Supplementary Material

1. Daily Aggregation of IVAN Measurements

In this study, soft criteria were adopted for the aggregation of hourly IVAN measurements into daily data. Specifically, all available hourly measurements were used for aggregation without concerning the hourly coverage. IVAN is a well-maintained low-cost sensor network. Before the daily aggregation, the completeness and continuity of IVAN hourly measurements were examined. We found that the sensors had very high hourly coverage during the study period. The mean coverage of each site in each day was > 22 hours with an interquartile range of 0 (median: 24 hours; 25% and 75% percentiles: 24 hours). Only < 3% of the sensor-days (the total days during the study period multiplies the number of sensors) had the hourly coverage < 5 hours. Although this small number of daily measurements aggregated from < 5 hourly measurements might bias the predictions at the daily level, this uncertainty may have limited influence on our major conclusions which were mainly based on the overall prediction pattern during the study period.

2. AOD Missingness and Gap-Filling

Satellite AOD has a large proportion of missing retrievals over land due to cloud cover and high surface brightness (*e.g.,* standing water). Missing AOD retrievals have significantly hindered the generation of fully-covered $PM_{2.5}$ estimates. Recent studies overcome this obstacle by estimating missing AOD data before predicting $PM_{2.5}$, a process called AOD gap-filling (Bi et al. 2019; Li et al. 2012; Van Donkelaar et al. 2011; Xiao et al. 2017). In this study, we followed the approach proposed by Bi et al. (2019) to conduct AOD gap-filling, in which random forest models with AOD-related predictors were established at a daily level. The predictors consisted of 1) cloud fraction, 2) 2-meter specific humidity, 3) 2-meter temperature, 4) planetary boundary layer height, 5) surface wind speed, and 6) spatial coordinates of AOD retrievals. The AOD gap-filling was separately conducted for Terra and Aqua datasets. A three-day moving time window was applied to increase the sample size (Xiao et al. 2017). As in Bi et al. (2019), the random forest-specific out-of-bag (OOB) R^2 and root-mean-square error (RMSE) were used to evaluate the performance of AOD gap-filling.

During the study period (September 2016 to November 2017), Terra AOD had a mean daily missing rate of 36.9% with an interquartile range (IQR) from 6.3% to 69.3%. Aqua AOD had a mean daily missing rate of 40.8% with an IQR from 8.0% to 76.3%. Figure S4 shows the patterns of missing AOD retrievals of Terra and Aqua during the study period. The grid cells over water bodies (*e.g.,* the Salton Sea) were filtered out (Chu et al. 2002). Besides water bodies, missing AOD was evenly distributed throughout the study domain with minor increases in the regions covered by dense vegetation. The gap-filling models had good performance with a mean OOB R^2 of 0.95 and a mean RMSE of 0.013 for both Terra and Aqua datasets. As in Xiao et al. (2017) and Bi et al. (2019), we also found that the gap-filled AOD values tended to be higher than the original AOD retrieved under clear-sky condition. Xiao et al. (2017) and Bi et al. (2019) suggested that this pattern may be caused by aerosol hygroscopic growth.

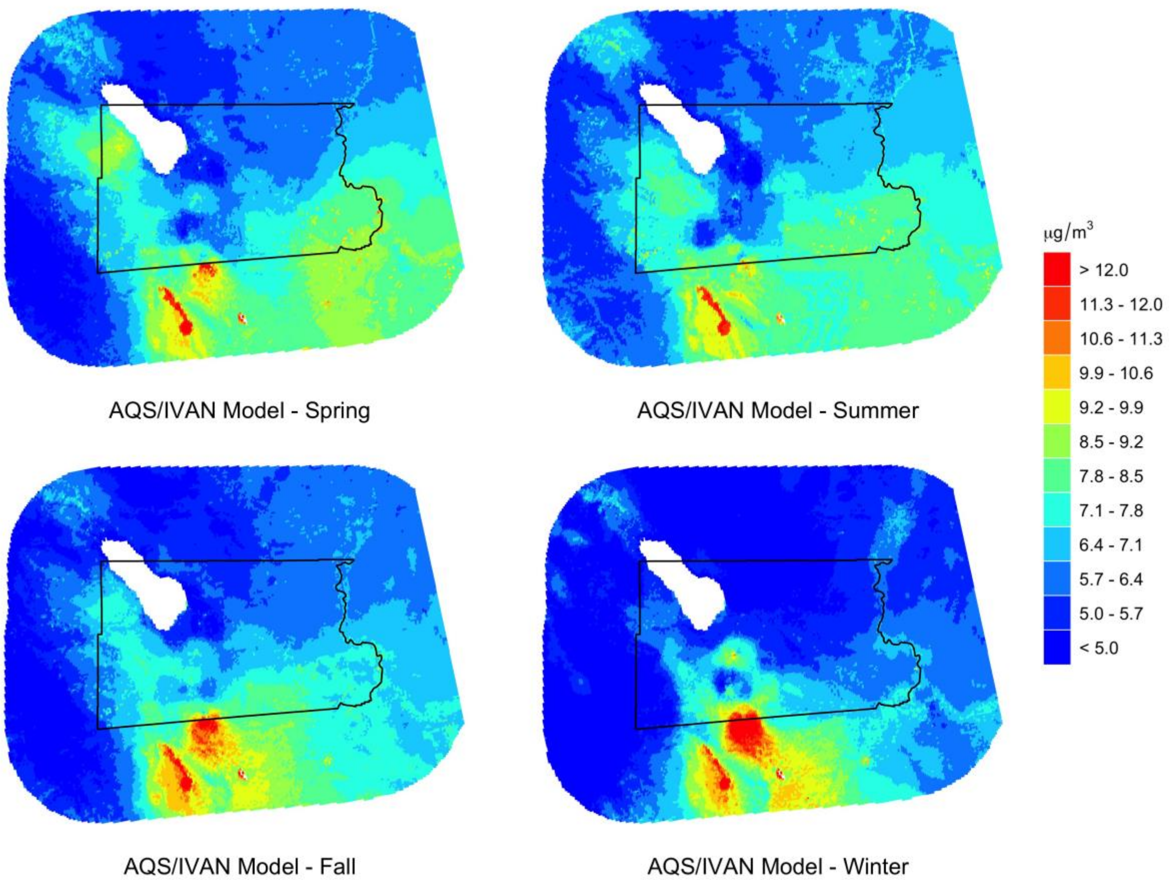

Figure S1 Mean $PM_{2.5}$ distributions in different seasons generated by the AQS/IVAN model during the study period.

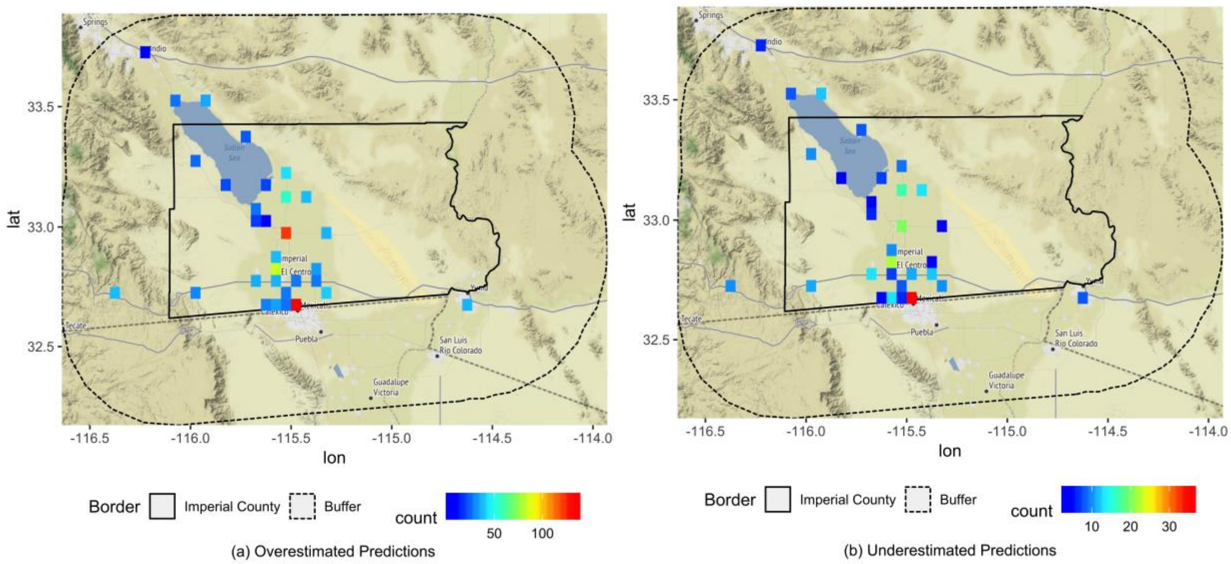

Figure S2 Frequencies of prediction outliers at the locations of AQS and IVAN stations (left: overestimated predictions; right: underestimated predictions).

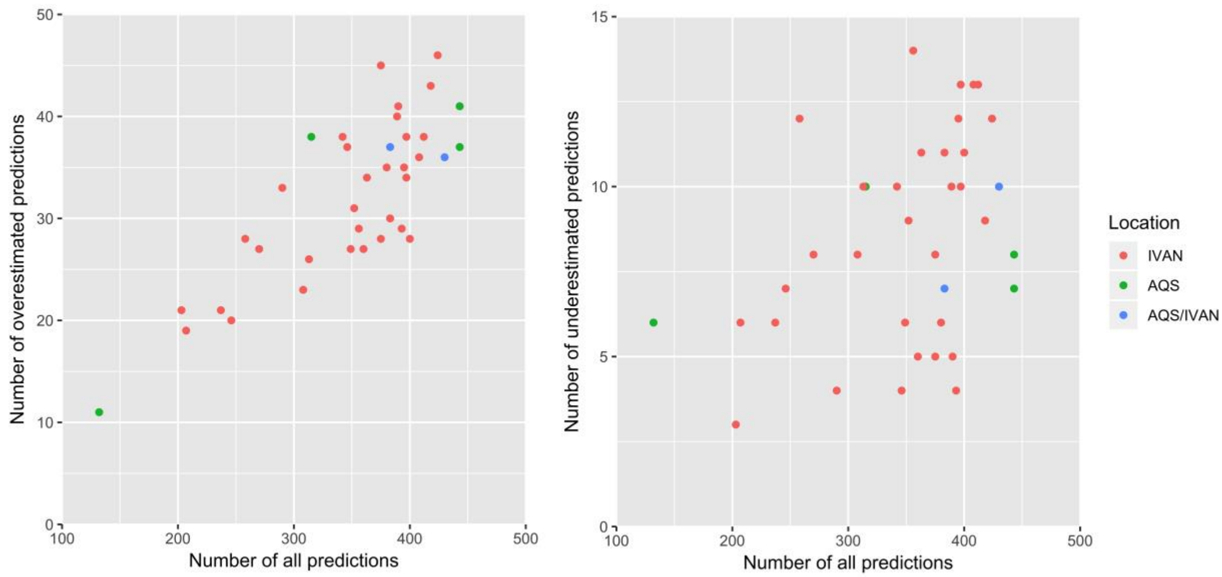

Figure S3 Correlations between the number of outliers and the number of predictions (left: overestimated predictions; right: underestimated predictions).

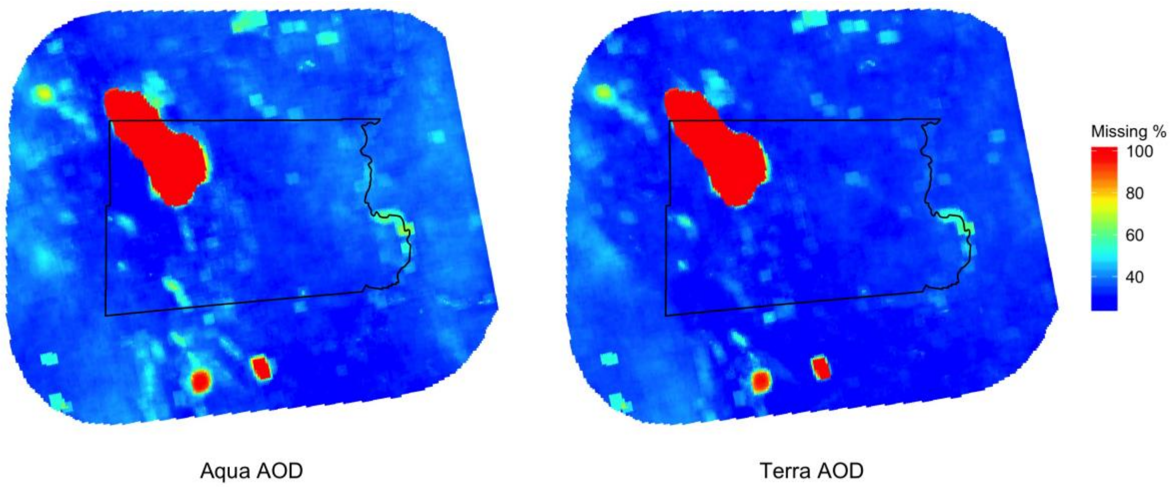

Figure S4 Distributions of the average missing rates of Terra/Aqua AOD during the study period.

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