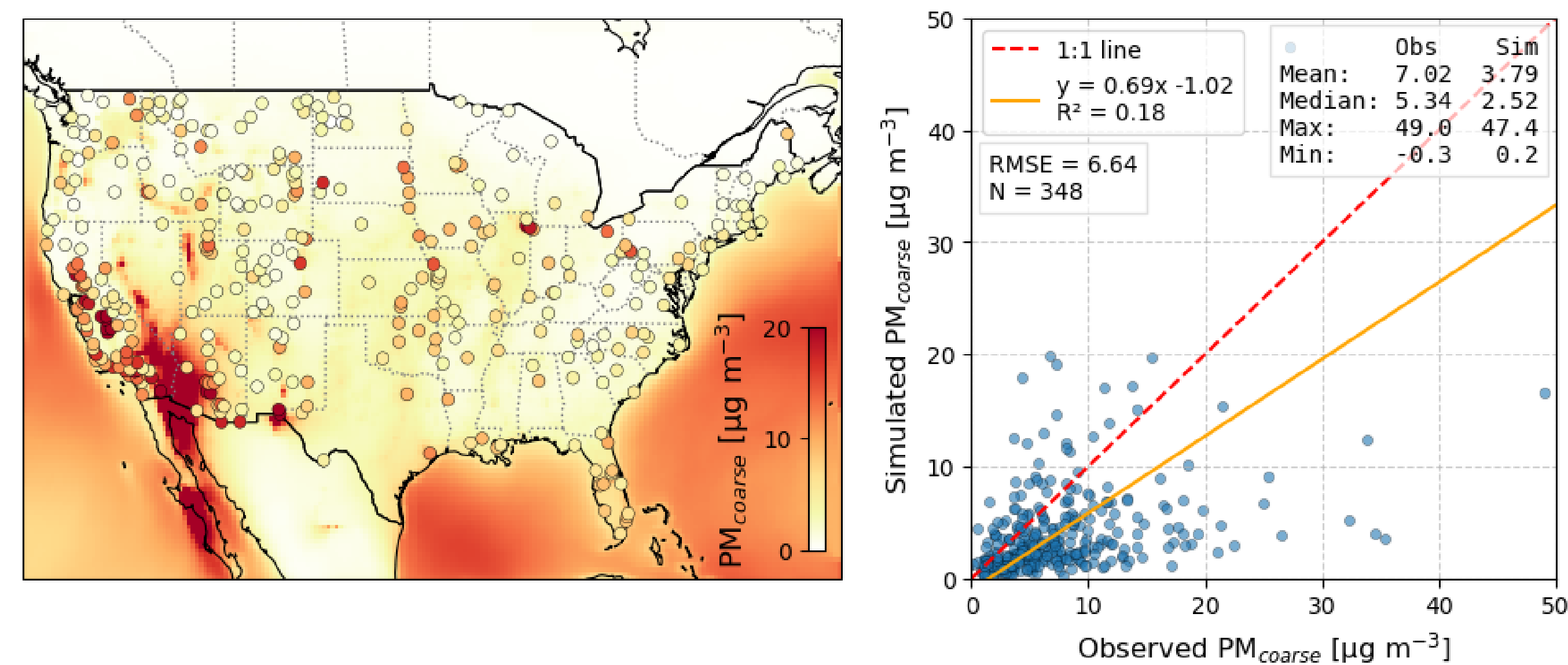


Figure 1. (left) PM<sub>coarse</sub> across the CONUS for 2019, observed by EPA monitors (dots) and estimated by the GEOS-Chem simulation using the NEI for anthropogenic dust emissions and Leung et al. (2023) for natural dust emissions (background). (right) Scatter plot comparing the two datasets.

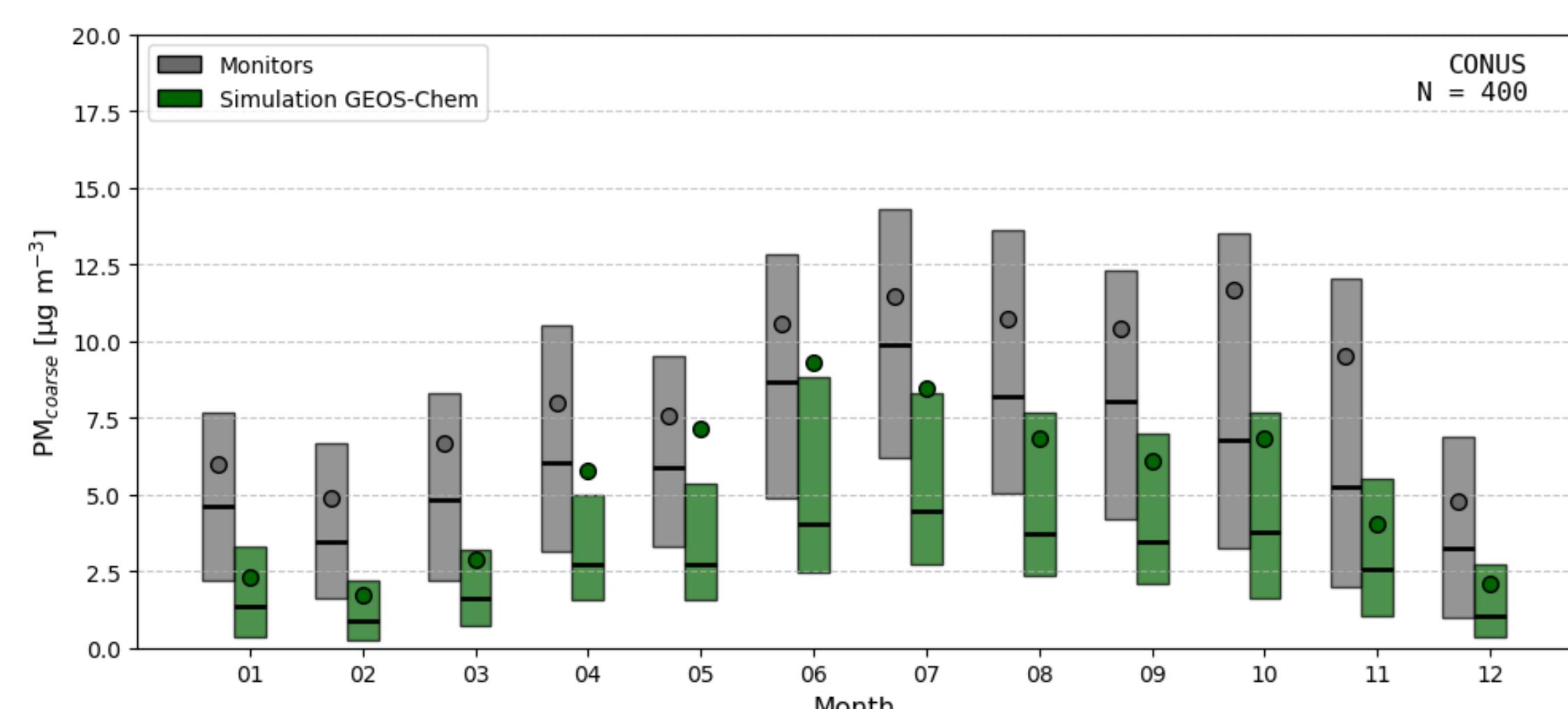


Figure 2. Seasonality of observed (grey) and simulated (green) PM<sub>coarse</sub> across the contiguous U.S. The upper and lower limits represent the 25th and 75th percentiles of the data within each bin, while the center lines indicate the median values and the dots represent the mean values.

## 3) The biases show strong anthropogenic features

- We found that bias increases with population density (figure 3), crop land fraction, and fallow land fraction (figure 4). Fallow lands are arable land intentionally left unseeded to recover nutrients, store moisture, and for other purposes. They are more strongly associated with dust emission bias (figure 4).

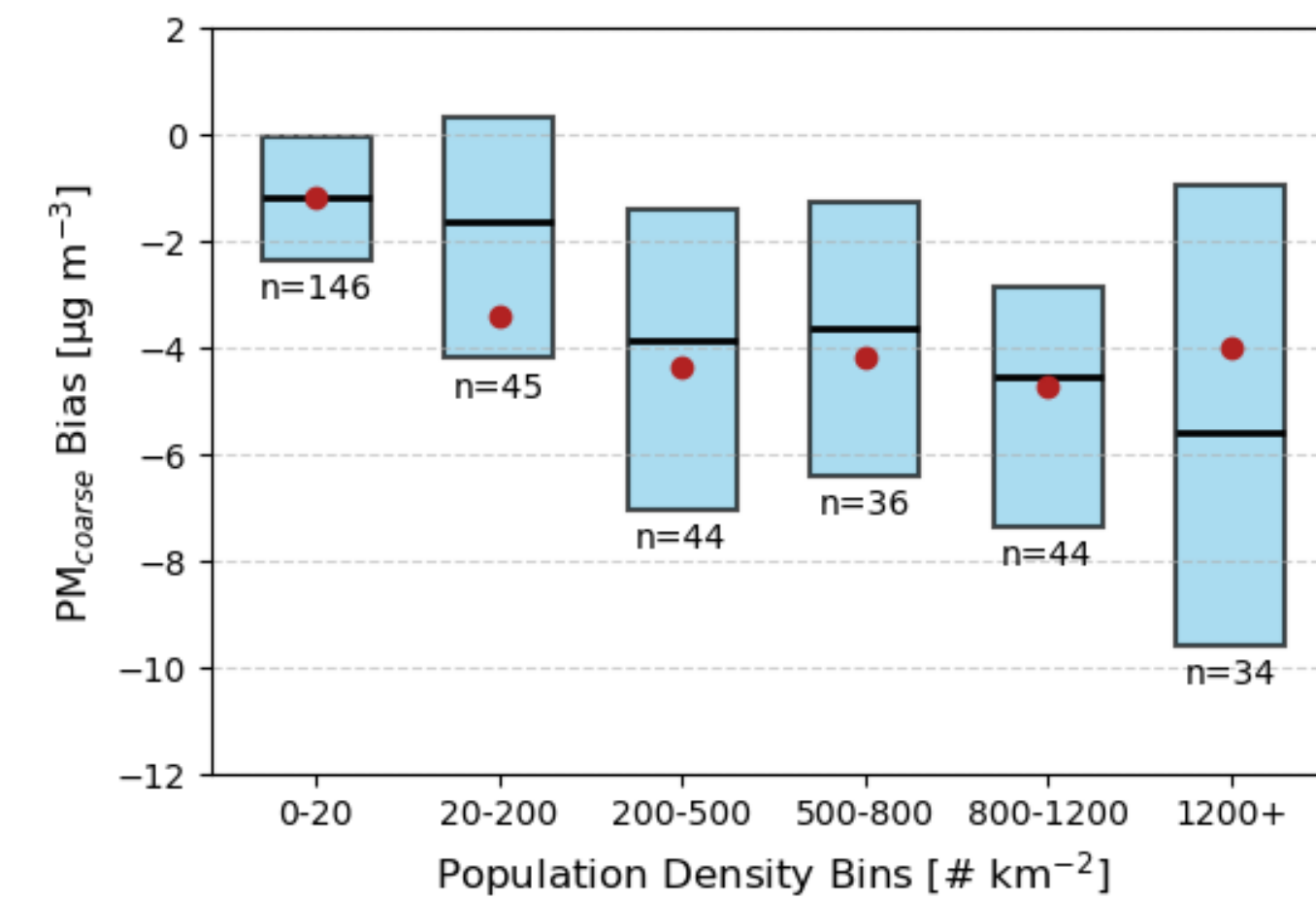


Figure 3. The annual mean model-observation bias in PM<sub>coarse</sub> is shown as a function of the population density within a 9 km × 9 km box around an observation site. The upper and lower limits represent the 25<sup>th</sup> and 75<sup>th</sup> percentiles of the data within each bin, the center lines indicate the median values, and the red dots represent the mean values.

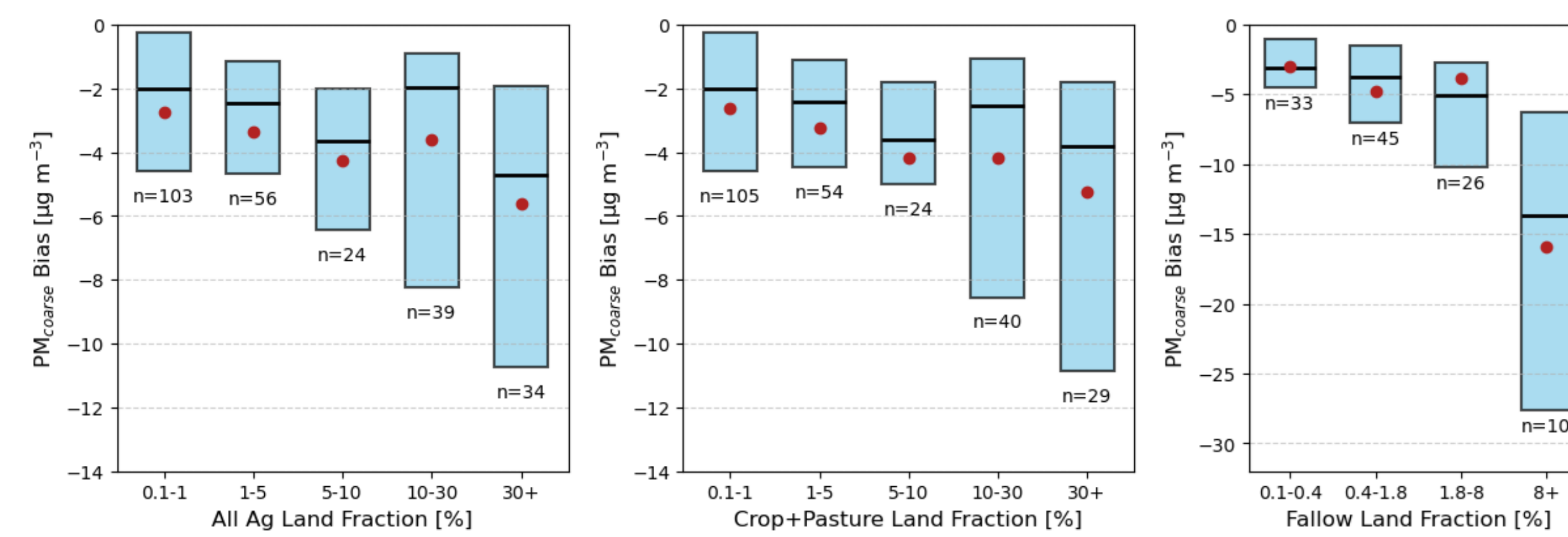


Figure 4. The annual mean model-observation bias in PM<sub>coarse</sub> is shown as a function of the fraction of agricultural lands (crop+pasture+fallow) within a 9 km × 9 km box around an observation site for 2019. The upper and lower limits represent the 25<sup>th</sup> and 75<sup>th</sup> percentiles of the data within each bin, the center lines indicate the median values, and the red dots represent the mean values.

## 4) Anthropogenic dust emission is the main driver of the bias

- To examine drivers the the bias, we run sensitivity simulations by perturbing major factors that could affect PM<sub>coarse</sub> concentration. The study period is Spring 2019 (March and April).
- Factors include dry deposition rate, natural dust emission, anthropogenic dust emission from agricultural and non-agricultural sources.
- Scaling agricultural emissions by a factor of 10 improves model accuracy and precision more efficiently than other perturbations.
- Scaling non-agricultural dust emission by a factor of 5 yields 25% and 50% percentiles that are close to the observations, but the mean values and 75% percentiles are too high.
- Scaling dry deposition and natural dust emission are inefficient for model improvement.

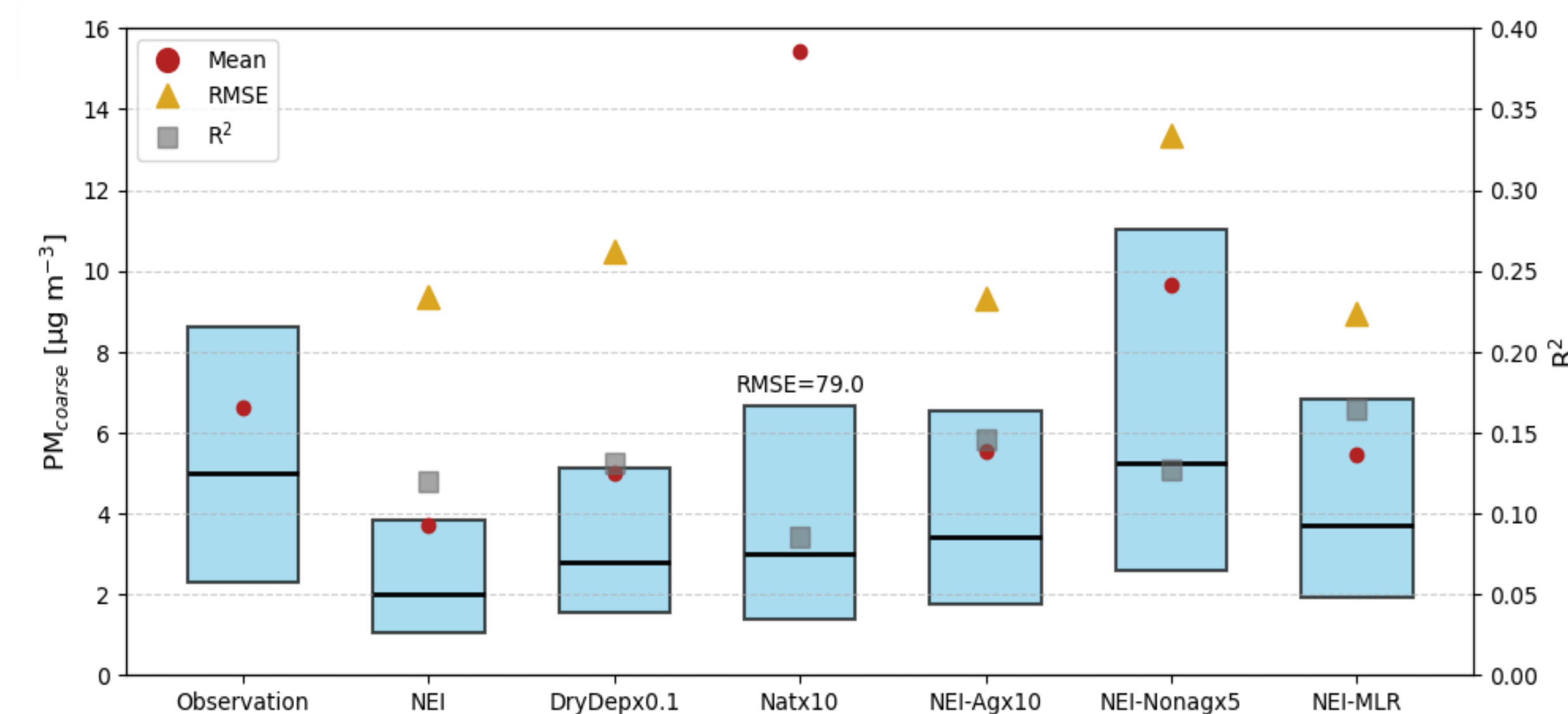


Figure 5. Box plot showing the statistical summary of PM<sub>coarse</sub> concentrations from:

- (1) Observations,
- (2) GEOS-Chem simulation with NEI,
- (3) simulation with NEI and scaled dry deposition speed by 0.1,
- (4) simulation with NEI and scaled natural dust emission by 10,
- (5) simulation with NEI and scaled agricultural dust emission by 10,
- (6) simulation with NEI and scaled non-agricultural dust emission by 5, and
- (7) simulation with NEI and scaled emission based on multiple linear regression (MLR), scaling factors are in the next section.

## 5) Regional bias estimation

- We use the following equation, which includes spatially varying predictors such as land types and population in multiple linear regression (MLR), to examine bias in each state. Coefficients are bounded so that emissions are positive.

$$PM_{coarse,adjusted} = PM_{coarse,sim} + (a + b \times F_{crop} + c \times F_{fallow}) \times \Delta PM_{coarse,ag\ emission} + (d + e \times F_{urban} + f \times P) \times \Delta PM_{coarse,non-ag\ emission}$$

- This statistical method suggests significant spatial and sectoral heterogeneity in the biases. Agricultural dust emissions along the East Coast and in the Great Plains are substantially underestimated, while non-agricultural anthropogenic dust emissions exhibit relatively less bias nationally.

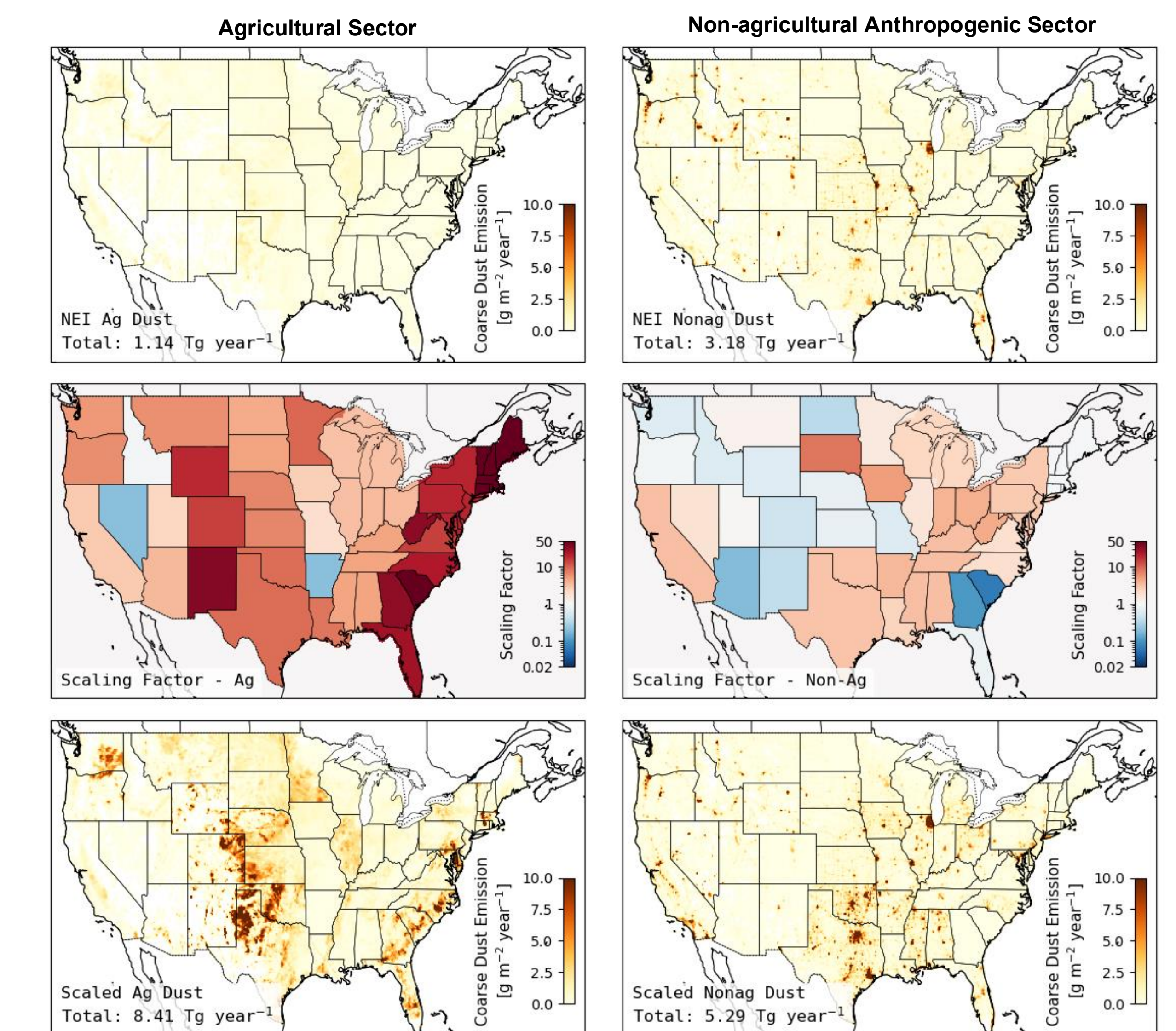


Figure 6. (Top) Maps of 2019 annual mean coarse dust emissions from the agricultural and non-agricultural anthropogenic sources. (Middle) Maps of state average scaling factors for NEI anthropogenic dust emission for the two. (Bottom) Scaled annual mean coarse dust emissions.

## 6) Conclusions and Future Works

- We found notable bias in GEOS-Chem simulated PM<sub>coarse</sub>, even when the National Emissions Inventory (NEI), the most advanced bottom-up anthropogenic dust emission inventory, is applied.
- We found that the bias is associated with crop lands, fallow lands, and population.
- Through sensitivity test and statistical analysis, we found the bias is mostly due to underestimated anthropogenic dust emissions, rather than dry deposition or natural dust emission.
- Multiple linear regression indicates that the agricultural dust emissions are substantially underestimated, particularly along the East Coast and the Great Plains. Dust emissions from other anthropogenic sectors are also being underestimated.
- Future efforts to improve simulated PM<sub>coarse</sub> could be: (1) developing a process-based agricultural dust emission scheme; (2) advancing satellite remote sensing techniques for dust; and (3) including additional ground-based measurements, such as aerosol concentration, emission flux, and particle size distribution. These improvements will help reduce model uncertainties and reconcile the discrepancies identified in this study.

## Acknowledgements and References

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