A continuous 2011-2022 record of fine particulate matter (PM_{2.5}) in East Asia at daily 2-km resolution from geostationary satellite observations: population exposure and long-term trends

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Abstract. We construct a continuous 24-h daily fine particulate matter $(PM_{2.5})$ record with $2 \times 2 \text{ km}^2$ resolution over eastern China, South Korea, and Japan for 2011-2022 by applying a random forest (RF)

- 20 algorithm to aerosol optical depth (AOD) observations from the Geostationary Ocean Color Imager (GOCI) I and II satellite instruments. The RF uses PM_{2.5} observations from the national surface networks as training data. PM_{2.5} network data starting in 2015 in South Korea are extended to pre-2015 with a RF trained on other air quality data available from the network including PM₁₀. PM_{2.5} network data starting in 2014 in China are supplemented by pre-2014 data from the US embassy and consulates.
- 25 Missing AODs in the GOCI data are gap-filled by a separate RF fit. We show that the resulting GOCI PM_{2.5} dataset is successful in reproducing the surface network observations including extreme events, and that the network data in the different countries are representative of population-weighted exposure. We find that PM_{2.5} peaked in 2014 (China) and 2013 (South Korea, Japan), and has been decreasing steadily since with no region left behind. We quantify the population in each country exposed to annual
- 30 PM_{2.5} in excess of national ambient air quality standards and how this exposure evolves with time. The long record for the Seoul Metropolitan Area (SMA) shows a steady decrease from 2013 to 2022 that was not present in the first five years of AirKorea network PM_{2.5} measurements. Mapping of an extreme pollution event in Seoul with GOCI PM_{2.5} shows a predicted distribution indistinguishable from the dense urban network observations, while our previous 6×6 km² product smoothed local features. Our
- 35 product should be useful for public health studies where long-term spatial continuity of PM_{2.5} information is essential.

1. Introduction

Outdoor fine particulate matter ($PM_{2.5}$, less than 2.5 μ m in diameter) is a leading cause of morbidity and mortality, with exposure leading to 8.9 million deaths worldwide in 2015 and increased diagnoses of

- 40 respiratory, cardiovascular, and neurodegenerative diseases (Dominici et. al., 2006; Kioumourtzoglou et. al., 2016; Burnett et. al., 2018 Wei et. al., 2019). East Asian countries experience particularly high concentrations of PM_{2.5}. China introduced its Action Plan on Prevention and Control of Air Pollution in 2013 (Chinese State Council, 2013) and South Korea introduced its Comprehensive Action Plan on Fine Dust in 2017 (Joo, 2018). As a result, concentrations declined in the latter half of the 2010s
- 45 (Pendergrass et al., 2022). However, the publicly available archive of PM_{2.5} measurements from national surface networks only started in 2014 in China and in 2015 in South Korea, and even today they are relatively sparse for public health applications. Here we use geostationary satellite observations of aerosol optical depth (AOD) from the Geostationary Ocean Color Imager (GOCI) I and II instruments, trained with surface PM_{2.5} data using a machine learning algorithm, to provide complete
- 50 2011-2022 daily 24-h coverage of surface $PM_{2.5}$ concentrations at $2 \times 2 \text{ km}^2$ resolution for eastern China, South Korea, and Japan.

Satellite retrievals of AOD from backscattered solar radiation have long been used to expand surface PM_{2.5} coverage beyond that provided by network sites. Early applications used AOD/PM_{2.5} ratios computed with a chemical transport model (CTM) to infer surface PM_{2.5} from observed AOD

- 55 (Liu et al., 2004; van Donkelaar et al., 2006; van Donkelaar et al., 2021) but this may be affected by CTM biases. More recent applications have used machine learning algorithms to train satellite AODs on PM_{2.5} network measurements (Guo et al., 2021; Pendergrass et al., 2022; Wongnakae et al., 2023). Commonly used machine learning algorithms include XGBoost and Random Forest (RF), both based on decision trees, and neural networks; precision tends to be similar across algorithms (Di et al., 2019;
- 60 Kulkarni et al., 2022). RF approaches are widely used due to their explainability and consistently strong performance with minimal hyperparameter tuning (Breiman, 2001).

In East Asia, studies inferring PM_{2.5} from satellite AOD data have benefited from new geostationary instruments including GOCI (launched 2010), the Advanced Himawari Imager (AHI, launched 2014), the Advanced Meteorological Imager (AMI, launched 2018), GOCI-II and GEMS

- 65 (launched in 2020), which provide continuous hourly or subhourly measurements during daytime (Choi et al., 2018; Lim et al., 2018; Lee et al., 2023; Kim et al., 2023; Cho et al., 2023a). The RF method has been used to infer hourly PM_{2.5} from geostationary AOD (Liu et al., 2022; Tan et al., 2023; Cho et al., 2023b), but geostationary AOD also improves inference of 24-h mean PM_{2.5}; Park et al. (2019) found that an RF algorithm trained to predict PM_{2.5} from GOCI AOD outperformed an otherwise identical one
- 70 trained on the MODIS low-earth orbit instrument. Our previous work (Pendergrass et al., 2022) used GOCI I observations to produce a continuous 24-h 6×6 km² PM_{2.5} product for eastern China, South Korea and Japan for the network observation periods (starting in 2014 in China and 2015 in South Korea) and extending to 2019.

Here we use a continuous, gap-filled record of AOD retrieved from GOCI I and its successor
GOCI II on a consistent 2×2 km² grid to infer surface PM_{2.5} at 24-h temporal resolution from March
2011 through the end of 2022 for eastern China, Japan, and South Korea. We make use of an improved
AOD gap-filling procedure by using a separate RF fit trained to reproduce AOD data. To provide

continuity in training across the study domain from 2011 to present, we additionally develop and

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evaluate a virtual network PM_{2.5} record prior to 2015 in South Korea by training an RF on network observations of coarse particulate matter (PM_{10}) and other air pollutants. In China, we make use of US embassy and consulate PM_{2.5} data to train the RF before 2014. We use the resulting GOCI PM_{2.5} dataset to analyze urban and regional trends in PM_{2.5} including population exposures.

2. Methods

- Pendergrass et al. (2022) used GOCI I AOD observations to produce a continuous 24-h 6×6 km² PM_{2.5} product for eastern China, South Korea and Japan for the surface network observation periods 85 (starting in 2014 in China, 2015 in South Korea, and 2011 in Japan) and extending to the end of 2019. It gap-filled missing GOCI I AOD data by blending a CTM simulation with statistical interpolation (inverse distance weighted means).
- Here we improve on Pendergrass et al. (2022) in several major ways. First, we extend the AOD record using the GOCI II instrument to cover the 2011-2022 period, and replace the 6×6 km² GOCI I 90 AOD with a $2 \times 2 \text{ km}^2$ GOCI I AOD retrieval (section 2.1). We replace the statistical AOD gap-filling method of Pendergrass et al. (2022) with an additional AOD RF fit (section 2.2). To avoid biased PM_{2.5} estimates in South Korea prior to the beginning of the AirKorea national network observations in 2015, we use an additional RF to infer surface observations of PM_{25} in South Korea at the network sites using
- measurements of other air quality variables including PM₁₀ starting from 2011 (section 2.3). In China, 95 we avoid extrapolation bias by supplementing surface network data with data from the US embassy and consulates, which date from 2011. Finally, we train an RF on the gap-filled AOD and other predictor variables to construct a continuous 24-h 2×2 km² PM₂ 5 product for China. South Korea, and Japan covering the 2011-2022 period (section 2.4). Table 1 lists the predictor variables for all of the RFs used
- 100 in this work.

We evaluate how each RF performs and learns via a 10-fold crossvalidation procedure and Shapley analysis. The crossvalidation measures how well an RF can make predictions based on more limited training data. For each fold of the crossvalidation, we leave out a randomly selected 10% of sites entirely from training; in this way, the crossvalidation measures the ability of the RF to generalize

- 105 spatially to unseen sites. We compare RF-predicted AOD and PM_{2.5} (24-h and annual) to the withheld observed AOD and PM2.5 using four metrics: the root mean square error (RMSE); the RMSE divided by mean observed value (relative RMSE, or RRMSE); the coefficient of determination (R²); and the mean bias computed by averaging the difference between predicted and observed values (MB). To determine the contributions of training variables to the overall RF estimate, we use the SHapley Additive
- exPlanations (SHAP) analysis as implemented by the TreeExplainer algorithm (Lundberg et al., 2020). 110 This method allocates a SHAP value, in the same unit as the target variable ($\mu g m^{-3}$ for PM_{2.5}, unitless for AOD), to each predictor variable and can be interpreted as the importance of that variable to the trained RF algorithm. All RFs are produced using the Python module scikit-learn (Pedregosa et. al., 2011).
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Table 1. Random Forest predictor variables^a

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GOCI (gap-filled) and GEOS-Chem
   GOCI I AOD 8-h average (0:30-7:30 UTC) at 550 nm wavelength (2011-2020)
   GOCI II AOD 10-h average (23:15-8:15 UTC) at 550 nm wavelength (2021-2022)
    Gaspari-Cohn missingness factor \alpha^{b}
    Bias-corrected GEOS-Chem monthly mean AOD<sup>c</sup>
Meteorology<sup>d</sup>
    Boundary layer height (m)<sup>†</sup>
    10-m meridional wind (m s^{-1})^*
    10-m zonal wind (m s^{-1})^*
    2-m temperature (K)*
   2-m relative humidity<sup>e</sup> (%)*
   Sea-level pressure (Pa)<sup>†</sup>
KORUSv5 emissions<sup>f</sup>
   NO_x (molec m<sup>-2</sup> s<sup>-1</sup>)
   SO_2 (molec m<sup>-2</sup> s<sup>-1</sup>)
   NH_3 (molec m<sup>-2</sup> s<sup>-1</sup>)
Land use
   Land cover type (cropland, urban, rural)<sup>g</sup>
   Population density<sup>h</sup>
   Elevation<sup>i</sup>
   Normalized Difference Vegetation Index (NDVI)<sup>j</sup>
Metadata
   Country categorical variables<sup>k</sup>
   Day of year
   Year
AirKorea surface air quality data<sup>1</sup>
   CO (ppm)
   NO<sub>2</sub> (ppm)
   O_3 (ppm)
   SO_2 (ppm)
   PM<sub>10</sub> (µg m<sup>-3</sup>)
    Yellow dust categorical variable (T/F)
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^bWeighting factor with a value of 1 if AOD is retrieved successfully at least once in a given day in a given 2×2 km² grid cell and descending to 0 as distance to the nearest successful retrieval increases. Not used to in the GOCI AOD gap-filling RF. See section 2.2.

^cSimulation from Zhai et al. (2021) at 0.5° × 0.625° resolution and corrected to annual mean GOCI observations on the 2×2 km² grid.
 ^dMeteorological data from either the ECMWF hourly 9×9 km² resolution ERA5-Land replay of the ERA5 global reanalysis (denoted *) or hourly 30×30 km² from ERA5 (†), interpolated bilinearly to the GOCI grid and averaged over 24 hours. For coastal pixels missing from the ERA5-Land data, we impute values from ERA5.

^aThese predictor variables are used for three separate RF fits: (1) GOCI $PM_{2.5}$, (2) imputing pre-2015 $PM_{2.5}$ at AirKorea sites from PM_{10} and other predictors, and (3) gap-filling GOCI AOD. Unless otherwise noted, the data are used in all three RFs and are mapped onto the 2x2 km² GOCI grid cells.

^{125 °}Inferred from temperature and dewpoint using the August-Roche-Magnus approximation (Alduchov and Eskridge, 1996). f 2015 emissions for East Asia on a 0.1° × 0.1° grid (Woo et al., 2020).

^g Land cover data at 300 m resolution for 2015 is obtained from the from the PROBA-Vegetation (PROBA-V) and Sentinel-3 OLCI (S3 OLCI) time series (<u>https://cds.climate.copernicus.eu/cdsapp#!/dataset/satellite-land-cover?tab=overview;</u> CDS, 2019). We aggregate the data to one of three categories based on the most prevalent land cover type within a 2×2 km² GOCI grid cell; urban areas, cropland

- 130 (irrigated, rainfed, and mosaic but majority cropland), and rural (all other non-water pixels with minimal human modification). ^h2015 population density at 30 arc second resolution from the Gridded Population of the World v4.11 dataset (CIESIN, 2018) ⁱElevation from the global multi-resolution terrain elevation data 2010 digital elevation model (GMTED2010), corrected and aggregated to 0.0625° resolution by the Tropospheric Emission Monitoring Internet Service (<u>https://www.temis.nl/data/gmted2010/index.php</u>; Danielson and Gesch, 2011).
- 135 ^jDaily Normalized Difference Vegetation Index (NDVI) derived from the NOAA Climate Data Record (CDR) of Advanced Very High Resolution Radiometer (AVHRR) Surface Reflectance and reported at 0.05°×0.05° resolution (Vermote 2019). A small number of NDVI pixels are missing, which are imputed by first looking for a successful retrieval within two weeks of the day in question and if that fails by inverse distance weighting.
- ^kThree variables that, for each of eastern China, South Korea, and Japan, have value 1 if a grid cell is within those national borders and 0 otherwise.

¹Used as input in the pre-2015 AirKorea PM_{2.5} RF. Yellow dust variable is true if a dust event (due to transport from China/Mongolia) is observed at a given site that day.

2.1 GOCI, GEOS-Chem, and PM_{2.5} input datasets

GOCI I and II AOD. GOCI I was launched in 2010 by the Korea Aerospace Research Institute (KARI)
and recorded data every hour eight times daily at 0.5×0.5 km² pixel resolution over eastern China, the Korean peninsula, and Japan (Choi et. al., 2018) until it was shut down in early 2021. GOCI II, launched in February 2020, continues the GOCI mission with improved 0.25×0.25 km² pixel resolution, four additional spectral bands, and ten times daily retrievals over an expanded daytime window (Lee et al., 2023). The Yonsei aerosol retrieval (YAER) algorithm family computes AOD from GOCI

- 150 measurements by aggregating the native GOCI pixels to improve accuracy and cloud clearing into a 6×6 km² AOD product for GOCI I (GOCI YAER v2; Choi et al., 2018) and a 2.5×2.5 km² AOD product for GOCI-II (GOCI-II YAER; Lee et al., 2023). Lee et al. (2017) showed that fewer GOCI I pixels could be aggregated to produce a higher resolution AOD product with a modest tradeoff in precision. In this work, we use their 2×2 km² GOCI I AOD product (produced from 4×4 GOCI I pixels) which exhibits
- 155 an R² of 0.825 relative to AERONET for 2016 as compared to 0.858 for the standard GOCI YAER v2 6×6 km² AOD product (Lee et al., 2017).

To produce a continuous GOCI AOD training dataset, we first aggregate GOCI I AOD into an 8-h average (0:30-7:30 UTC) and GOCI II AOD into a 10-h average (23:15-8:15 UTC), representing the full daily records of each instrument, then regrid the 2.5×2.5 km² GOCI II AOD to the 2×2 km²

- 160 GOCI I grid by bilinear interpolation. We use the GOCI I AOD for March 2011 through December 2020 and the GOCI II AOD for January 2021 through December 2022. We remove 1.7% of pixels in the GOCI II record with an AOD outside the range observed by GOCI I (-0.05 to 3.6). The GOCI II AOD retrieval is biased low over land relative to AERONET while GOCI I shows no significant bias (Lee et al., 2023). To avoid spurious trends in the inferred PM_{2.5}, we incorporate relevant training data
 165 intercord with a DE exclosed by GOCI I (-0.05 to 3.6). The GOCI II
- 165 into the RF as described in Section 2.4.

Bias-corrected GEOS-Chem monthly mean AOD. Following Pendergrass et al. (2022), we use biascorrected GEOS-Chem CTM AODs to blend with GOCI I and II AODs in the gap-filling RF. The GEOS-Chem AODs are monthly means from a simulation by Zhai et al. (2021) for 2016 in East Asia with 0.5% × 0.625% resolution. We bigs correct the CEOS Chem AODs to metch the ensuel mean COCI

170 with $0.5^{\circ} \times 0.625^{\circ}$ resolution. We bias-correct the GEOS-Chem AODs to match the annual mean GOCI

I and II AODs on the $2 \times 2 \text{ km}^2$ grid for each year in the 2011–2022 period. In this way, we obtain a spatial distribution of monthly mean bias-corrected GEOS-Chem AOD values.

Surface PM_{2.5} data. We use hourly PM_{2.5} data from operational air quality networks in eastern China,
South Korea, and Japan, and average the data over 24 hours and over the 2×2 km² GOCI AOD grid to define targets for the RF algorithm. Data for eastern China are from the National Environmental Monitoring Center (CNEMC; <u>https://quotsoft.net/air/</u>), with measurements in Beijing beginning in December 2013 and for the rest of the country in May 2014. Following Zhai et. al. (2019) we remove values with more than 24 consecutive repeats in the hourly timeseries as likely in error. Data in China

- 180 are supplemented by US embassy data in Beijing (beginning in March 2011) and US consulates data in Shanghai (beginning in December 2011) and Shenyang (January 2013) (<u>https://www.airnow.gov/international/us-embassies-and-consulates</u>). These US embassy and consulates data have been used in previous air quality studies (Li et al., 2018; Pendergrass et al., 2019). Data for South Korea are from the AirKorea surface network (https://www.airkorea.or.kr/), which added PM_{2.5}
- 185 beginning in January 2015. Data for Japan are from the Japanese National Institute for Environmental Studies (NIES) for 2011-2021 (<u>https://tenbou.nies.go.jp/download/</u>) and for 2022 by the AEROS network (<u>https://soramame.env.go.jp/download</u>).

2.2 Gap-filled AOD and AOD missingness metric

The GOCI AOD records have gaps from clouds, snow cover, and other causes. Following Di et al. (2019), we perform gap-filling by using a separate GOCI AOD RF fit trained on the predictor variables of Table 1 except GOCI I and II AOD (the target variables in this case), the Gaspari-Cohn factor α (which has value 1 for all successful AOD retrievals), and the AirKorea surface air quality data. Because of the size of the AOD gap-filling problem, we use a separate RF for each year of data for computational economy. Training the GOCI AOD RF with annually disaggregated input data also

- 195 avoids bias from gap-filling GOCI I based on information from GOCI II and vice versa. As shown in Figure 1, we find using a ten-fold crossvalidation that our GOCI AOD RF explains 91% of 24-h variability ($R^2 = 0.91$; annual $R^2 = 0.96$) with no significant mean bias. Our approach here improves on the statistical gap-filling method used in Pendergrass et al. (2022) which led to smooth AOD interpolation over large missing areas which may have been unphysical. To understand the variables
- 200 driving the gap-filling prediction, we perform a SHAP analysis (lower panel) for a random sample of 0.1% of AOD data for 2016. NDVI is the most important predictor, perhaps because NDVI is predictive of AOD biases in both the GOCI I and II products (Choi et al., 2018; Lee et al., 2023), followed by GEOS-Chem modeled AOD and the six meteorological input variables; 2-m temperature and day of year are likely metrics for the seasonal variability of AOD.
- 205 The GOCI AOD gaps are non-random as they result from specific conditions that would not be part of the training dataset. However, Brokamp et al. (2018) found that when inferring $PM_{2.5}$ from AOD the non-randomness of AOD retrieval failure could be exploited to improve $PM_{2.5}$ predictions. Following Pendergrass et al. (2022), we compute an AOD missingness factor α that takes on a value of 1 if AOD is retrieved successfully at least once in a given day in a given grid cell and descending to 0 as
- 210 distance to the nearest successful retrieval increases. We compute α with the Gaspari-Cohn function, a

polynomial with a single radial argument r that takes on a maximum value of 1 for r = 0 and a minimum value of 0 for $r \ge 2$ (Gaspari and Cohn, 1999). We obtain r for a given grid cell and day by normalizing the distance from the grid cell to that of the nearest AOD retrieval against an empirically determined spatial correlation length scale ranging from 110 km to 170 km across the domain

215 (Pendergrass et al., 2022). By passing the Gaspari-Cohn factor α to the GOCI PM_{2.5} RF, we allow the algorithm to learn the optimal correction strategy in cases of AOD retrieval failure.

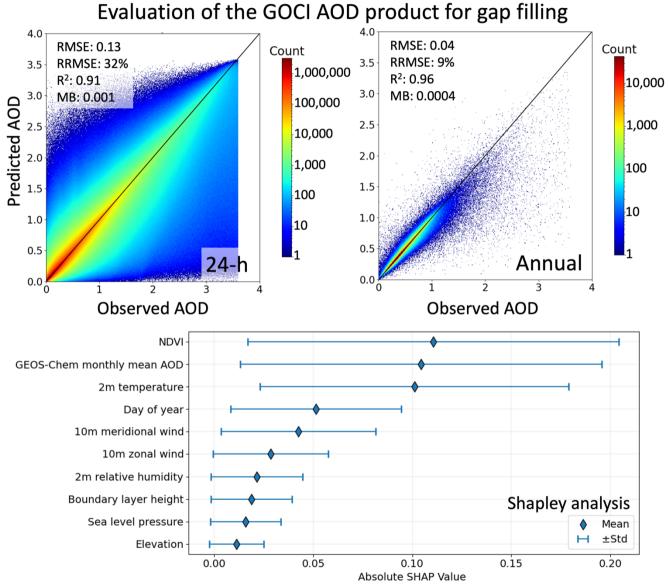


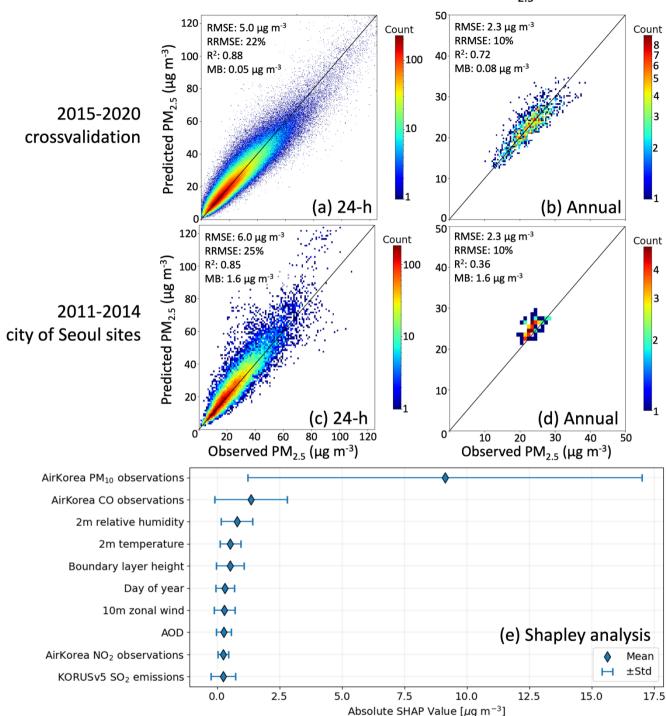
Figure 1: Evaluation of the GOCI AOD RF predictions. The top panels evaluate the GOCI AOD RF predictions in the 2011-22 training period at grid cells withheld entirely from training in a ten-fold crossvalidation procedure, aggregated at (a) 24-h and (b) annual resolution. Results are shown as two-dimensional histograms where pixel color corresponds to the count of observation/prediction correspondences within the corresponding bin, with statistics inset and the identity line shown in black. The bottom panel shows the top ten predictors of

AOD ranked by importance by the SHAP analysis. Predictor variable contributions are shown by mean absolute SHAP values and standard deviations.

225 2.3 Inferring South Korea PM_{2.5} before 2015

Prior to the January 2015 addition of PM_{2.5} measurements, the AirKorea surface network measured CO, O₃, NO₂, SO₂, and PM₁₀ concentrations. Many sites also recorded events of "yellow dust" transported from deserts in Mongolia and northern China (categorical true/false variable). We train a separate AirKorea PM_{2.5} RF on the 2015-2020 data, with all predictor variables in Table 1 except year and

230 country categorical variables, to predict 24-h 2011-2014 PM_{2.5} at AirKorea sites. Figure 2 evaluates the ability of the AirKorea PM_{2.5} RF product by its ability to match observed PM_{2.5} in the 2015-2020 period. We find using a ten-fold crossvalidation that the AirKorea PM_{2.5} RF is able to predict 88% of 24-h PM_{2.5} variability in the 2015-2020 record with no significant bias.



Evaluation of the AirKorea PM_{2.5} RF

Figure 2: Evaluation of the AirKorea PM_{2.5} RF predictions. The top panels evaluate the AirKorea PM_{2.5} RF predictions in the 2015-2020 training period at grid cells withheld entirely from training in a ten-fold crossvalidation procedure, aggregated at (a) 24-h and (b) annual resolution. Middle panels show an independent evaluation with observed 2011-2014 PM_{2.5} from the Seoul Research Institute surface network

in the city of Seoul, selecting the 20 sites that are collocated with AirKorea sites on the 2×2 km² GOCI grid. Panels (a-d) show two-dimensional histograms where pixel color corresponds to the count of observation/prediction correspondences within the corresponding bin, with statistics inset and the identity line shown in black. The bottom panel shows the top ten predictors of AirKorea PM_{2.5} ranked by importance by the SHAP analysis. Predictor variable contributions are shown by mean absolute SHAP values and standard deviations.

To independently evaluate the AirKorea PM_{2.5} RF for the pre-2015 period, we use 2011-2014 hourly PM_{2.5} data collected at 25 sites in the city of Seoul by the Seoul Research Institute of Public Health and Environment (NIER 2022) and select sites that are collocated with AirKorea sites within a 2×2 km² GOCI grid cell (20 sites). We find that the AirKorea PM_{2.5} RF reproduces successfully the 2011-2014 city of Seoul data (Figure 2), with statistics similar to the 2015-2020 crossvalidation. The annual R² is weak but this can be explained by the small sample size and small dynamic range. The most important predictor variable by far is PM₁₀, followed by CO and relative humidity. Figure 3 shows how the AirKorea PM_{2.5} RF maps 2011-2014 PM₁₀ data to infer PM_{2.5}.

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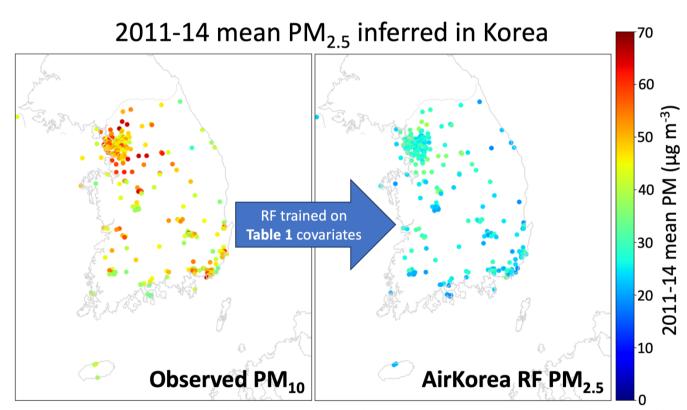


Figure 3: 2011-14 mean observed PM_{10} and inferred $PM_{2.5}$ at AirKorea sites. The AirKorea $PM_{2.5}$ RF is trained on data in Table 1 and its Shapley analysis is in Figure 2.

255 2.4 RF inference of PM_{2.5} from GOCI AOD

After producing a gap-filled AOD dataset with the GOCI AOD RF and a 2011-2014 $PM_{2.5}$ target dataset for South Korea with the AirKorea $PM_{2.5}$ RF, we can infer continuous 24-h 2011-2022 $PM_{2.5}$ in the study domain at 2×2 km² resolution. We train a GOCI $PM_{2.5}$ RF on all predictor variables in Table 1

for which we have gap-free coverage. The GOCI PM_{2.5} RF includes as its target all PM_{2.5} measurements
 from national networks, supplemented by PM_{2.5} from the US embassy and consulates in China and by
 the pre-2015 PM_{2.5} inferred in South Korea by the AirKorea PM_{2.5} RF.

We find that using year as a predictor variable substantially improves the GOCI PM_{2.5} RF fit, as its inclusion avoids an artificially large drop in PM_{2.5} concentrations from the 2020 to 2021-2022 period, corresponding with the switch from the GOCI I instrument to GOCI II (section 2.1). However,

- 265 in China prior to the 2014 start of surface network data, the use of year as a predictor is problematic because in that period $PM_{2.5}$ is only available from the US embassy and consulates which is too sparse. To solve this problem, we train a separate China-rebalanced GOCI $PM_{2.5}$ RF without year as a covariate and stopping in 2020 to avoid the GOCI II bias. In the China-rebalanced GOCI $PM_{2.5}$ RF, we also apply training data weights to increase the penalty to the RF if the US embassy and consulate $PM_{2.5}$ are poorly
- 270 modeled prior to 2014. We use the output of the China-rebalanced GOCI PM_{2.5} RF to overwrite the GOCI PM_{2.5} RF output prior to May 2014 in China.

Figure 4 compares 24-h and annual mean $PM_{2.5}$ network observations to predictions from the GOCI $PM_{2.5}$ RF for sites in 2×2 km² grid cells withheld from training. Annual mean values are obtained by averaging the 24-h predictions. We find using a ten-fold crossvalidation that our prediction captures

- 275 86% of the observed 24-h variance ($R^2 = 0.86$) and 95% of annual ($R^2 = 0.95$). Overall mean bias is only 0.26 µg m⁻³ but there are tail biases discussed later in this section. Applying the SHAP analysis to a random sample of 1% of the training data, we find that whether a grid cell is located in China is the most important predictor; this presumably serves as a proxy for the different vertical distribution of aerosols in the column relative to South Korea and Japan and also reflects the large dynamic range of
- 280 PM_{2.5} in China. Boundary layer height and AOD are the most important physical predictors, as would be expected.

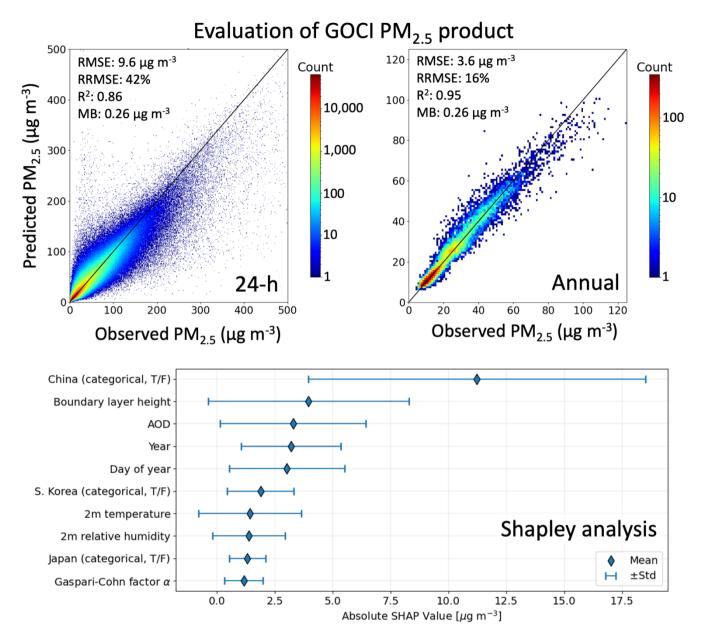


Figure 4: Evaluation of the GOCI PM_{2.5} RF predictions. The top panels evaluate the GOCI PM_{2.5} RF predictions in the 2011-2022 training period at grid cells withheld entirely from training in a ten-fold crossvalidation procedure, aggregated at (a) 24-h and (b) annual resolution.
 The panels show two-dimensional histograms where pixel color corresponds to the count of observation/prediction correspondences within the corresponding bin, with statistics inset and the identity line shown in black. The bottom panel shows the top ten predictors of GOCI PM_{2.5} ranked by importance by the SHAP analysis. Predictor variable contributions are shown by mean absolute SHAP values and standard deviations.

Figure 5 shows the performance of the GOCI PM_{2.5} product in the high tail of the distribution which is of particular interest for air pollution exposure but is notoriously difficult for RF algorithms to fit (*Zhang and Lu*, 2012; Pendergrass et al., 2022). Here, perhaps due to the very large training set, we find that the RF extends the successful fit to the high tail. Averaging data into bins each containing 0.1% of ordered observations, we find that the observed 24-h 99th percentile of 129 μ g m⁻³ is underestimated by 13.5% (annual by 7.6%) in the corresponding GOCI PM_{2.5} predictions. The observed 24-h 99.9th percentile of 319 μ g m⁻³ is underestimated by GOCI PM_{2.5} by 26.5% (annual by 21.0%).

These are relatively good RF performances for such high extremes.

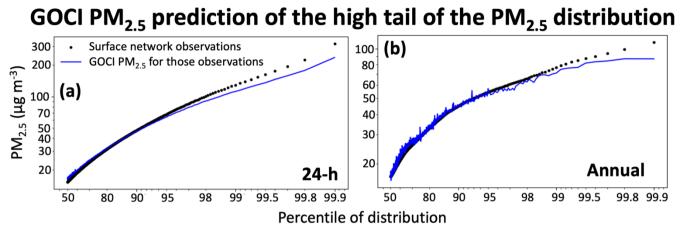


Figure 5: High tail of the $PM_{2.5}$ distribution in China, South Korea, and Japan for 2011-2022. The figure shows the mean binned percentiles of the 24-h and annual $PM_{2.5}$ concentrations measured at the surface networks, together with the corresponding mean GOCI $PM_{2.5}$ predictions sampled at those observed percentiles.

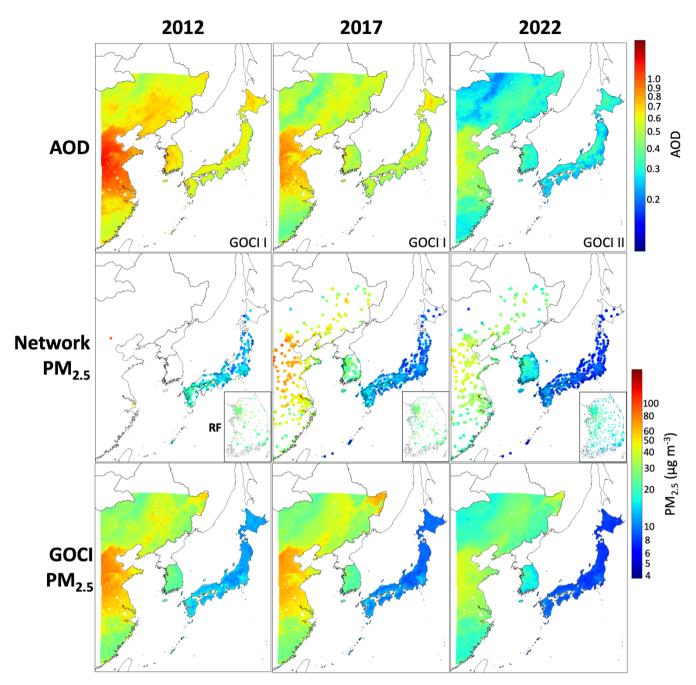
3 Results and discussion

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Here we present features and insights from our spatially and temporally continuous PM_{2.5} product generated from the GOCI AOD data from March 2011 to December 2022 with 2×2 km² spatial resolution and 24-h temporal resolution. We refer to this product as GOCI PM_{2.5} in what follows. Results for annual data are presented starting in 2012 as the first full calendar year of data.

Figure 6 (top row) shows gap-filled GOCI AODs in 2012, 2017, and 2022. AOD declined steadily in East Asia over the lifetime of the GOCI I instrument (2011-2020) and drops sharply in the transition to GOCI II (2021-2022) but this is due partly to a low bias in GOCI II AOD. The middle row

- 310 shows the PM_{2.5} network data, highlighting the spatial limitations as well as the temporal limitations before 2015. The bottom row shows our GOCI PM_{2.5} product, highlighting the spatial and temporal continuity over the period. The bias between GOCI I and II does not affect our GOCI PM_{2.5} product because the RF is given information to fit the GOCI data for individual years. The GOCI PM_{2.5} product shows high concentrations at the northeastern tip of China where there are no surface network data.
- 315 Such high concentrations are not predicted in air quality models (Zhai et al., 2021) but have been previously inferred from MODIS AOD satellite observations (Van Donkelaar et al., 2021). They may be due to high ammonia emissions missing from current inventories (Kong et al., 2019), but also could be overestimated due to regional biases in vertical distributions of aerosols such as from Russian wildfires.



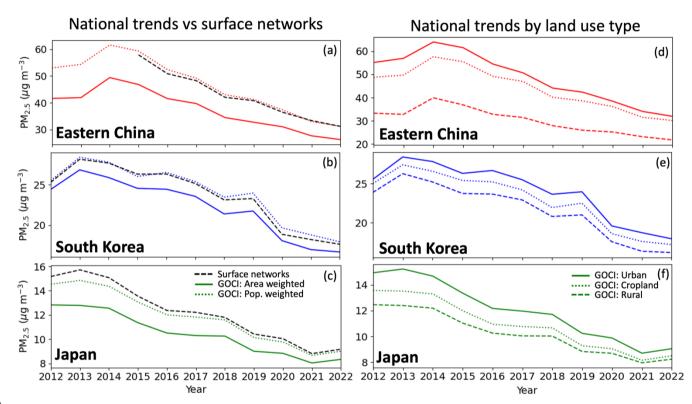
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Figure 6: GOCI gap-filled aerosol optical depth (AOD), $PM_{2.5}$ from air quality networks, and GOCI $PM_{2.5}$ obtained by applying a RF algorithm to the GOCI AOD data. Data are annual means for 2012 (the first year with complete GOCI data), 2017, and 2022. The gap-filled AOD data provide continuous 2×2 km² coverage of eastern China, S. Korea, and Japan for 2011-2022. The $PM_{2.5}$ network data are from individual sites and enlarged for visibility. The S. Korea insets in the middle panels provide greater resolution of network data gaps. $PM_{2.5}$ measurements from the AirKorea network started in 2015, and the S. Korea $PM_{2.5}$ network data shown for 2012 are from a RF

325 PM_{2.5} measurements from the AirKorea ne reconstruction as described in Section 2.3.

Figure 7 shows long-term trends of annual GOCI $PM_{2.5}$ for each country with averaging weighted by area, population, and land type (Table 1). Also shown are the trends from the $PM_{2.5}$ networks, including pre-2015 data for Korea from our RF fit of other network data (Section 2.3). The

- 330 GOCI PM_{2.5} trends for the population-weighted average mirror the network trends and extrapolate them to before the start of the records. Peak concentrations were in 2014 (China) and 2013 (South Korea, Japan) and have been decreasing steadily since. The anomalous peak in South Korea PM_{2.5} in 2019 is driven in part by unfavorable winter meteorological conditions (Cha et al., 2023). We find no COVID-19 anomaly in 2020, except perhaps in South Korea, possibly because emission decreases were offset
- 335 by increase in oxidants producing secondary aerosol (Chang et al., 2020; Huang et al., 2021; Yang et al., 2022). We also see a narrowing spread with time across land use types and averaging method (areal or population-weighted), consistent with more rapid improvements in polluted urban areas.



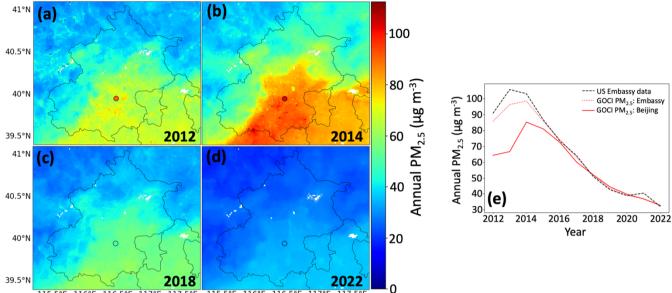
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Figure 7: Trends in annual mean GOCI PM_{2.5} concentrations averaged over eastern China, South Korea, and Japan for years with complete data (2012-2022). Also shown are the trends from the national PM_{2.5} networks (dashed black lines) averaged over 2×2 km² grid cells and requiring at least 80% of data for a given year. Surface network data in South Korea prior to 2015 are generated from the AirKorea PM_{2.5} RF using PM₁₀ and other covariates (Table 1). GOCI PM_{2.5} are shown as averages weighted by area, population, and land type.

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Figure 8 compares our GOCI $PM_{2.5}$ product for Beijing to the US embassy observations going back to 2012, and places them in the context of $PM_{2.5}$ concentrations in the broader city. GOCI $PM_{2.5}$ tracks the observations at the US embassy well, peaking in 2013-2014 and then rapidly decreasing, a pattern consistent with the 2012-14 increase in $PM_{2.5}$ in East China shown in Figure 7. From the GOCI

350 PM_{2.5} map we see that the US embassy was in a particularly polluted location in Beijing during the early part of the record but became more typical of the population-weighted city average after 2015. Improvements in PM_{2.5} air quality in Beijing have been relatively greater than in other urban areas (Zhai et al., 2019), as is apparent from Figure 6.



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Figure 8: Annual mean GOCI PM_{2.5} in Beijing compared with US embassy PM_{2.5} observations in individual years. The left panels show the distribution of PM_{2.5} in the city of Beijing (centered black outline) and surrounding area, with the location of the US embassy shown as a black circle. The right panel shows long-term trends at the US embassy site and averaged within Beijing city limits.

The long-term record produced in this work provides improved local information on 2011-2022 360 trends. Figure 9 shows trends in annual mean PM_{2.5} concentrations in South Korea derived from a linear regression applied to the annual GOCI PM_{2.5} in each 2×2 km² grid cell, as well as monthly trends in Seoul/Incheon starting in March 2011. The first five years of AirKorea PM_{2.5} records (2015-19) showed no decrease in the Seoul metropolitan area (SMA) despite local emissions controls as well as controls upwind in China, and an increase in winter (Pendergrass et al., 2022). However, the 2012-2022 record

- 365 shows steady improvements in PM_{2.5} across the country including the SMA. The lack of trend in the 2015-2019 period in the SMA reflected the brevity of the record, as seen by the addition of the 2011-2015 years with the AirKorea PM_{2.5} RF showing a decrease starting in 2013. Winter decrease after 2019 may have been further driven by a seasonal fine dust management program launched by the government of Seoul in 2019 that limits vehicle use, coal-fired power plants, and industrial activity from December
- 370 through March (Ministry of the Environment, 2019; Yonhap News, 2021), but also may show an impact from COVID-19 lockdowns (Koo et al., 2020).

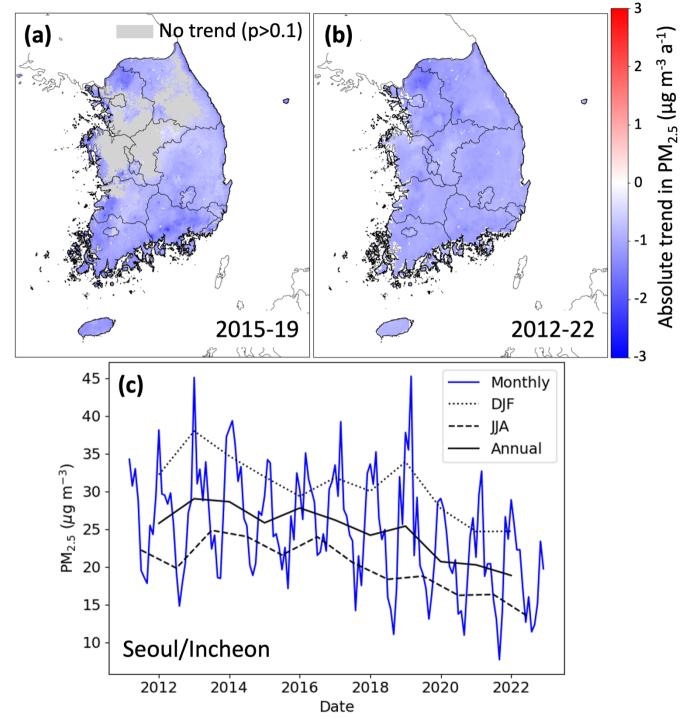


Figure 9: Trends in PM_{2.5} concentrations in South Korea. Panels in the top row show annual trends for (a) 2015-2019 and (b) 2012-2022. The trends are obtained by ordinary linear regression of the annual mean GOCI PM_{2.5} in each 2×2 km² grid cell with significant regression slope (p < 0.10). Grid cells with insignificant trends are plotted in gray. The bottom panel shows population-weighted GOCI PM_{2.5}

concentrations in Seoul and Incheon. Lines represent monthly (solid blue line), DJF (black dotted), JJA (black dashed), and annual (black solid) mean concentrations.

Figure 10 expresses the national trends in PM_{2.5} in terms of population exposure. In China, 380 where PM_{2.5} air quality is worst, we find the greatest improvements for the populations exposed to the highest pollution, leading to a narrowing spread of exposures across the country that is illustrated by the sharpening slope of the cumulative distribution. While in 2014 97% of the population in China within the GOCI domain was exposed to annual PM_{2.5} exceeding the national ambient air quality standard (NAAOS; 35 µg m⁻³), by 2022 the figure declined to 29%. However, over 99% of the population was

- 385 still exposed to annual $PM_{2.5}$ greater than 15 µg m⁻³, the NAAQS in Japan and South Korea. In 2022 in South Korea 92% of the population still was exposed to annual $PM_{2.5}$ greater than the NAAQS but all would have met the pre-2018 NAAQS of 25 µg m⁻³. Japan was fully compliant with its NAAQS by 2018 and its air quality has continued to improve since, consistent with an observed shift from urban to marine aerosols over the study period (Kobayashi et al., 2023). Across the domain, the maximum to
- 390 which any population is exposed decreases everywhere, which means that no population has been left behind in the improvements in PM_{2.5} air quality.

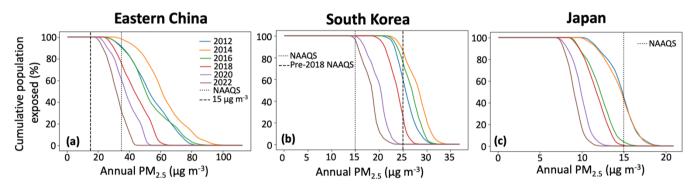


Figure 10: Trends in cumulative population exposure in countries within the study domain. The *y* axis shows the cumulative populations exposed to at least the annual PM_{2.5} level given on the *x* axis, with year indicated by color. Panel (a) shows Eastern China, (b) South Korea, and (c) Japan. Note different scales for the different panels. National ambient air quality standards (NAAQS) are shown in the vertical black dotted line.

The 2×2 km² resolution of our new GOCI PM_{2.5} product (compared to 6×6 km² in Pendergrass et al. (2022)) improves the representation of urban scale pollution events. This is illustrated in Figure 11 400 with a severe event in the SMA on 24–29 May 2016 previously shown by Pendergrass et al. (2022). Extreme concentrations and local gradients are better represented in the new product. Over the six-day period for the shown sites, we find an overall R² of 0.97 versus observations as compared with 0.77 for the 6×6 km² product in part because the resolution is now sharp enough to individually resolve all sites. A two-sample Kolmogorov-Smirnov test indicates that the 6×6 km² product has a statistically 405 significantly different distribution than the observations (p < 0.001) while the improved 2×2 km²

product is indistinguishable (p = 0.52).

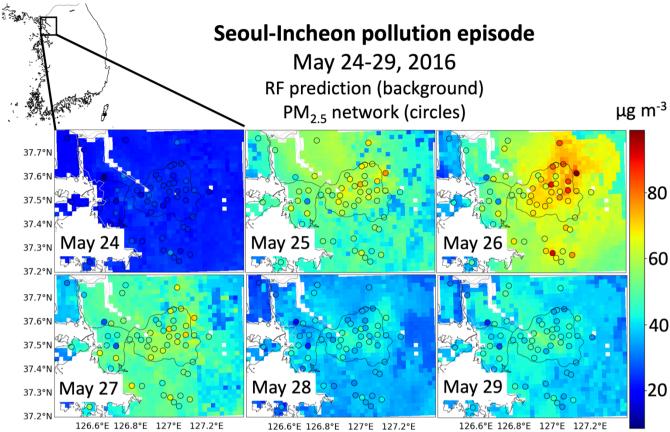


Figure 11: 24 h $PM_{2.5}$ concentrations during a pollution event in the Seoul Metropolitan Area (24–29 May 2016). Observations from the AirKorea surface network (circles) are overlaid on GOCI $PM_{2.5}$ produced in this work (2×2 km² grid). Seoul city limits are shown by the black outline in the panel center.

4 Conclusions

We produced a continuous 24-h data set of fine particulate matter (PM_{2.5}) concentrations over East Asia at 2×2 km² resolution for 2011-2022 by training a random forest (RF) machine learning algorithm on GOCI I and II geostationary satellite observations of aerosol optical depth (AOD) to predict PM_{2.5}
d15 observations from surface networks. The resulting GOCI PM_{2.5} dataset offers high-resolution coverage of the region over a twelve-year period of rapid change. It improves on our previous GOCI PM_{2.5} product (Pendergrass et al., 2022) in spatial resolution, record duration, and RF method. We produced the GOCI PM_{2.5} data in a three-step process. First, we gap-filled missing GOCI I and II AOD retrievals using an RF algorithm trained on covariates including gap size, chemical
transport model (CTM) output, meteorology, and land use variables. Second, to train on the GOCI I data starting in March 2011, before the start of PM_{2.5} monitoring in South Korea (2015), we trained

another RF to predict 2011-2014 $PM_{2.5}$ at AirKorea network sites using the pre-2015 data available at those sites and most notably PM_{10} . Finally, we used the gap-filled GOCI AOD along with the target

PM_{2.5} set expanded by the inferred 2011-2014 AirKorea PM_{2.5} and US embassy and consulate data in pre-2014 China to train an RF to predict PM_{2.5} across the study domain.

- 425 pre-2014 China to train an RF to predict PM_{2.5} across the study domain. The continuous 2011-2022 GOCI PM_{2.5} record at 2×2 km² resolution constructed in this manner reproduces the PM_{2.5} network observations with no significant bias and a relative root-mean-square error (RRMSE) of 22% for 24-h data and 10% for annual data. Its success extends to the high tail of the PM_{2.5} frequency distribution (severe pollution episodes). It shows that the air quality networks in all
- 430 three countries are representative of population-weighted exposure. The 2012-2022 full-year time series show PM_{2.5} peaking in 2014 (China) and 2013 (South Korea and Japan) and then steadily declining through the end of 2022 with steepest improvements in the most polluted regions. Population exposure over that period decreases for all quantiles of the distributions, implying that no region has been left behind in air quality improvement. While the Seoul Metropolitan Area (SMA) does not show a decrease
- 435 over the first five years of the PM_{2.5} network record (2015-2019), the longer 2012-2022 record shows a decline consistent with the rest of the country.

The $2 \times 2 \text{ km}^2$ resolution of our GOCI PM_{2.5} product enables successful representation of the fine-scale structure and statistical distribution of concentrations during urban pollution episodes, improving significantly on our previous $6 \times 6 \text{ km}^2$ product that had excessive smoothing. It should be of value for long term public health studies where continuity of PM₄ = data is essential.

440 value for long-term public health studies where continuity of $PM_{2.5}$ data is essential.

Data availability 24-h $2 \times 2 \text{ km}^2$ resolution 24-h GOCI PM_{2.5} from March 1, 2011 through December 31, 2022 are made publicly available on DataVerse (<u>https://doi.org/10.7910/DVN/0G07BS</u>, Pendergrass et al., 2024). PM_{2.5} data for eastern China are publicly available from the National

- 445 Environmental Monitoring Center (<u>https://quotsoft.net/air/</u>); data for South Korea are available from AirKorea (<u>https://www.airkorea.or.kr/</u>); data for Japan for 2011-2021 are available from the National Institute for Environmental Studies (NIES) (<u>https://tenbou.nies.go.jp/download/</u>) and for 2022 from the AEROS network (<u>https://soramame.env.go.jp/download</u>). The versions of GOCI I and GOCI II AOD data used in this paper are available on request to the corresponding author. All other input data to the
- 450 RF is publicly available at links given in the text and can also be provided on request.

Author Contributions DP and DJJ designed the study. DP developed the RF and performed analysis. SZ ran and analyzed chemical transport model data. SL and JL aided in satellite data processing. MK, HI, and YO aided in surface data processing. JK, DJJ, and HL provided scientific interpretation and discussion. All authors provided input on the paper for revision before submission.

Competing interests The authors declare that they have no conflict of interest.

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