

# Improved daily PM<sub>2.5</sub> estimates in India reveal inequalities in recent enhancement of air quality

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# 1 Abstract

2 Poor ambient air quality represents a substantial threat to public health globally. However, ac-  
3 curate measurement of air quality remains challenging in many parts of the world, including in  
4 populous countries like India, where ground monitors are scarce yet exposure and health burdens  
5 are expected to be high. This lack of precise measurement impedes understanding of how pollu-  
6 tion exposure changes over time and varies across different populations, and inhibits monitoring  
7 of interventions to improve air quality. Here we develop open-source daily fine particulate matter  
8 ( $PM_{2.5}$ ) datasets at a 10 km resolution for India from 2005 to 2023, using a region-specific two-stage  
9 machine learning model carefully validated on held-out monitor data that it was not trained on.  
10 Our model demonstrates robust out-of-sample performance, substantially outperforming existing  
11 publicly-available monthly  $PM_{2.5}$  datasets. We use model output to analyze long-term air quality  
12 trends, finding that  $PM_{2.5}$  increased across most of the country until around 2016 and then began  
13 to decline thereafter, partially driven by favorable meteorology in southern India. Importantly, re-  
14 cent  $PM_{2.5}$  reductions were substantially larger in wealthier areas, albeit from a higher initial level,  
15 but we find no evidence that the recently-adopted National Clean Air Program has improved air  
16 quality in targeted urban areas to date. Our results highlight the urgency of air quality control  
17 policies that effectively target both lower and higher socioeconomic groups. To further enhance air  
18 quality monitoring across populations in India and other countries, we use model output to propose  
19 locations where new ground monitors should be installed in India, and examine the adaptability of  
20 our method to other settings with scarce ground monitoring data.

## 21 INTRODUCTION

22 Exposure to ambient fine particulate matter (PM<sub>2.5</sub>) is a recognized global health concern. India,  
23 with its large population and high average pollution levels, bears a substantial share of the global  
24 health burden from poor air quality (1). Importantly, the health burden of air pollution often varies  
25 across individuals and groups with different socio-economic status, due to differences in pollution  
26 concentrations, as well as the increased sensitivity of health outcomes to pollution exposure in lower-  
27 income communities (2). While these disparities in air pollution exposure across wealth groups  
28 have been well documented in high-income countries (2–4), evidence from low- and middle-income  
29 countries remains limited, primarily due to sparse air quality monitoring networks, especially in  
30 rural areas where large proportions of the population live, along with a lack of data on wealth at  
31 fine temporal and spatial resolutions (3). A population-scale understanding of trends and exposure  
32 to air pollution, including in wealthier and poorer areas of low- and middle-income countries such  
33 as India, is urgently needed to understand and address the impacts of air pollution exposure across  
34 diverse socioeconomic groups.

35 Despite recent efforts in expanding the air quality monitoring network in the country, India  
36 still faces a great challenge in comprehensive measurement of surface air quality. The Central  
37 Pollution Control Board (CPCB) initiated the National Ambient Air Quality Monitoring (NAAQM)  
38 Network in 1984, beginning with the installation of manual monitoring stations, where pollutants are  
39 subsequently analyzed in the laboratory (5). Continuous Ambient Air Quality Monitoring System  
40 (CAAQMS), which provides real-time data, was first introduced in Delhi in 2006 and expanded to  
41 other cities after 2016 (6). The CPCB manages 883 manual stations and 438 continuous monitoring  
42 stations as of February 2023 (7), and this relatively small number of monitors results in a much  
43 higher ratio of population to continuous air quality monitors in India (3.2 million people per monitor)  
44 compared to the US (0.1 - 0.5 million people per monitor (8)), EU (0.1 million people per monitor  
45 (9)), and China (0.9 million people per monitor (10)). Moreover, most current CAAQMS monitors  
46 are situated in populous urban areas where wealthier people reside (Figure S1). The government of  
47 India has committed to expanding the CAAQMS network up to 1,000 monitors under the National  
48 Clean Air Program (NCAP), which started in 2019 (11); however, the placement of these additional  
49 continuous monitors remains an ongoing policy question, and it is uncertain whether environmental  
50 inequalities are considered or prioritized in determining their locations.

51 Previous population-based studies (12, 13) have explored the disproportionate exposure to air  
52 pollution and associated health impacts in low- and middle-income countries, including India, by  
53 utilizing publicly available modeled PM<sub>2.5</sub> estimates. One widely utilized dataset for such analyses is  
54 global monthly estimates of PM<sub>2.5</sub>, integrating satellite retrievals of Aerosol Optical Depth (AOD),  
55 atmospheric chemical transport models, and ground-based measurements (14). While this dataset  
56 has proven valuable on a global scale, significant uncertainties persist in regions with limited ground

57 monitoring stations, including India (14). Furthermore, it is likely that region-specific models could  
58 substantially outperform global models in measuring air pollution in a target region of interest, as  
59 has been found in the US (15).

60 In addition to the uncertainty in estimating PM<sub>2.5</sub> concentrations, the coarse temporal resolution  
61 of existing datasets (i.e monthly) hinders the assessment of short-term health effects of PM<sub>2.5</sub>, such  
62 as effects on all-cause, respiratory, and cardiovascular mortality (16–23). Furthermore, monthly  
63 estimates fail to capture short-term spikes in PM<sub>2.5</sub> emissions at the local to regional scales, such as  
64 crop residue burning. Acknowledging the need for a dataset with finer temporal resolution, a growing  
65 number of studies (24–26) have worked on developing daily PM<sub>2.5</sub> estimates for India. However,  
66 their datasets are not publicly accessible, inhibiting their use by both researchers and a host of  
67 governmental and non-governmental actors. Using up-to-date data to comprehensively characterize  
68 temporal and spatial trends in exposure, and potential differences in exposure by socioeconomic  
69 status, is also critical for understanding how and why exposures are changing and who is being  
70 most impacted.

71 Here we develop an open-source daily PM<sub>2.5</sub> dataset at 10 km resolution for India over 2005  
72 - 2023 by training a machine learning model to predict the limited available ground monitor data  
73 with abundant data from remote sensing. Previous studies have employed machine learning al-  
74 gorithms, such as neural networks, random forest, and extreme gradient boosting (XGBoost), to  
75 predict ambient PM<sub>2.5</sub> concentrations (27, 28). Due to the theoretical relationship between satellite  
76 AOD and surface PM<sub>2.5</sub> (27, 29), satellite-derived AOD has long been a key input feature used  
77 to train machine learning models for air pollution measurement, often in combination with data  
78 on meteorology, land cover, elevation, and population density (27, 28). However, the substantial  
79 amount of missing AOD values due to clouds and bright surfaces has posed challenges in reducing  
80 predictive errors when estimating PM<sub>2.5</sub> concentrations through machine learning methods. (27,  
81 30). To address this issue, a common recent approach involves imputing missing data in AOD  
82 observations using machine learning methods (24, 25, 31). Additionally, new satellite sensors pro-  
83 vide alternative input features for predicting PM<sub>2.5</sub> concentrations without relying on AOD, such as  
84 data from the Sentinel-5 Precursor (Sentinel-5P) mission’s TROPospheric Monitoring Instrument  
85 (TROPOMI) (30, 32, 33). However, it is still unknown how much the predictive performance  
86 differs between satellite-derived AOD and TROPOMI-based features in estimating PM<sub>2.5</sub> concen-  
87 trations, and whether the two sources of data independently add value in predicting surface PM<sub>2.5</sub>  
88 concentrations.

89 To take advantage of both the longer available time series of AOD data and information from  
90 these newer sensors, we use available ground monitoring data to train two separate models, which  
91 we term as the "Full model" and the "AOD model". The Full model combines both Moderate  
92 Resolution Imaging Spectroradiometer (MODIS) AOD (34) and TROPOMI satellite inputs (nitro-  
93 gen dioxide (NO<sub>2</sub>) (35) and carbon monoxide (CO) (36) along with other inputs, Figure S2 and

94 Materials and Methods) and is trained on data from July 2018, when TROPOMI features become  
95 available, through September 2023 (Figure S2, S3). It produces daily estimates across the coun-  
96 try for the period corresponding to its training period. The AOD model retains all inputs except  
97 TROPOMI and trains on data beginning January 2013 (Figure S2, S3), and is used to generate  
98 daily predictions from January 2005 to September 2023. In both the Full and AOD models, we  
99 first fill the missing satellite observations in either AOD or TROPOMI using a separate machine  
100 learning model (see Materials and Methods). The gap-filled estimates are then combined with the  
101 other features in a second-stage model that predicts surface PM<sub>2.5</sub> concentrations as measured by  
102 CAAQMS monitors (n = 435) (Figure S4).

103 Critically, and in contrast to related work(24, 25), our second stage model is evaluated using  
104 spatial cross-validation (CV) (Figure S5) – i.e. we evaluate model predictions on entirely held-out  
105 monitoring stations – rather than conventional random CV, in which a given station can contribute  
106 data to both the training and test sets. This more challenging performance metric is meant to ensure  
107 that the model generalizes to locations where it has no data to train, which is most of India. Using  
108 spatial CV, we then calculate two performance metrics: (1) the total R<sup>2</sup>, or the percent of variation  
109 in observed PM<sub>2.5</sub> explained by model predictions, as well as (2) the "within" R<sup>2</sup>, or the percent of  
110 variation in observed PM<sub>2.5</sub> explained by predictions after accounting for both differences in average  
111 PM<sub>2.5</sub> across locations as well as seasonal differences in PM<sub>2.5</sub> within a given location. In essence,  
112 the within R<sup>2</sup> measures whether our model can predict daily PM<sub>2.5</sub> anomalies relative to location-  
113 and season-specific averages, rather than simply predict spatial or seasonal patterns correctly. This  
114 "within" variation is often exploited in studies of the impact of air pollution on health and related  
115 societal outcomes, and thus is a highly-relevant, if rarely-reported, performance metric.

116 We utilize model-derived predictions to better characterize nationwide air quality trends, in-  
117 cluding inequalities in PM<sub>2.5</sub> exposure by region and wealth level, and to identify locations with  
118 extreme PM<sub>2.5</sub> concentrations. We then use our predictions, along with emissions inventories, me-  
119 teorological data, and administrative data on national air quality programs, to better understand  
120 why pollution concentrations are changing. This includes, to our knowledge, the first evaluation  
121 of whether the recently-adopted National Clean Air Program (NCAP) is improving air quality in  
122 targeted areas relative to a comparable set of non-targeted areas. Subsequently, to address current  
123 gaps in the air quality monitoring network in India, we use our PM<sub>2.5</sub> estimates and compressed  
124 sensing methods (37) (see Materials and Methods) to propose where additional air quality monitors  
125 could be installed to maximize the ability of the ground network to capture variability in surface  
126 air pollution across both wealthier and lower-wealth regions. Finally, we explore how our approach  
127 could supplement limited monitor data in other low- and middle-income countries by investigating  
128 the number of air quality monitors required to achieve reasonable model performance using our  
129 machine learning approach.

## 130 RESULTS

### 131 Model performance

132 The spatial out-of-sample performance of the Full model, assessed across daily  $\text{PM}_{2.5}$  observations  
133 in the held-out fold, yielded an  $R^2$  of 0.68 (Figure 1A). Importantly, our model effectively predicts  
134 local and temporal  $\text{PM}_{2.5}$  variation rather than average differences in  $\text{PM}_{2.5}$  concentrations between  
135 locations, months, or years (within  $R^2 = 0.49$ ) (Figure 1A). The AOD model demonstrated a  
136 comparable performance, with an  $R^2$  of 0.64 and within  $R^2$  of 0.45 (Figure S6). When aggregated  
137 at the monthly level, our model substantially outperforms the existing publicly-available monthly  
138  $\text{PM}_{2.5}$  dataset (14) when evaluated on Indian monitoring data, with an  $R^2$  of 0.74 and within  
139  $R^2$  of 0.52 (Figure S7). Examining location-specific performance, the out-of-sample within  $R^2$  for  
140 each 10 km grid suggests that the northwest, northeast, and south regions exhibit higher within  
141  $R^2$  (Figure 1B) (for the performance of AOD model, Figure S8). We find that these differences  
142 in regional performance are substantially driven by differences in the level and variance of  $\text{PM}_{2.5}$   
143 concentrations at the station level and the number of air quality monitors within 100 km (Figure  
144 S9); performance is much higher in locations with higher and more variable  $\text{PM}_{2.5}$  and with more  
145 stations nearby, consistent with the model having an easier time learning patterns in these higher  
146 signal-to-noise areas.

147 To further evaluate the out-of-sample spatio-temporal performance of the Full model in different  
148 locations, the daily variations in the observed and predicted  $\text{PM}_{2.5}$  are compared in five mega-cities  
149 in India: New Delhi, Mumbai, Bangalore, Chennai, and Kolkata (Figure 1C) (for the AOD model,  
150 Figure S10). These five mega-cities are selected based on their population size, with each being  
151 among the most populated cities in India (38). Among these cities, New Delhi shows the highest  
152 performance,  $R^2$  of 0.83 and within  $R^2$  of 0.69 (Figure 1C). This aligns with our prior analysis  
153 on model performance because New Delhi demonstrates dense CAAQMS network and significant  
154 variance in  $\text{PM}_{2.5}$  concentrations (Figure S11), primarily driven by distinct seasonal patterns caused  
155 by meteorological conditions restricting pollutant dispersion and the concurrent operation of brick  
156 manufacturing around Delhi during winter. (39, 40).

157 We observe variations in model performance driven by distinct seasonality throughout the year  
158 across the winter (December to February), spring (March), summer (April to May), monsoon (June  
159 to September), and post-monsoon (October to November). Both the Full and AOD models demon-  
160 strate strong performance in dry seasons (winter and post-monsoon) (Figure S12). However, their  
161 performance declines during spring, summer, and monsoon periods (Figure S12), likely the result  
162 of clouds introducing noise in the remotely-sensed input features during these wetter periods.

163 We find consistent results in sensitivity analysis, yielding an  $R^2$  of 0.62 and within  $R^2$  of 0.44  
164 when the Full model underwent training and testing on significantly larger blocks based on latitude

165 (Figure S13, S14), which helps rule out the possibility that our high model performance is simply  
166 being driven by auto-correlation between nearby train and test locations. When evaluated using 10-  
167 fold random CV rather than spatial CV, our model showed notably higher performance ( $R^2$  of 0.85  
168 and within  $R^2$  of 0.72) (Figure S15), highlighting the potential of random CV to overstate model  
169 performance on critical real-world applications (i.e. accurately predicting variation in locations  
170 where the model was not trained).

171 Furthermore, as a result of model comparison using the same sets of training and test data from  
172 July 10, 2018, to September 30, 2023, the Full model reported the highest performance, achieving  
173 an  $R^2$  of 0.67 and within  $R^2$  of 0.55 (Figure S16). The TROPOMI model, which excludes AOD  
174 but incorporates other features present in the Full model, slightly outperformed the AOD model  
175 by 0.01 in  $R^2$  and within  $R^2$  (Figure S16). This suggests that, at least in our setting, TROPOMI  
176 data can be a substitute for AOD in predicting  $PM_{2.5}$  concentrations, which is perhaps appealing  
177 given their lower amount of missing data compared to AOD. Nevertheless, the combined use of both  
178 TROPOMI and AOD features provides the strongest predictive power (Figure S16).

### 179 **Long-term spatiotemporal trends in predicted $PM_{2.5}$ concentrations**

180 To better understand longer-term shifts in  $PM_{2.5}$  concentrations, we calculate the changes in average  
181  $PM_{2.5}$  concentrations between six-year blocks, beginning in 2005-2010 (Figure 2A) and ending in  
182 either 2011-2016 (Figure 2B) or 2017-2022 (Figure 2C). We find that much of India experienced  
183 substantial increases in  $PM_{2.5}$  concentrations between 2011-2016 compared to 2005-2010, except for  
184 regions such as Jammu and Kashmir, Punjab, and Rajasthan states. However, the pace of increase  
185 moderated in more recent years, and during 2017-2022, a higher percentage of locations across  
186 the country showed decreases in  $PM_{2.5}$  concentrations (45 % of grid cells at a 10 km resolution)  
187 compared to 2011-2016 (16 % of grid cells), with notable decreases observed in Jammu and Kashmir,  
188 Punjab, Haryana, Delhi, and Rajasthan states and union territories.

189 To characterize trends in  $PM_{2.5}$  concentrations, we quantify population-weighted annual average  
190  $PM_{2.5}$  concentrations for the country overall and five mega-cities from 2005 to 2022 (Figure 2D) by  
191 combining our 10km  $PM_{2.5}$  estimates with gridded population data. Among the five mega-cities,  
192 the average New Delhi resident has consistently faced the highest population-weighted average of  
193  $PM_{2.5}$  concentrations, with  $88.67 \mu\text{g}/\text{m}^3$  in 2022, more than double India's annual national air  
194 quality guideline of  $40 \mu\text{g}/\text{m}^3$  (41) (Figure 2D). Similarly, residents in India overall, as well as those  
195 in Kolkata and Mumbai, have consistently experienced  $PM_{2.5}$  levels exceeding the national annual  
196 threshold (Figure 2D). Notably, we estimate that residents in all mega-cities have experienced a  
197 moderate decline in  $PM_{2.5}$  exposure since 2016-2018 (Figure 2D), as assessed by computing the 3-  
198 year rolling averages of population-weighted  $PM_{2.5}$  concentrations (Figure 2E). Mumbai exhibited  
199 the most substantial decline of 10 %, followed by New Delhi with 8 % in 2020-2022 (Figure 2E).

200 We further examine whether the observed declining trend since 2016-2018 is attributable to  
201 meteorological variability, including increasing trends in precipitation and relative humidity (42,  
202 43) observed in 70 % and 90 % of 10 km grid cells, respectively, from 2005-2015 to 2016-2022  
203 (Figure S17). Employing trend analysis (see Materials and Methods), we compare the observed  
204 average annual trend for 2005-2015 and 2016-2022 with the meteorology-corrected trend for the same  
205 periods. Our analysis revealed that the declining trend in PM<sub>2.5</sub> concentrations from 2016 to 2022  
206 was influenced by meteorological variability in the southern regions, but not in the northern regions  
207 such as Delhi, Haryana, Rajasthan, Uttar Pradesh, and Bihar (Figure S18). This suggests that  
208 without meteorological influence, the southern regions would have experienced fewer decreases in  
209 PM<sub>2.5</sub> concentrations from 2016 to 2022. In contrast, we find little evidence that the increasing trend  
210 from 2005 to 2015 was driven by changing meteorology (Figure S18), suggesting that these increases  
211 could be attributable to increased anthropogenic activities. We then confirm these trends using  
212 emissions data obtained from the Emissions Database for Global Atmospheric Research (EDGAR)  
213 (44, 45), focusing particularly on PM<sub>2.5</sub> and Black Carbon (BC) emissions. Notably, nationwide  
214 declines in BC emissions were observed in 2018 compared to 2016, and some locations experienced  
215 decreases in PM<sub>2.5</sub> emissions in 2018 (Figure S19). When emissions data beyond 2018 becomes  
216 available, further analysis can confirm whether the declining trend in PM<sub>2.5</sub> concentrations align  
217 with PM<sub>2.5</sub> and BC emissions in more recent years.

218 As declines in PM<sub>2.5</sub> since 2016 are not attributed to favorable meteorology, we examine whether  
219 India’s air quality control policies, particularly the National Clean Air Programme (NCAP), might  
220 have contributed to recent reductions in ambient PM<sub>2.5</sub> concentrations. Acknowledging the need for  
221 improved air quality to reduce health and societal burdens in the country, the government of India  
222 has recently proposed and implemented a number of air quality control measures, such as NCAP,  
223 the Bharat Stage-VI (BS-VI) emission standards for vehicles that mandated vehicles to adhere to  
224 PM emission limits as strict as European standards, which went into effect in Delhi in 2018 and  
225 other parts of India in 2020 (46), and the closure of multiple coal-fired power plants located near  
226 the Delhi National Capital Region (NCR) (47). However, the specific contribution of each policy  
227 to improving nationwide and regional air quality remains uncertain.

228 NCAP was initiated in 2019 with the goal of reducing key air pollutants, including PM<sub>2.5</sub>, by  
229 20 to 30% by 2024, using the pollution levels observed in 2017 as a baseline (7). Focusing on 131  
230 non-attainment cities across 28 states and union territories, selected based on air quality data from  
231 2015 to 2019 (7), one of the primary objectives of NCAP is to prompt each non-attainment city to  
232 prepare and implement a clean air action plan that details sector-specific interventions to improve air  
233 quality with predetermined timelines and an agency responsible for execution of each intervention  
234 (7, 11). Under the NCAP, 102 out of the 131 non-attainment cities submitted comprehensive city  
235 action plans, which were approved by the CPCB in July 2020 (11).

236 We use a difference-in-differences approach to assess whether the implementation of the NCAP



237 has affected changes in ambient  $\text{PM}_{2.5}$  concentrations to date. Our approach compares within-  
238 subdistrict changes in  $\text{PM}_{2.5}$  over time, in targeted and non-targeted areas, before and after initiation  
239 of NCAP. Our analysis is constrained to the subdistrict level, rather than the city level, due to the  
240 limited availability of reliable city-level shapefiles in India. We denote "treated" subdistricts as  
241 the 102 non-attainment cities whose city clean action plans were approved in 2020, and select a  
242 set of corresponding control subdistricts that were not targeted by NCAP using a propensity score  
243 method that matches pre-treatment trends in air pollution and covariates between later-treated and  
244 never-treated subdistricts (see Materials and Methods). To account for spillover effects, we exclude  
245 subdistricts adjacent to treatment subdistricts and any others within a 50 km buffer. Consequently,  
246 88 treatment subdistricts and 74 control subdistricts are included (Figure S20), and the effect of  
247 NCAP is estimated by comparing whether trends in air pollution diverged between treated and  
248 control units after 2020. Our analysis reveals that there is no evidence that NCAP contributed to  
249 reducing  $\text{PM}_{2.5}$  concentrations both in 2021 and 2022 (Figure S21) in targeted cities.

250 The observed decreases in  $\text{PM}_{2.5}$  concentrations, especially in the mega-cities in 2020, are instead  
251 more consistent with previous studies examining the impact of the COVID-19 pandemic on air  
252 quality in India (48–52), which highlighted a significant decrease (43%) in  $\text{PM}_{2.5}$  in 2020 compared  
253 to 2017-2019 in urban areas, after controlling for meteorological variability (48). Other various  
254 air quality control policies in India, including the implementation of BS-VI emission standards and  
255 the closure of power plants near Delhi, may have contributed to the nationwide or regional  $\text{PM}_{2.5}$   
256 declines. Our  $\text{PM}_{2.5}$  estimates could be used to evaluate the impact of these programs in future  
257 work.

## 258 Population exposure to $\text{PM}_{2.5}$ concentrations

259 To understand the population exposure to daily high levels of  $\text{PM}_{2.5}$  concentrations, we calculate  
260 the average number of days that each grid cell exceeded the WHO guideline of  $15 \mu\text{g}/\text{m}^3$ , the  
261 national guideline of  $60 \mu\text{g}/\text{m}^3$ , and the extreme concentration of  $100 \mu\text{g}/\text{m}^3$  between 2018 and  
262 2022. Notably, much of India experienced over 300 days above the WHO daily threshold, except for  
263 the northeastern region (Figure 3A). Delhi, Rajasthan, Uttar Pradesh, and Bihar encountered days  
264 exceeding the national guideline of  $60 \mu\text{g}/\text{m}^3$  for at least 250 days (Figure 3A). Moreover, certain  
265 locations in Delhi, Uttar Pradesh, and Bihar observed extreme days with  $\text{PM}_{2.5}$  concentrations  
266 exceeding  $100 \mu\text{g}/\text{m}^3$  for 100 to 150 days (Figure 3A).

267 We also assess the proportion of the overall population exposed to elevated annual concentrations  
268 of  $\text{PM}_{2.5}$  over our study period. Notably, the entire population in India has consistently faced  
269 exposure above the WHO annual threshold of  $5 \mu\text{g}/\text{m}^3$  over the 17 years (Figure 3B). Although  
270 there was a decrease in 2020, approximately 60% of the population consistently experienced exposure  
271 exceeding the national annual guideline of  $40 \mu\text{g}/\text{m}^3$ , and 10% experienced extreme levels of  $\text{PM}_{2.5}$ ,

272 with an annual average of  $80 \mu\text{g}/\text{m}^3$  (Figure 3B). Further analysis of the spatial distribution of these  
273 exposed populations revealed that 63% of the locations exceeded the national guideline between 2018  
274 and 2022 (Figure 3C). Areas with annual average  $\text{PM}_{2.5}$  concentrations exceeding  $80 \mu\text{g}/\text{m}^3$  were  
275 predominantly observed around Delhi and in Bihar state (Figure 3C).

## 276 **Inequalities in $\text{PM}_{2.5}$ exposure**

277 Identifying disparities in  $\text{PM}_{2.5}$  exposure by socio-economic status is essential to understanding  
278 pollution burdens and developing policy measures to alleviate them. To understand how pollution  
279 concentrations vary with socioeconomic status in India, we combine recent high-resolution estimates  
280 of local-level asset wealth (54), a common proxy for socioeconomic status, with spatial and temporal  
281 variation in our  $\text{PM}_{2.5}$  predictions at a 10 km resolution.

282 Utilizing 5-year average  $\text{PM}_{2.5}$  concentrations from 2015 to 2019 (see Materials and Methods), we  
283 find that the wealthiest quintile of the population is slightly less likely to experience concentrations  
284 above the national annual guideline of  $40 \mu\text{g}/\text{m}^3$  as compared to other quintiles of the wealth  
285 distribution (Figure 4A) – although substantial majorities in all quintiles are exposed to levels  
286 above this guideline. However, areas in the top two wealth quintiles are substantially more likely  
287 to live in areas with extreme  $\text{PM}_{2.5}$  concentrations above  $80 \mu\text{g}/\text{m}^3$  annually (23.0 % and 21.0 %,  
288 respectively) (Figure 4B), consistent with a previous study revealing that wealthy populations live in  
289 polluted urban centers in low- and middle-income countries (3). Due to these extreme exposures, an  
290 average person at the 90th percentile of wealth in India has consistently faced higher  $\text{PM}_{2.5}$  exposure  
291 than the average Indian or someone at the 10th percentile of wealth from 2005 to 2022, holding  
292 wealth constant across years (Figure 4C). Notably, since 2016, an average wealthy individual has also  
293 experienced faster declines in  $\text{PM}_{2.5}$  concentrations than an average poor individual, particularly  
294 evident since the start of the COVID-19 pandemic, yet notably apparent as early as 2017-2019.  
295 (Figure 4C, 4D, S23). This has shrunk the wealth gap in average  $\text{PM}_{2.5}$  over time. These  $\text{PM}_{2.5}$   
296 reductions disproportionately experienced by wealthier individuals highlights the urgent need of air  
297 quality mitigation policies that effectively target both the poor and the rich.

## 298 **Assessing the optimal placement of air quality monitors ensuring equality**

299 The current CAAQMS network is sparser in poorer communities, limiting understanding of dispro-  
300 portionate  $\text{PM}_{2.5}$  exposure. While our predictions enable investigation of nationwide trends and  
301 exposure across wealth groups, ground monitor data would enhance precision of such monitoring  
302 and ground monitor data will likely remain the basis for official evaluation of air quality trends  
303 and policy attainment. The government of India has committed to expanding the CAAQMS up to  
304 1,000 monitors under NCAP(11) to aid in more comprehensive air pollution monitoring. However,  
305 it is uncertain whether placement decisions account for the ability to accurately monitor pollution

306 concentrations across the socioeconomic spectrum.

307 We use compressed sensing methods (37, 55) to propose strategic placement of additional 565  
308 CAAQMS monitors, or the remainder of the 1,000 total monitors proposed to be installed under  
309 NCAP, using data from our AOD model to inform monitor placement (see Materials and Methods).  
310 Our approach identifies baseline national-scale long-term variability of PM<sub>2.5</sub> concentrations, and  
311 then chooses locations of additional monitors that would optimally capture local and short-term  
312 anomaly spikes in PM<sub>2.5</sub> exceeding this baseline across the country. When placing monitors, we  
313 prioritize low-wealth locations to ensure that sudden spikes occurring in poorer communities are also  
314 captured. The identified placement of monitors (Figure 5A) highlights the need for an additional  
315 dense network in northern India, particularly in Rajasthan, Delhi, Haryana, Uttar Pradesh, Bihar,  
316 and Assam states and union territories, as well as in the southeastern region, including West Bengal  
317 and Telangana states. The proposed additional network would help promote equality in nationwide  
318 exposure assessment while also enabling the network to maximally capture spatial and temporal  
319 variation in surface PM<sub>2.5</sub> concentrations.

## 320 **Examining applicability of our model to other low- and middle-income settings**

321 Other low- and middle-income countries also face challenges in understanding overall levels and  
322 trends in population exposure to PM<sub>2.5</sub> as well as inequalities in these exposures, due to limited  
323 ground-monitoring data. Our method could provide resource-efficient alternative to establishing  
324 extensive monitoring network by generating predictions for non-monitored locations, but it relies  
325 on having at least some amount of ground monitoring data to train and validate predictions. To  
326 understand how the performance of our machine learning approach changes as the ground network  
327 becomes sparser, we vary the number of monitors our model is allowed to see in training and quantify  
328 the relationship between the number of monitors in training and model performance. To estimate  
329 uncertainty in performance, we repeat this experiment a thousand times, resampling a fixed number  
330 of stations for training each time and re-estimating model performance on a disjoint set of sampled  
331 test stations (for more details, see Materials and Methods).

332 We observed an nonlinear increase in model performance, as evaluated on held-out test monitors,  
333 ranging from an R<sup>2</sup> value of 0.50 to 0.33 with 25 monitors, and reaching R<sup>2</sup> values of 0.68 and 0.54  
334 with 300 monitors (Figure 5B). When evaluated at the monthly level using the same sets of daily  
335 predictions derived from this experiment, we achieved R<sup>2</sup> and within R<sup>2</sup> values comparable to those  
336 of the existing benchmark publicly-available monthly PM<sub>2.5</sub> dataset(14) (Figure S7) while training  
337 only on 25 and 50 monitors, respectively (Figure 5C). These results indicate that investment in a  
338 moderate number of reference-grade air quality monitors, when combined with information from  
339 satellites, can enable training of machine-learning-based model that can predict PM<sub>2.5</sub> concentra-  
340 tions with performance that exceeds benchmark datasets commonly used for health impact analysis

341 in low- and middle-income countries. While variations in  $\text{PM}_{2.5}$  concentrations and country sizes  
342 differ, these findings offer valuable insights for other countries designing their monitoring network  
343 and implementing our machine learning method to understand nationwide trends and exposure to  
344  $\text{PM}_{2.5}$  across different populations.

## 345 DISCUSSION

346 Here, we generate daily  $\text{PM}_{2.5}$  predictions at a spatial resolution of 10 km across India from 2005  
347 to 2023. These daily estimates perform well over the range of observed monitor  $\text{PM}_{2.5}$  measure-  
348 ments, and very accurately capture temporal variations in  $\text{PM}_{2.5}$  concentrations, including daily  
349 peaks, within mega-cities in India. We find a declining trend in average  $\text{PM}_{2.5}$  concentrations since  
350 2016-2018, particularly in northern India, and confirm that these reductions are not attributable  
351 to meteorological variability nor to NCAP, a recently-begun nationwide air quality improvement  
352 program; smaller declines in southern India are driven in part by favorable trends in meteorology.  
353 Our analysis also provides a comprehensive characterization of the spatial distribution of popula-  
354 tions exposed to elevated levels of daily and annual  $\text{PM}_{2.5}$ , revealing that wealthier people are more  
355 likely to live in areas with extreme  $\text{PM}_{2.5}$  concentrations but that they have also experienced faster  
356 reductions in exposure in recent years. We propose the strategic placement of additional CAAQMS  
357 monitors to more effectively capture high  $\text{PM}_{2.5}$  episodes occurring in both poorer and wealthier  
358 locations, and we study the applicability of our approach in settings where existing or proposed  
359 monitoring networks could be even sparser than in our Indian study context, finding that only a  
360 relatively small number of monitors are needed to train a relatively accurate prediction model.

361 In comparison to many existing efforts to estimate  $\text{PM}_{2.5}$  concentrations using machine learning  
362 (27, 28), we incorporate data from multiple recent satellite sensors to estimate pollution concen-  
363 trations across the country. Additionally, we spatially validate predictions against time series of  
364 held-out monitor observations, which stands in contrast to the random CV used in many previous  
365 machine learning-based efforts (24, 25, 56, 57). Finally, our work complements recent machine  
366 learning-based studies to estimate  $\text{PM}_{2.5}$  concentrations by providing insights into the predictive  
367 power of TROPOMI features in contrast to AOD.

368 Our  $\text{PM}_{2.5}$  predictions could likely be further improved through improvements in both the  
369 monitor-based ground truth data and in the remotely-sensed input features. Our method relies on  
370 ground  $\text{PM}_{2.5}$  observations acquired from CAAQMS monitors for training; however, the manage-  
371 ment of these monitors and quality of the collected data has not been verified by a third-party insti-  
372 tution (5). For instance, in the UK, all regulatory air quality data collected as part of the Automatic  
373 Urban and Rural Network is validated by an independent agency (5). Using the quality-assured  
374 ground measurements could help improve our predictions by reducing noise in both model training  
375 and validation. Additionally, future model development could benefit from additional monitor data,

376 including PurpleAir monitors and CPCB’s manual monitoring stations. Calibration is imperative  
377 for these data as they are not considered as reference grade or regularly calibrated. Calibration of  
378 PurpleAir measurements to estimate  $PM_{2.5}$  concentrations has been widely explored by previous  
379 studies (58–62). A recent study (63) highlights the significance of seasonally-optimized calibra-  
380 tion for PurpleAir sensors in enhancing prediction performance, especially in India. Calibration of  
381 manual monitor measurements is also crucial since  $PM_{2.5}$  samples are collected for 8 hours twice a  
382 week, providing only a snapshot of actual concentrations (5). Moreover, the remotely-sensed input  
383 features utilized in our model, particularly TROPOMI data, do not directly represent air pollution  
384 concentrations at the ground level. A recent study (64) revealed a mean relative and absolute bias  
385 of approximately 10 % between TROPOMI  $NO_2$  products and ground-based observations, high-  
386 lighting a tendency for frequent underestimation of elevated  $NO_2$  levels on the ground (64). The  
387 robust predictive power of TROPOMI features indicates that the calibration of these data could  
388 lead to improved performance of machine learning-based estimations for  $PM_{2.5}$  concentrations, al-  
389 though our machine-learning-based approach is implicitly calibrating these satellite observations to  
390 ground data already. Finally, uncertainty quantification from machine learning models is currently  
391 an active area of research. Subsequent enhancements to  $PM_{2.5}$  estimates may involve more granular  
392 quantification of uncertainty.

393 Another limitation of our study is that we relied on cross-sectional wealth estimates for investi-  
394 gating temporal changes in wealth disparities in  $PM_{2.5}$  exposure. The wealth data we used for India  
395 were derived from machine learning models trained on ground data from 2015 and 2019, and thus  
396 might not capture shifts in the wealth distribution in other years. While these wealth estimates  
397 represent the most comprehensive and up-to-date local-level estimates of income or wealth in India  
398 that we are aware of (3, 54), future improvements to these data could further improve our under-  
399 standing of spatial distribution of and temporal changes in income disparities in  $PM_{2.5}$  exposure  
400 across the country.

401 Our publicly available  $PM_{2.5}$  predictions serve as a platform for evaluating specific policies or  
402 interventions aimed at improving air quality. We utilized our daily  $PM_{2.5}$  estimates in the initial  
403 evaluation of India’s NCAP, finding a limited impact of the program to date in targeted cities. This  
404 null result could be because city-level clean air action plans did not yet have time to take effect,  
405 or because they are not effective, and our  $PM_{2.5}$  data – which can be updated in future years –  
406 offer a platform for understanding which explanation is more likely true. They also offer the critical  
407 opportunity to evaluate other air quality control measures being rolled out across the country, as  
408 well as an opportunity to identify the contribution to local  $PM_{2.5}$  concentrations of emissions from  
409 specific sources such as brick kilns that exhibit distinct spatial and temporal patterns. Finally, our  
410 data could also be used to better assess the health burden on air pollution in the country – a task  
411 often accomplished using existing monthly  $PM_{2.5}$  datasets that are more temporally coarse and less  
412 accurate than the data we produce here.

## 413 MATERIALS AND METHODS

### 414 Model inputs

415 We collect daily  $\text{PM}_{2.5}$  observations from January 1, 2013, to September 30, 2023, from 435  
416 CAAQMS monitors with accessible geo-coordinate information, serving as ground truth for our  
417 machine learning model. We construct two main machine learning models to predict  $\text{PM}_{2.5}$  concen-  
418 trations: the AOD model and the Full model. The input features of the AOD model include MODIS  
419 AOD, meteorology, land cover, and elevation collected from the Google Earth Engine (GEE) plat-  
420 form, along with atmospheric reanalysis data retrieved from NASA’s Earthdata portal. In addition  
421 to these features, the Full model incorporates Sentinel-5P mission’s TROPOMI for  $\text{NO}_2$  and CO,  
422 launched on October 13, 2017, by the European Space Agency. The AOD model is trained on  
423 ground-measured  $\text{PM}_{2.5}$  from January 1, 2013, to September 30, 2023 and used to generate daily  
424  $\text{PM}_{2.5}$  predictions starting from January 1, 2005, which corresponds to the earliest available input  
425 feature for the model. Conversely, the Full model is trained from July 10, 2018, to September 30,  
426 2023 due to the limited availability of TROPOMI features, and used to produce predictions for the  
427 corresponding period. Both  $\text{PM}_{2.5}$  measurements and input features are merged to a consistent 10  
428 km grid for model training and validation. The grid is constructed to cover country borders of the  
429 Republic of India as per survey of India records.

430 From the collected  $\text{PM}_{2.5}$  observations, we exclude values of  $999.99 \mu\text{g}/\text{m}^3$ , the upper detection  
431 limit of CCAQM monitors, as it does not accurately represent the actual concentration on the  
432 ground (65). Additionally, we filter out  $\text{PM}_{2.5}$  measurements if the difference between the rolling  
433 average of the preceding 5 days and the  $\text{PM}_{2.5}$  concentration on the current day is less than 0.05  
434 to ensure that air quality monitors exhibit valid variations in daily concentration changes (i.e. we  
435 remove observations when they were at an unrealistic constant level for over five days). As a result,  
436 4,213 observations (1.2%) were removed. Subsequently, for each 10 km grid, we exclude extreme  
437 outliers identified by an interquartile range (IQR) that fall below 15 times the first quartile or exceed  
438 15 times the third quartile, resulting in the removal of 24 observations. The use of a threshold of  
439 15 times allows us to identify  $\text{PM}_{2.5}$  measurements that significantly deviate from the IQR within  
440 each 10 km grid, and helps retain elevated observations that may reflect local variations, such as  
441 those caused by agricultural fires.

442 The complete list of input features for the AOD and Full models can be found in Table S1.  
443 TROPOMI  $\text{NO}_2$  (tropospheric vertical column of  $\text{NO}_2$ ) (35) and CO (vertically integrated CO col-  
444 umn density) (36) are derived from the Sentinel-5P gridded level 3 product at a 1.11 km resolution.  
445 AOD is collected from the MODIS Multi-angle Implementation of Atmospheric Correction (MAIAC)  
446 Land gridded Level 2 product ( $0.55 \mu\text{m}$ ), produced daily at a 1 km resolution (34). Meteorological  
447 input features comprise the daily mean of temperature and dewpoint temperature at 2 meters, wind

448 speed in the eastward and northward directions, total precipitation, net thermal radiation at the  
449 surface, and surface pressure. These input features are drawn from the daily aggregate of European  
450 Centre for Medium-Range Weather Forecasts (ECMWF) Reanalysis 5th Generation (ERA5) land  
451 hourly assets at an 11.13 km resolution (66). Additionally, the daily mean of wind degree and  
452 relative humidity per 10 km grid is calculated using wind speed in the eastward and northward  
453 directions, temperature, and dewpoint temperature. Furthermore, we incorporate MODIS land  
454 cover type data produced yearly at a 500-meter resolution (67). In addition to MODIS land cover  
455 data, low and high vegetation indices obtained from ECMWF ERA5 are included in the model.  
456 Elevation data is sourced from the Shuttle Radar Topography Mission (SRTM) at a resolution of  
457 approximately 30 meters (68).

458 To account for missing observations in TROPOMI features and AOD, atmospheric reanaly-  
459 sis data such as the Modern-Era Retrospective Analysis for Research and Applications, version  
460 2 (MERRA-2) aerosol optical thickness (AOT at 550nm) (69) and CO (70), as well as Aura  
461 Ozone Monitoring Instrument (OMI) NO<sub>2</sub> (71), are included. Importantly, those missing data  
462 in TROPOMI features and AOD are imputed using machine learning methods before being incor-  
463 porated into the main machine learning model, which predicts PM<sub>2.5</sub> concentrations. Furthermore,  
464 the missingness of TROPOMI NO<sub>2</sub>, CO, and AOD is computed based on the amount of missing  
465 observations for each day, and this information is included as model input. Finally, month and day  
466 of the year, a dummy variable indicating a monsoon season, and centroids of each 10 km grid are  
467 incorporated as model input features. Weekly rolling averages of TROPOMI NO<sub>2</sub> and CO, AOD,  
468 MERRA-2 variables, and meteorological variables are also calculated and included in the model to  
469 capture the potential time dependency between input features and PM<sub>2.5</sub> observations to improve  
470 the model predictability.

## 471 **Imputation of TROPOMI and AOD**

472 For the Full model, 26.0% of TROPOMI NO<sub>2</sub>, 18.7% of TROPOMI CO, and 49.0% of AOD had  
473 missing observations from July 10, 2018, to September 30, 2023 across the country (Figure S24).  
474 In the AOD model, 41.8% of AOD data was missing from January 1, 2013, to September 30,  
475 2023 (Figure S25). These missing observations are predicted using light gradient-boosting machine  
476 (LightGBM) and XGBoost with input features, including MERRA-2 AOT and CO, OMI NO<sub>2</sub>,  
477 meteorology, land cover, elevation, month and day of the year, a dummy variable indicating a  
478 monsoon season, centroids of each 10 km grid, and weekly rolling averages and annual averages of  
479 MERRA-2 variables, as well as meteorological variables (Table S2). We conduct pairwise correlation  
480 analysis for feature selection to ensure no variable is highly correlated ( $R^2 > 0.9$ ) with each other,  
481 resulting in the removal of redundant features and increased efficiency in learning tasks (72).

482 Models are trained on TROPOMI NO<sub>2</sub>, TROPOMI CO, and AOD observations for each machine

483 learning task to predict and impute missing data using randomly-selected training data (2% for  
484 models predicting TROPOMI NO<sub>2</sub> and TROPOMI CO, and 3% for the model predicting AOD)  
485 based on 50 km grid cells, month of the year, and year. For model selection and hyperparameter  
486 tuning, we conduct 5-fold inner spatial CV using training data in one of the 10-fold outer spatial  
487 CV to prevent any final test data from leaking into training tasks during model selection and  
488 hyperparameter tuning. Both the inner and outer spatial CV are conducted based on 50 km grid  
489 to account for spatial auto-correction within the input features, especially MERRA-2 reanalysis  
490 data, which has the most coarse spatial resolution of approximately 50 km (0.5° latitude × 0.625°  
491 longitude). Splitting data into training and test sets based on the 50 km grid, rather than the  
492 more conventional method of random splitting by observation is a more demanding prediction task  
493 because a given monitor can contribute data to both training and test sets in case of random split.  
494 Using coarser spatial blocks, rather than 10 km grid, further increases the difficulty of such a task;  
495 however, spatial CV is a more realistic test of how well the model would perform in predicting  
496 time series of missing observations in a new location with no training data. We fit LightGBM and  
497 XGBoost for each model predicting TROPOMI NO<sub>2</sub>, TROPOMI CO, and AOD while implementing  
498 GridSearchCV to search over the LightGBM and XGBoost hyperparameter ranges (Table S3) and  
499 identify the optimal combination of hyperparameters. As a result, LightGBM was used for predicting  
500 missing TROPOMI NO<sub>2</sub> and TROPOMI CO, and XGBoost was used for predicting missing AOD  
501 (Table S4). The final predictions derived from held-out test data in each of the 10 folds are compared  
502 with observations using the evaluation metrics, such as overall R<sup>2</sup>, within R<sup>2</sup>, and RMSE. The  
503 within R<sup>2</sup> is calculated by regressing observed TROPOMI NO<sub>2</sub>, TROPOMI CO, and AOD on their  
504 respective predicted values while incorporating fixed effects for locations with observations, month  
505 of the year, and year.

506 For the Full model, the predictions of missing TROPOMI NO<sub>2</sub> explained 52%, predictions of  
507 missing TROPOMI CO explained 92%, and predictions of missing AOD explained 82% of out-  
508 of-sample variation (Table S5). For the AOD model, the predictions of AOD explained 82% of  
509 out-of-sample variation (Table S5). We then predicted TROPOMI NO<sub>2</sub>, TROPOMI CO, and AOD  
510 values for all 10 km grid cells over India, and used these imputed variables as input features for  
511 the main machine learning model predicting PM<sub>2.5</sub> concentrations. Binary variables indicating  
512 whether each of TROPOMI NO<sub>2</sub>, TROPOMI CO, and AOD is imputed (0 or 1) is also included.  
513 Finally, we create additional weighted variables by assigning weights to the imputed TROPOMI  
514 NO<sub>2</sub>, TROPOMI CO, and AOD values based on their respective imputation performances (1.0 for  
515 actual observations, 0.5 for imputed TROPOMI NO<sub>2</sub>, 0.9 for imputed TROPOMI CO, and 0.8 for  
516 imputed AOD). The weighted variables are incorporated into the main machine learning model.



## 517 **Model tuning and validation**

518 The Full model is trained and validated on 301,355 daily ground-measured  $\text{PM}_{2.5}$  concentrations  
519 from July 10, 2018, to September 30, 2023. The AOD model is trained on 345,559  $\text{PM}_{2.5}$  observations  
520 from January 1, 2013, to September 30, 2023. Similar to our models for the imputation of TROPOMI  
521 features and AOD, we conduct pairwise correlation analysis for feature selection to ensure no variable  
522 is highly correlated ( $R^2 > 0.9$ ) with each other. For model selection and hyperparameter tuning, we  
523 conduct 5-fold inner spatial CV using training data in one of the 10-fold outer spatial CV. Both the  
524 inner and outer spatial CV are constructed based on 50 km grid. When splitting data into training  
525 and test sets through inner and outer spatial CV, we ensure that each test fold includes a nearly  
526 equal number of 50 km grid cells from each of three environmental regions (Figure S5), identified  
527 based on k-means clustering using imputed TROPOMI  $\text{NO}_2$  and CO, imputed AOD, MERRA-2  
528 AOT and CO, and OMI  $\text{NO}_2$ . This helps balance environmental characteristics, such as urban  
529 versus rural, within training and test data in each fold. For the inner 5-fold CV, each 50 km block  
530 of 10 km grid cells goes into only one of the 5 test folds, and for the outer 10-fold CV, each 50  
531 km block goes into only one of the 10 test folds. We fit LightGBM and XGBoost using the inner  
532 CV while implementing GridSearchCV to search over the hyperparameter ranges and identify the  
533 optimal combination of hyperparameters (Table S6). Based on the model performances using the  
534 inner CV, we selected XGBoost for both Full and AOD models (Table S7). We apply XGBoost to  
535 the outer 10-fold spatial CV, utilizing the RMSE as the objective function.

536 We measure model performance by comparing observed  $\text{PM}_{2.5}$  with model predictions derived  
537 from held-out test data in each of the 10 folds. For the model evaluation, we calculate overall  $R^2$ ,  
538 within  $R^2$ , and RMSE on the held-out test set for each of the 10 folds. Similar to the models  
539 for imputing missing observations in TROPOMI  $\text{NO}_2$ , TROPOMI CO, and AOD, the within  $R^2$   
540 is calculated by regressing observed  $\text{PM}_{2.5}$  concentrations on predicted values while incorporating  
541 fixed effects for locations with observations, month of the year, and year.

## 542 **Sensitivity analysis**

543 As part of the sensitivity analysis, we aim to assess the robustness of the Full model by exposing  
544 it to a more spatially challenging task through the implementation of a 10-fold spatial CV with  
545 larger test blocks based on latitude (Figure S13). Additionally, we conduct another iteration of  
546 the Full model using a 10-fold random CV where data is randomly split into 10 folds without  
547 specific consideration for spatial distribution. This approach enables us to assess and confirm the  
548 potential underestimation of spatial prediction errors and the optimistic overall results associated  
549 with random k-fold CV, which does not account for spatial auto-correction within spatial data.

## 550 **Predictive power of AOD and TROPOMI features**

551 To evaluate and identify the predictive power of TROPOMI features, we construct another model,  
552 the TROPOMI model, which excludes AOD but incorporates other features present in the Full  
553 model. We compare the performance of the Full, AOD, and TROPOMI models by fitting XGBoost  
554 using the same training and test sets from July 10, 2018, to September 30, 2023. Evaluation metrics  
555 include the overall  $R^2$ , within  $R^2$ , and RMSE.

## 556 **Assessing long-term spatiotemporal trends in predicted $PM_{2.5}$ concentrations**

557 Utilizing the  $PM_{2.5}$  predictions derived from the AOD model, We calculate the average  $PM_{2.5}$   
558 concentrations per 10 km grid between 2005 and 2010, and changes in average concentrations  
559 from 2005-2010 to 2011-2016 and to 2017-2022. Employing six-year windows helps control for  
560 meteorological variability between years and mitigates undue influence from extreme years, such as  
561 2020 when India experienced a nationwide lockdown, similar to other countries (49). Additionally,  
562 population-weighted annual average  $PM_{2.5}$  concentrations are computed for India as a whole and  
563 five mega-cities from 2005 to 2022, combining population counts (73) with annual average  $PM_{2.5}$   
564 concentrations per 10 km grid. To elucidate the long-term temporal trend in population-weighted  
565 averages, we also calculate the percentage changes in population-weighted annual average  $PM_{2.5}$   
566 concentrations for each 3-year window from 2006-2008 to 2020-2022 relative to the 3-year average  
567 from 2005-2007.

## 568 **Identifying the contribution of meteorological variability to long-term trends in 569 predicted $PM_{2.5}$ concentrations**

570 To identify the contribution of meteorological variability to the observed declining trend in  $PM_{2.5}$   
571 concentrations since 2016-2018, we model the  $PM_{2.5}$  concentrations of each individual 10 km grid  
572 cell using an additive form of a trend component, a meteorology component, and time fixed effects  
573 (42). More specifically, we employ the following regression equation for each grid cell  $i$ :

$$y_{it} = \beta_i^{\text{obs}} \times t + f_i(X_{it}) + \eta_{it} + \varepsilon_{it}$$

574 where  $y_{it}$  denotes the daily  $PM_{2.5}$  concentration at grid cell  $i$  on day  $t$ .  $t$  is the time index  
575 (e.g.,  $t = 1$  for 1 January 2005,  $t = 2$  for 2 January 2005, and  $t = 32$  for 1 February 2005).  $X_{it}$   
576 denotes the local meteorology variables in grid cell  $i$  on day  $t$ , including temperature and dewpoint  
577 temperature at 2 meters, wind speed in the eastward and northward directions, total precipitation,  
578 and surface pressure.  $\eta_{it}$  is the month-of-year and day-of-month fixed effects to capture daily and  
579 monthly variability in pollutant concentrations that are not related to the meteorological variability  
580 (e.g., seasonal cycle in  $PM_{2.5}$ ).  $\varepsilon_{it}$  is the normally distributed error term.  $\beta_i^{\text{obs}}$  represents the

581 meteorology-corrected daily trend in  $PM_{2.5}$  concentration for grid cell  $i$  estimated with the standard  
582 ordinary least-squares method. We apply the above regression equation to  $PM_{2.5}$  daily predictions  
583 derived from the AOD model for 2005-2015 and 2016-2022, respectively, and obtain the meteorology-  
584 corrected annual trends of  $PM_{2.5}$  concentrations for each grid cell for the corresponding periods by  
585 multiplying the estimated  $\beta_i^{\text{obs}}$  by 365 days.

586 In contrast, the observed annual trend is estimated by the following regression equation for each  
587 grid cell  $i$ :

$$y_{it} = \beta_i^{\text{obs}} \times t + \eta_{it} + \varepsilon_{it}$$

588 where  $y_{it}$  denotes the daily  $PM_{2.5}$  concentration at grid cell  $i$  on day  $t$ .  $t$  is the time index and  
589  $\eta_{it}$  is the month-of-year and day-of-month fixed effects.  $\varepsilon_{it}$  is the normally distributed error term.  
590 Utilizing the  $\beta_i^{\text{obs}}$ , representing the daily trend in predicted  $PM_{2.5}$  concentrations, annual trends for  
591 2005-2015 and 2016-2022 are estimated.

## 592 Evaluation of NCAP

593 Our evaluation of NCAP is based on a matched difference-in-difference analysis, where we first  
594 identify a set of "control" urban areas that were not initially targeted by NCAP that look similar  
595 across a range of covariates to the "treated" cities that were targeted by the program, and then  
596 we compare trends in ambient  $PM_{2.5}$  concentrations between treated and control areas, before and  
597 after program implementation. We implement our analysis at the subdistrict level due to the limited  
598 availability of city-level shapefiles in India. Specifically, we first identify the treatment subdistricts  
599 corresponding to the 102 non-attainment cities whose city clean action plans were approved in 2020.  
600 Next, we calculate the propensity score —the probability of becoming a city targeted by NCAP  
601 — based on population count, proportion of urban area, proportion of forest area, and total area  
602 per subdistrict using logistic regression. We apply this to non-treated subdistricts that are not  
603 adjacent to the treatment subdistricts and are located outside a 50 km radius from them to account  
604 for potential spillover effects of NCAP on neighboring subdistricts. Finally, we identify control  
605 subdistricts as those with propensity closest to those of the treatment subdistricts. Propensity  
606 score methods are commonly used to minimize selection bias in identifying control groups that share  
607 observed baseline characteristics with treatment groups as similar as possible (74). Consequently,  
608 our analysis includes 88 treatment subdistricts and 74 control subdistricts (Figure S20).

609 To establish the causal effect of NCAP, the trends in  $PM_{2.5}$  concentrations in subdistricts that  
610 do not include NCAP's non-attainment cities must provide valid counterfactuals for the trends that  
611 we would have observed in the treatment subdistricts. We evaluate whether the parallel trends  
612 assumption may be reasonable in our case by plotting yearly average treatment effects on  $PM_{2.5}$   
613 concentrations prior to 2020, confirming that there was no systematic difference in treatment and

614 control groups before city action plans were approved by the CPCB in 2020. More specifically, we  
 615 employ the following regression equation to estimate the average treatment effects for each year  
 616 from 2005 to 2022:

$$y_{it} = \sum_{j=t-15}^{t+2} \beta_j D_i * 1(\text{year} = j) + \alpha_i + \gamma_t + \theta x_{it} + \varepsilon_{it}$$

617 where  $y_{it}$  denotes the daily  $\text{PM}_{2.5}$  concentration at subdistrict  $i$  on day  $t$ , and the  $\beta_j$  coefficients  
 618 estimate the difference between treated and control subdistricts in the 15 years prior to treatment  
 619 ( $t = 0$ , which is 2020 in our data, the year in which city clean action plans were approved by the  
 620 CPCB), and the two observed years following treatment (2021 and 2022). The estimate in  $t = 0$   
 621 is omitted to avoid collinearity, and so the  $\beta_j$  coefficients are interpreted as the difference between  
 622 treated and control concentrations relative to the reference year of 2020.  $\alpha_i$  represents a vector of  
 623 subdistrict fixed effects, and  $\gamma_t$  vectors of month-of-year and year fixed effects.  $\theta x_{it}$  denotes a vector  
 624 of additional time-varying controls at the subdistrict level, including temperature and dewpoint  
 625 temperature at 2 meters, wind speed in the eastward and northward directions, total precipitation,  
 626 and surface pressure.  $\varepsilon_{it}$  is the error term. Because our chosen treated units all adopted city clean  
 627 action plans in the same year, our treatment is not staggered across time and thus our analysis  
 628 avoids the inference issues that accompany difference-in-difference designs with staggered adoption  
 629 that have been highlighted in recent literature.

### 630 **Investigating population exposure to $\text{PM}_{2.5}$ concentrations**

631 To identify locations with elevated levels of daily  $\text{PM}_{2.5}$  concentrations, we employ predictions from  
 632 the AOD model to calculate the average number of days each 10 km grid cell exceeding the WHO  
 633 daily guideline of  $15 \mu\text{g}/\text{m}^3$ , national daily guideline of  $60 \mu\text{g}/\text{m}^3$ , and extreme value of  $100 \mu\text{g}/\text{m}^3$   
 634 for 2018-2022.

635 Furthermore, we examine proportional changes in populations exposed to high levels of  $\text{PM}_{2.5}$   
 636 annual average concentrations from 2005 to 2022. For this analysis, we firstly calculate annual  
 637 average  $\text{PM}_{2.5}$  concentrations per 10 km grid. Subsequently, we identify the 10 km grids above  
 638 the thresholds of the WHO annual guideline ( $5 \mu\text{g}/\text{m}^3$ ), national annual guideline ( $40 \mu\text{g}/\text{m}^3$ ), and  
 639 extreme annual concentration ( $80 \mu\text{g}/\text{m}^3$ ). We then aggregate the population counts residing in  
 640 those grid cells exceeding each threshold to calculate the percentage of whole population.

### 641 **Examining inequalities in $\text{PM}_{2.5}$ exposure**

642 We combine estimates of relative wealth (Relative Wealth Indices (RWIs)) at a 2.4 km resolution  
 643 (54) with our  $\text{PM}_{2.5}$  predictions derived from the AOD model by averaging the wealth estimates  
 644 for each 10 km grid. RWIs were generated using machine learning algorithms that utilized satellite

645 data, mobile phone data, and data from Facebook users (54). For India, their machine learning  
646 algorithms were trained on ground measurements of wealth collected by DHS surveys in 2015 and  
647 2019 (54). The gridded wealth data provide estimates for each grid cell of how wealthy that grid  
648 cell is relative to others in the same country as well as an estimated error for that estimate (3). We  
649 further combine population counts (73) for each grid, along with RWIs and daily PM<sub>2.5</sub> predictions  
650 from January 2005 to December 2022.

651 To understand the proportion of population exposed to elevated levels of PM<sub>2.5</sub> concentrations  
652 across wealth groups, we aggregate the population counts for 10 km grid cells whose average PM<sub>2.5</sub>  
653 concentrations for 2018-2022 are exceeding the national guideline of 40  $\mu\text{g}/\text{m}^3$  and extreme threshold  
654 of 80  $\mu\text{g}/\text{m}^3$  annually, respectively. We then calculate the percentage of population for each of the  
655 five wealth categories (Poorest, Poorer, Middle, Richer, and Richest), which are created based  
656 on quartiles of RWIs within the country. Additionally, to understand the temporal changes in  
657 wealth disparities in PM<sub>2.5</sub> exposure from 2005 to 2022, population-weighted annual average PM<sub>2.5</sub>  
658 concentrations are computed for all the wealth levels across the country, grid cells at 90th percentile  
659 of RWIs, and grid cells at 10th percentile of RWIs. To elucidate the long-term temporal trend  
660 in population-weighted averages, we also calculate the percentage changes in population-weighted  
661 annual average PM<sub>2.5</sub> concentrations for each 3-year window from 2006-2008 to 2020-2022 relative  
662 to the 3-year average from 2005-2007.

### 663 **Assessing the optimal placement of air quality monitors ensuring equality**

664 To calculate where additional ground monitors could be placed to optimally enhance the ability of  
665 the entire ground-monitoring network to capture spatial and temporal variation in ambient PM<sub>2.5</sub>  
666 concentrations, we use multi-resolution dynamic mode decomposition (mrDMD), which recursively  
667 decomposes a data set into low-rank spatial modes and their temporal Fourier dynamics. mrDMD  
668 has been shown to capture PM<sub>2.5</sub> concentrations spatially and temporally on short (daily) and  
669 long-term (years to decade) timescales, and to incorporate information from transient phenomena,  
670 such as wildfires and temperature inversions, that would otherwise be discarded or averaged out  
671 using similar data reduction techniques (37). The algorithm can thus capture a finer level of spatial  
672 and temporal variability in a data set that would otherwise be averaged out using traditional mean  
673 or maximum PM<sub>2.5</sub> metrics. mrDMD is a dimensionality reduction algorithm, similar to Principal  
674 Components Analysis (PCA), but mrDMD is more precise in capturing spatio-temporal variability  
675 than methods based on singular value decomposition such as PCA.

676 Here, we present an extension to the mrDMD framework that considers cost-constraining func-  
677 tions to optimize sensor placement based on wealth estimates. The mrDMD algorithm is based on  
678 a column-pivoted QR algorithm, where the pivot column is modified to balance (a) the decrease  
679 in accuracy of capturing the largest air pollution modal signals with (b) the increase in capturing

680 pollution exposure in grid cells with low RWI values, representing poorer communities. The cost  
681 function used here is a step function that penalizes placing sensors too far from poorer locations.  
682 For the cost function vector, all data is a binary of 0 or 1, with 0 representing wealthy locations  
683 and 1 representing poor locations. More details of the method can be found in (55).

684 We apply all mrDMD methods to the  $PM_{2.5}$  estimates derived from the AOD model from  
685 January 1, 2005, to September 30, 2023. The resulting sensor network adds additional monitors  
686 to the existing ground network in order to better constrain variation in surface pollution, and we  
687 select a number of additional monitors such that the entire monitoring network would have 1,000  
688 ground monitors, an NCAP’s target.

### 689 **Examining applicability of our model to other low- and middle-income settings**

690 To investigate the association between the quantity of ground truth data, represented by the number  
691 of 10 km grid cells with measured  $PM_{2.5}$  observations, and the predictive performance of our machine  
692 learning model for  $PM_{2.5}$  concentrations, we conduct an experiment involving multiple machine  
693 learning models trained with incremental training data. First, 21 out of 321 grid cells with  $PM_{2.5}$   
694 observations across the country are randomly selected and held out until the final evaluation of  
695 predictions. Second, from the remaining 300 grid cells, we randomly select grid cells in increments  
696 of 25, starting from 25 and up to 300 without replacement, respectively serving as training data for  
697 our Full model. This process result in the creation of 11 Full models with training data of different  
698 sizes. Each training dataset is then split into training and validation sets based on 50 km grid cells,  
699 creating a 10-fold spatial CV. For each model, XGBoost is applied with hyperparameter tuning,  
700 where the parameters include `learning_rate` (set to 0.01), `lambda` (1 or 10), `max_depth` (fixed at 10),  
701 and `n_estimators` (1,000 or 1,500), while keeping the remaining hyperparameters at their default  
702 values. The tuning process is performed using a 10-fold spatial CV. Finally, XGBoost with the  
703 best hyperparameters is applied to predict  $PM_{2.5}$  concentrations for the initially randomly-selected  
704 21 grid cells, and the final predictions are compared and evaluated with the test data. This entire  
705 process, from the random sampling without replacement of the 21 grid cells for the test dataset to  
706 making final predictions using the best hyperparameters for the 11 Full models, is repeated 1,000  
707 times to obtain the best possible unbiased estimates of prediction performances. The mean within  
708  $R^2$  is calculated out of 1,000 samples for each of the 11 Full models.

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## 720 **Author contributions**

721 A.K., M.K., M.Q., and M.B. designed research; A.K. performed research; A.K., and M.K. analyzed  
722 data; and A.K., M.K., M.Q., K.S., E.C., and M.B. wrote the paper.

## 723 **Competing interests**

724 The authors declare no conflict of interest.

## 725 **Data and materials availability**

726 Our daily PM<sub>2.5</sub> datasets derived from the Full and AOD models have been deposited in Zenodo  
727 (<https://doi.org/10.5281/zenodo.10807119>) and are publicly available. Code and source data  
728 needed to replicate the results have been deposited in GitHub ([https://github.com/ayako-kawano/  
729 pm\\_prediction](https://github.com/ayako-kawano/pm_prediction)).

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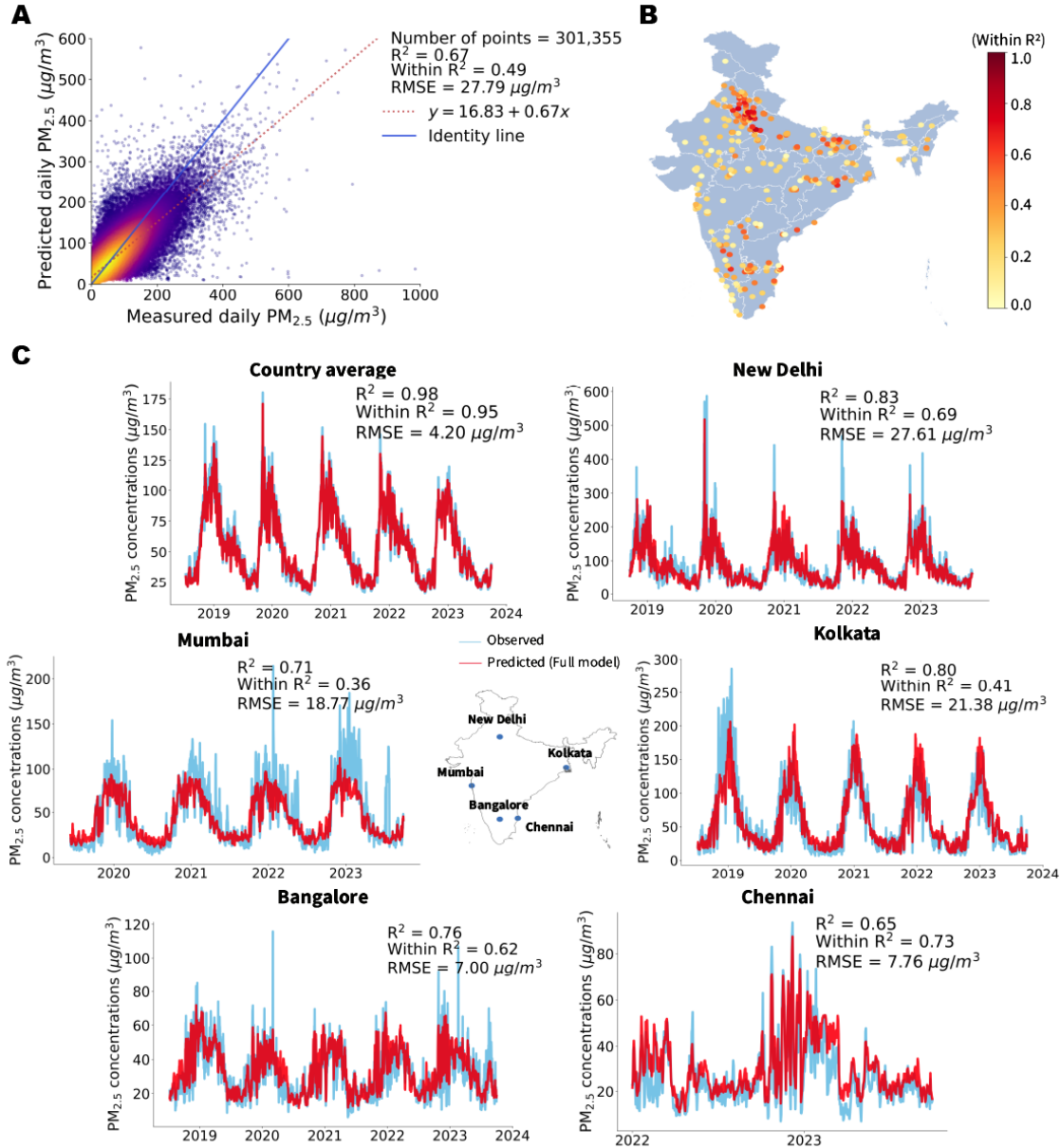


Figure 1: **Model performs well out-of-sample and across the range of observed  $\text{PM}_{2.5}$  concentrations.** Panel A: the overall performance of the Full model. Colors indicate the count of monitor observations, with observed  $\text{PM}_{2.5}$  on the horizontal axis and predicted  $\text{PM}_{2.5}$  concentrations on the vertical axis. The blue line represents the 1–1 line, indicating perfect match between predictions and observations. Panel B: out-of-sample model performance in each grid cell with at least one monitor reporting on at least 5 days. Performance measured using "within  $R^2$ " after removing local seasonality and year trends. It is calculated over observed  $\text{PM}_{2.5}$  using predicted  $\text{PM}_{2.5}$  from the Full model in which that station was out-of-sample, with month of the year and year fixed effects. Panel C: the out-of-sample performance of the Full model in predicting daily time-series of observed  $\text{PM}_{2.5}$  concentrations for the entire country, New Delhi, Mumbai, Bangalore, Chennai, and Kolkata. Different y-axis and x-axis scales are used in the figures to accommodate variations in  $\text{PM}_{2.5}$  concentrations and monitor availability across the cities.

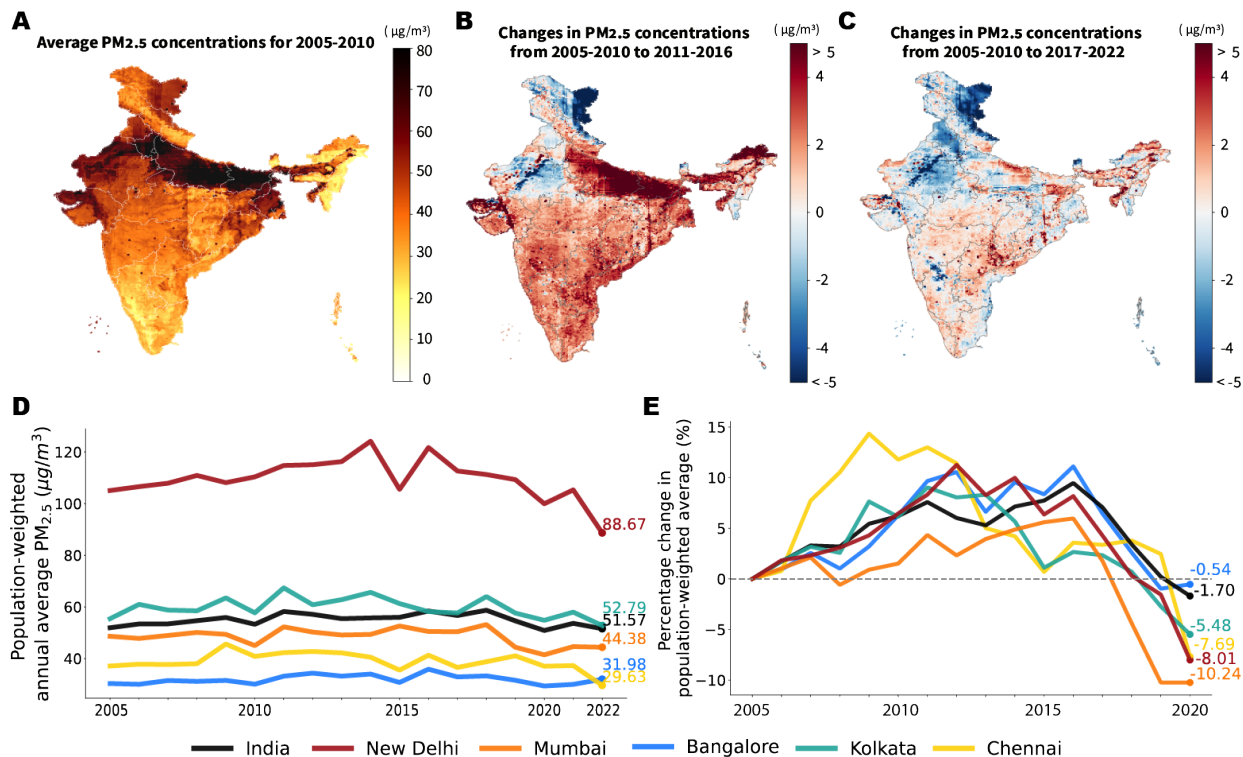
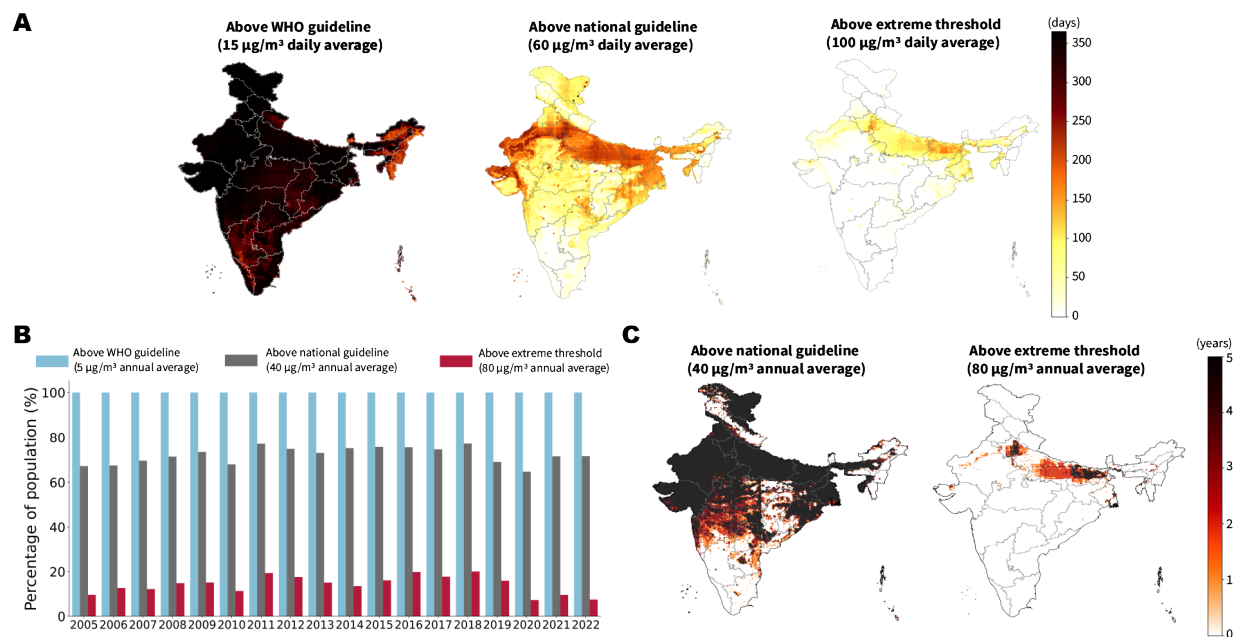


Figure 2: **PM<sub>2.5</sub> concentrations increased through 2016 but then declined through much of India thereafter.** Panel A: 6-year average PM<sub>2.5</sub> concentrations for 2005-2010 computed as the average over all days in each grid cell for 6 years. Panel B: changes in PM<sub>2.5</sub> concentrations between 2005-2010 and 2011-2016 show increases across most of the country. Panel C: changes in PM<sub>2.5</sub> concentrations between 2005-2010 and 2017-2022 show a mix of increases and declines. Panel D: population-weighted annual average PM<sub>2.5</sub> concentrations from 2005 to 2022, for all of India and selected mega-cities. Panel E: percentage changes in 3-year population-weighted averages relative to the 2005-2007 average, for all of India and selected mega-cities. The x-axis label represents the running means of years from 2005-2007 to 2020-2022.



**Figure 3: Widespread exposure to elevated  $PM_{2.5}$  concentrations across India, with daily extremes in the North.** Panel A: spatial distribution of grid cells with daily  $PM_{2.5}$  concentrations above WHO guideline of  $15 \mu\text{g}/\text{m}^3$  (53) (left panel), national guideline of  $60 \mu\text{g}/\text{m}^3$  (center panel), and daily extreme threshold of  $100 \mu\text{g}/\text{m}^3$  (right panel), demonstrating the average number of days for each 10 km grid cell for 2018-2022. Panel B: proportion of populations exposed to annual averages of  $PM_{2.5}$  concentrations exceeding WHO ( $5 \mu\text{g}/\text{m}^3$ ) (53), national ( $40 \mu\text{g}/\text{m}^3$ ), and extreme ( $80 \mu\text{g}/\text{m}^3$ ) thresholds from 2005 to 2022. The entire population in India is consistently exposed to  $PM_{2.5}$  concentrations above the WHO guideline during these years. Panel C: Locations highlighted in colors indicate areas where average  $PM_{2.5}$  concentrations exceeded the national guideline of  $40 \mu\text{g}/\text{m}^3$  (left panel) and extreme threshold of  $80 \mu\text{g}/\text{m}^3$  (right panel) from 2018 to 2022, respectively. Colored gradients depict the number of years each grid cell exceeded each threshold during these years. The entire country would be shaded in colors on a map denoting the areas exposed to above the WHO threshold.



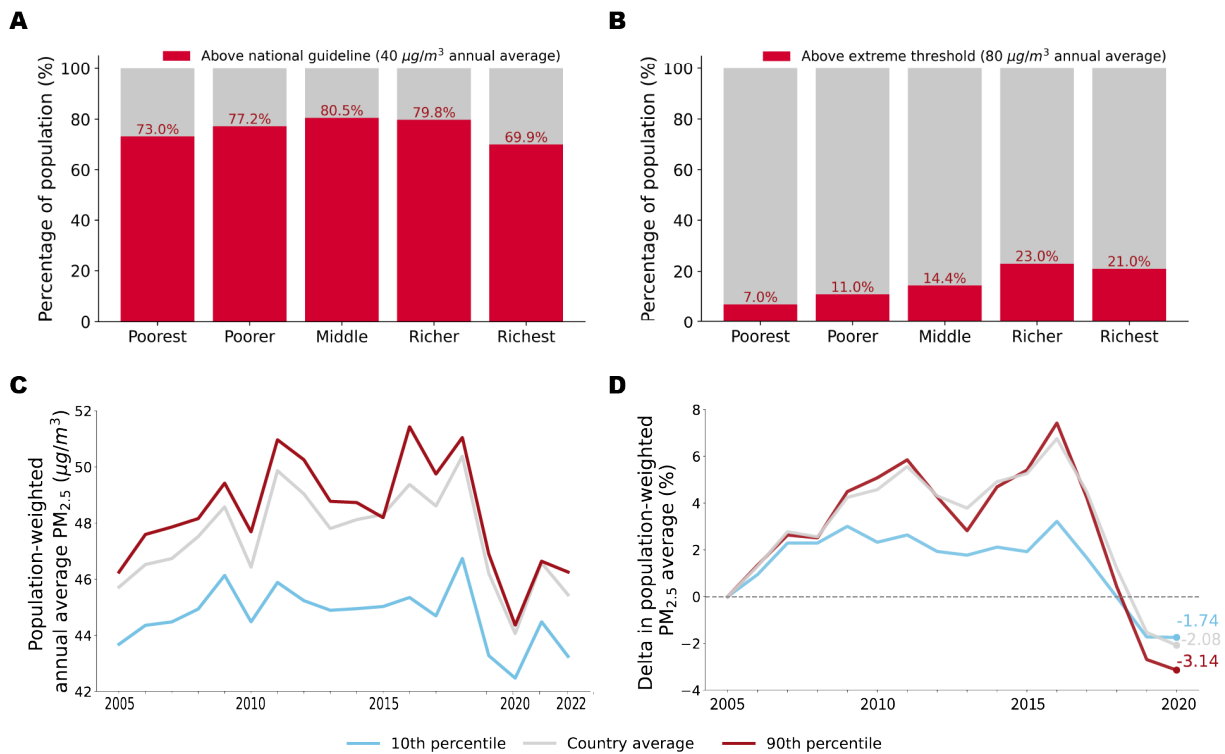


Figure 4: **Wealthier people experienced higher average  $\text{PM}_{2.5}$  concentrations but faster recent declines.** Panel A: percentage of the population in each wealth category exposed to ambient  $\text{PM}_{2.5}$  concentrations above the national guideline of  $40 \mu\text{g}/\text{m}^3$  for the years 2015-2019. These percentages were calculated using 5-year mean  $\text{PM}_{2.5}$  concentrations from 2015 to 2019 because machine learning algorithms to generate wealth estimates (54) were trained on ground data from that period. Panel B: percentage of the population in each wealth category exposed to concentrations above the extreme threshold of  $80 \mu\text{g}/\text{m}^3$  for the years 2015-2019. Panel C: population-weighted annual average of  $\text{PM}_{2.5}$  concentrations from 2005 to 2022 for the country average, locations with 90th percentile of wealth, and locations with 10th percentile of wealth. Panel D: percentage changes in 3-year population-weighted averages relative to the 2005-07 average. The x-axis label represents the running means of years from 2005-2007 to 2020-2022. Between 2020 and 2022, wealthier individuals experienced a 3.14% decline in  $\text{PM}_{2.5}$  exposure compared to the period of 2005-2007, while poorer individuals saw a 1.74% reduction. When compared to the period of 2015-2017, during which much of India began to experience an overall declining trend in  $\text{PM}_{2.5}$  concentrations, a wider disparity in reduction rates was observed, with the wealthiest experiencing an 8.11% reduction and the poorest experiencing a 3.60% reduction S23.

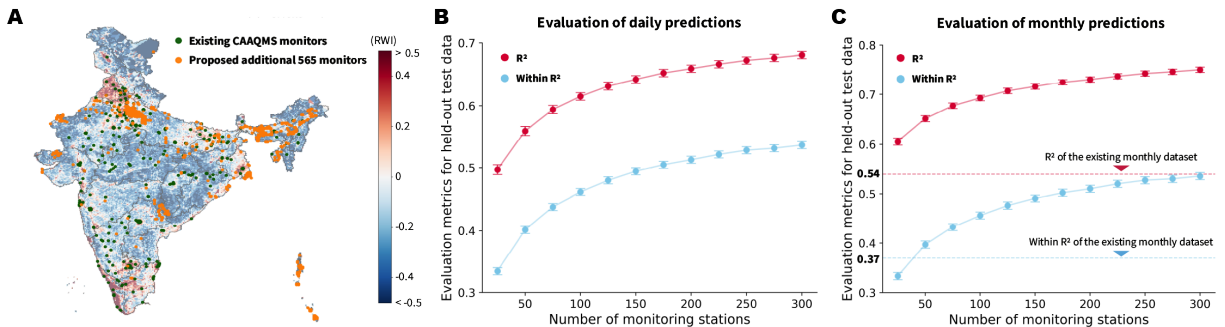


Figure 5: **Applications of our predictions for better air quality monitoring in India and other low- and middle-income countries.** Panel A: proposed locations of additional 565 CAAQMS identified by compressed sensing methods (see Materials and Methods), along with existing 435 monitor locations, ensuring increased equality in air quality monitoring across the country while capturing both baseline patterns and sudden anomaly spikes in  $PM_{2.5}$  concentrations. Colored gradients indicate wealth estimates (Relative Wealth Indices (RWIs) (54)) in each grid cell. Panel B: model performance at a daily level as a function of number of stations used in training. Performance is measured by  $R^2$  and "within  $R^2$ " as evaluated on a fixed number of held out stations (21 stations). Points represent the mean performance across 1,000 experiments (randomly re-sampling training and test sets) and whiskers denote the corresponding 95 % confidence intervals across experiments. Model performance increases with additional training data, but at a declining rate, with modest increases past 150 monitors. Panel C: model performance at a monthly level, estimated by aggregating the daily predictions and monitor data on a monthly basis. The dashed lines represent the  $R^2$  and within  $R^2$  values of the existing publicly-available monthly  $PM_{2.5}$  dataset, evaluated against monitor data in India, which are 0.54 and 0.37, respectively.

919 **Supplementary tables**

Table S1: Input features used for the Full and AOD models.

Feature	Temporal scale	Source	Native resolution
TROPOMI NO <sub>2</sub> <sup>*/**</sup>	daily, weekly rolling average	Sentinel-5P	1.11 km
TROPOMI CO <sup>*/**</sup>	daily, weekly rolling average	Sentinel-5P	1.11 km
AOD <sup>**</sup>	daily, weekly rolling average	MODIS MAIAC	1 km
AOT	daily, weekly rolling average	MERRA-2	50 km
CO (reanalysis data)	daily, weekly rolling average	MERRA-2	50 km
NO <sub>2</sub> (reanalysis data)	weekly rolling average <sup>***</sup>	OMI/Aura	25 km
Meteorology (temperature at 2 meters, dewpoint temperature at 2 meters, wind speed in the east ward and northward directions, total precipitation, net thermal radiation at the surface, surface pressure, relative humidity <sup>***</sup> , wind degree <sup>***</sup> )	daily, weekly rolling average	ECMWF ERA5	11.13 km
Land cover (water, shrub, urban, forest, savannas)	yearly	MODIS	500 meters
Land cover (low vegetation, high vegetation)	daily	ECMWF ERA5	11.13 km
Elevation	cross-sectional (as of 2000)	NASA SRTM	30 meters
Month and day of the year	daily	-	-
Binary variable indicating monsoon season (0 or 1)	daily	-	-
Latitude and longitude	cross-sectional	-	-
Percentage of missing observations in TROPOMI NO <sub>2</sub> <sup>*</sup> , TROPOMI CO <sup>*</sup> , AOD	daily	-	-
Binary variables indicating whether each of TROPOMI NO <sub>2</sub> <sup>*</sup> , TROPOMI CO <sup>*</sup> , AOD is imputed (0 or 1)	daily	-	-
Weighted TROPOMI NO <sub>2</sub> <sup>*</sup> (NO <sub>2</sub> values multiplied by 1.0 for observations, 0.5 for imputed values)	daily	-	-
Weighted TROPOMI CO <sup>*</sup> (CO values multiplied by 1.0 for observations, 0.9 for imputed values)	daily	-	-
Weighted AOD (AOD values multiplied by 1.0 for observations, 0.8 for imputed values)	daily	-	-

\*: Only included in the Full model.

\*\* : Missing observations are imputed using machine learning methods before being incorporated into the model.

\*\*\* : Calculated using the meteorological data derived from ECMWF ERA5.

\*\*\*\* : Only weekly rolling average is computed and included due to the missing data in daily observations.

Table S2: Input features used for machine learning models for imputation of missing data in TROPOMI NO<sub>2</sub>, TROPOMI CO, and AOD.

Feature	Temporal scale	Source	Native resolution
AOT	daily, weekly rolling average, annual average	MERRA-2	50 km
CO (reanalysis data)	daily, weekly rolling average, annual average, average of all days	MERRA-2	50 km
NO <sub>2</sub> (reanalysis data)	weekly rolling average, annual average**	OMI/Aura	25 km
Meteorology (temperature at 2 meters, dewpoint temperature at 2 meters, wind speed in the east ward and northward directions, total precipitation, net thermal radiation at the surface, surface pressure, relative humidity *, wind degree *)	daily, weekly rolling average, annual average	ECMWF ERA5	11.13 km
Land cover (water, shurub, urban, forest, savannas)	yearly	MODIS	500 meters
Land cover (low vegetation, high vegetation)	daily	ECMWF ERA5	11.13 km
Elevation	cross-sectional (as of 2000)	NASA SRTM	30 meters
Month, day of the year, cosine of day of the year	daily	-	-
Binary variable indicating monsoon season (0 or 1)	daily	-	-
Latitude and longitude	cross-sectional	-	-

\*: Calculated using the meteorological data derived from ECMWF ERA5.

\*\* : Only weekly rolling average and annual average are computed and included due to the missing data in daily observations.

Table S3: Hyperparameter ranges explored within inner 5-fold spatial CV using GridSearchCV for models imputing missing observations in TROPOMI NO<sub>2</sub>, TROPOMI CO, and AOD.

Algorithm	Hyperparameters	Range
LightGBM	max_depth	3, 5, 8, 10, 15, 20
	learning_rate	0.01, 0.1
	num_iterations	500, 800, 1000, 1500, 3000
	num_leaves	800, 1000, 1500
	max_bin	255, 350, 500, 1000
	min_data_in_leaf	10, 20, 30, 40, 50
	lambda_l2	0, 1, 10, 100, 500
	boosting	gbdt
	XGBoost	max_depth
	learning_rate	0.01, 0.1
	gamma	0.2, 0.8, 1.0
	subsample	0.2, 0.8, 1.0
	min_child_weight	0.2, 0.8, 1.0
	n_estimators	500, 1000, 1500
	lambda	0, 1, 10, 100, 500
	booster	gbtree

Table S4: Optimal hyperparameters and corresponding out-of-sample predictive performances obtained through GridSearchCV using a 5-fold inner spatial CV for each model imputing missing observations in TROPOMI NO<sub>2</sub>, TROPOMI CO, and AOD.

Model	Algorithm	Hyperparameters	Results		
Imputation for missing TROPOMI NO <sub>2</sub>	LightGBM	max_depth: 10	R <sup>2</sup> : 0.50		
		learning_rate: 0.1	RMSE: $1.82 \times 10^{-5}$		
		num_iterations: 3000			
		num_leaves: 1500			
		max_bin: 500			
		min_data_in_leaf: 10			
		lambda_l2: 10			
		boosting: gbdt			
		Imputation for missing TROPOMI CO	LightGBM	max_depth: 10	R <sup>2</sup> : 0.92
				learning_rate: 0.1	RMSE: 0.003
num_iterations: 3000					
num_leaves: 1500					
max_bin: 1000					
min_data_in_leaf: 10					
lambda_l2: 10					
boosting: gbdt					
Imputation for missing AOD	XGBoost			max_depth: 20	R <sup>2</sup> : 0.79
				learning_rate: 0.1	RMSE: 166.26
		gamma: 0.8			
		subsample: 0.8			
		min_child_weight: 1			
		n_estimators: 1000			
		lambda: 100			
		booster: gbtree			

Table S5: Out-of-sample performances of machine learning models for imputing missing observations in TROPOMI NO<sub>2</sub>, TROPOMI CO, and AOD.

Model	R <sup>2</sup>	Within R <sup>2</sup>	RMSE	Number of observations
TROPOMI NO <sub>2</sub>	0.52	0.27	$2 \times 10^{-5}$ (mol m <sup>2</sup> )	934,138
TROPOMI CO	0.92	0.75	0.003 (mol m <sup>2</sup> )	1,026,271
AOD (Full model)	0.82	0.76	155.21	965,224
AOD (AOD model)	0.82	0.75	147.23	1,264,830

Table S6: Hyperparameter ranges explored within inner 5-fold spatial CV using GridSearchCV for the Full and AOD models.

Algorithm	Hyperparameters	Range
LightGBM	max_depth	5, 8, 10, 15
	learning_rate	0.01, 0.1
	num_iterations	800, 1000, 1500
	num_leaves	800, 1023, 1500
	max_bin	255, 350, 500
	min_data_in_leaf	10, 20, 30, 40, 50
	lambda_l2	0, 1, 10, 100, 500
	boosting	gbdt
XGBoost	max_depth	5, 8, 10
	learning_rate	0.01, 0.1
	gamma	0.2, 0.8, 1.0
	subsample	0.2, 0.8, 1.0
	min_child_weight	0.2, 0.8, 1.0
	n_estimators	500, 1000, 1500
	lambda	0, 1, 10, 100, 500
	booster	gbtree



Table S7: Optimal hyperparameters and corresponding out-of-sample predictive performances obtained through GridSearchCV using a 5-fold inner spatial CV for the Full and AOD models.

Model	Algorithm	Hyperparameters	Results		
Full model	XGBoost	max_depth: 10	R <sup>2</sup> : 0.60		
		learning_rate: 0.1	RMSE: 29.12		
		gamma: 0.8			
		subsample: 0.8			
		min_child_weight: 0.8			
		n_estimators: 1500			
		lambda: 1			
		booster: gbtree			
		AOD model	XGBoost	max_depth: 10	R <sup>2</sup> : 0.60
				learning_rate: 0.1	RMSE: 29.37
		gamma: 0.8			
		subsample: 0.8			
		min_child_weight: 0.8			
		n_estimators: 1500			
		lambda: 1			
		booster: gbtree			

920 **Supplementary figures**

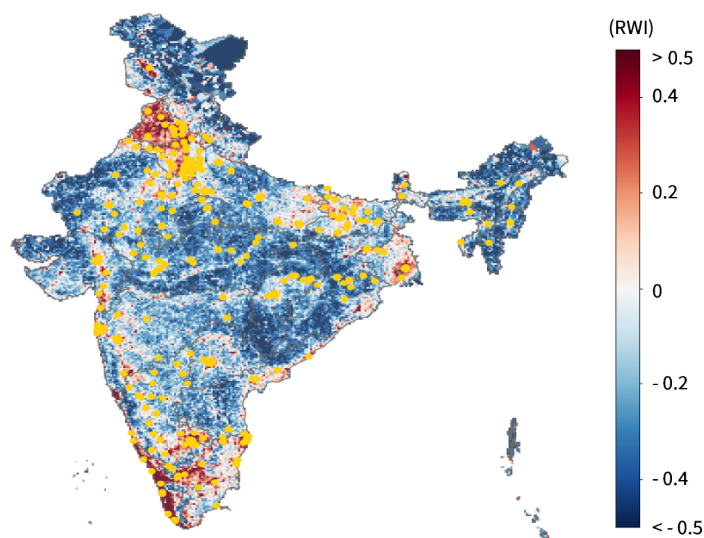


Figure S1: Location of CAAQMS monitors ( $n = 435$ ) mapped with Relative Wealth Indices (54) (RWIs), which represent the relative wealth of each grid cell compared to others in the same country.

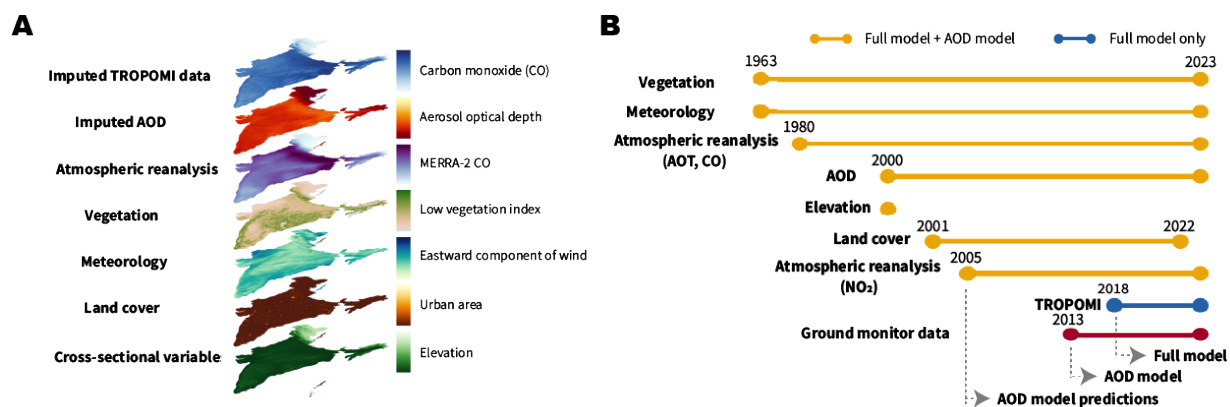


Figure S2: Panel A: input features utilized in the second-stage machine learning model predicting ambient PM<sub>2.5</sub> concentrations include both time-varying and cross-sectional variables. Gap-filled TROPOMI data are exclusively used for the Full model. Panel B: temporal availability of input features employed for the second-stage machine learning model. Due to the availability of TROPOMI data, the Full model is trained on ground monitor data from July 10, 2018, to September 30, 2023, and used to generate daily PM<sub>2.5</sub> predictions for the corresponding period. Additionally, the AOD model is trained from January 1, 2013, to September 30, 2023. The oldest availability of one of the input features, atmospheric reanalysis data (NO<sub>2</sub>), allows the AOD model to generate PM<sub>2.5</sub> estimates from January 1, 2005, to September 30, 2023.

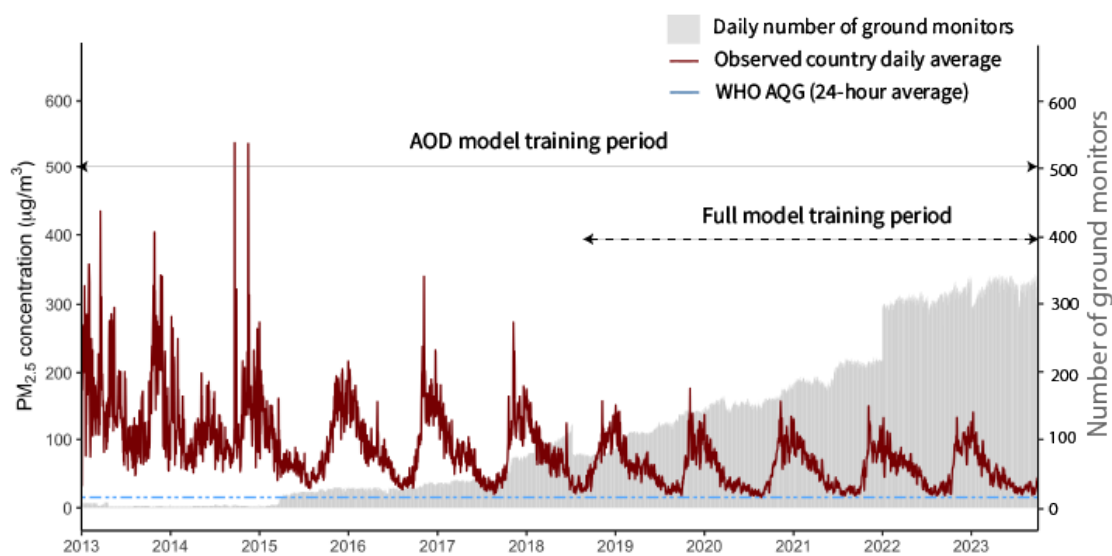


Figure S3: Daily country average of ground-measured  $\text{PM}_{2.5}$  concentrations demonstrates a slightly declining trend over the decade; however, it remains at an endangering level, exceeding the 24-hour average of the World Health Organization (WHO) air quality guideline of  $15 \mu\text{g}/\text{m}^3$  (53). The number of air quality monitors has progressively increased over time, aligning with government efforts to expand the CAAQMS network. Two dashed lines represent the training periods for the Full and AOD models, respectively.

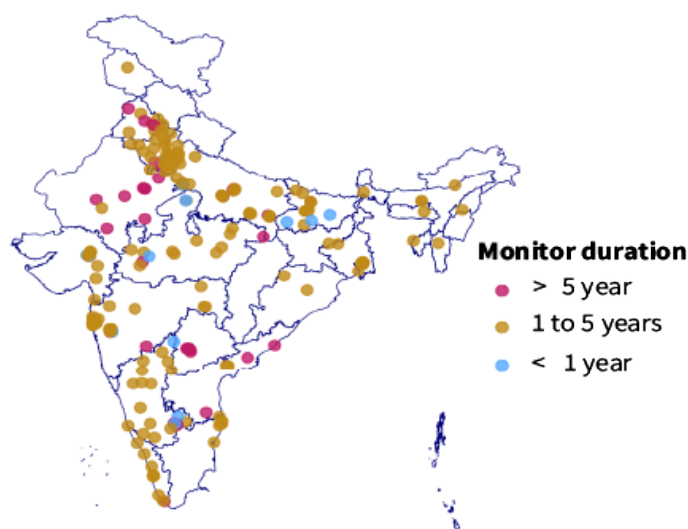


Figure S4: The location of 435 CAAQMS monitors used for this study, with data coverage ranging from less than 1 year to a maximum of 9 years of monitor data.

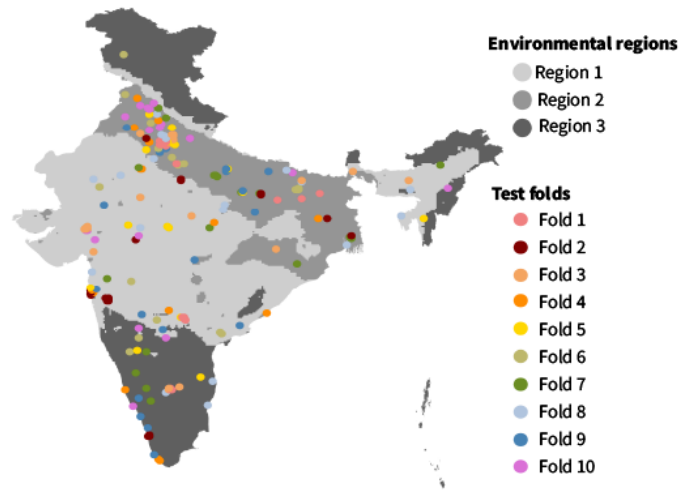


Figure S5: To balance environmental characteristics, such as urban versus rural areas, when splitting data into training and test sets using spatial 10-fold CV, three environmental regions are identified based on k-means clustering, utilizing imputed TROPOMI features, imputed AOD, and atmospheric reanalysis data. Each test fold includes a nearly equal number of 50 km blocks of 10 km grid cells from each of the three environmental regions. Each 50 km block of 10 km grid cells goes into only one of the 10 test folds.

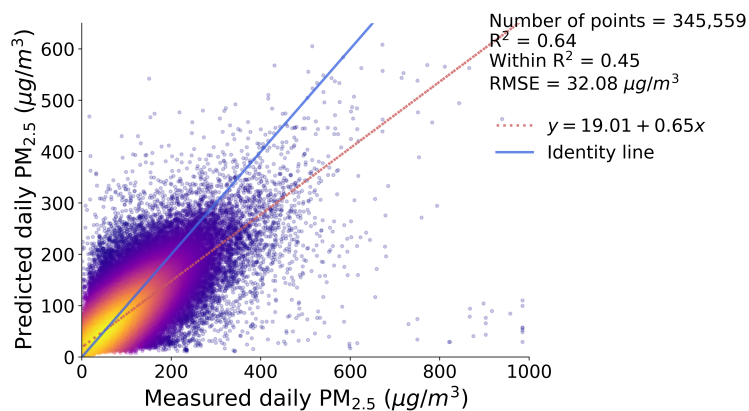


Figure S6: Performance evaluation of the AOD model, trained on  $PM_{2.5}$  measurements from January 1, 2013, to September 30, 2023.

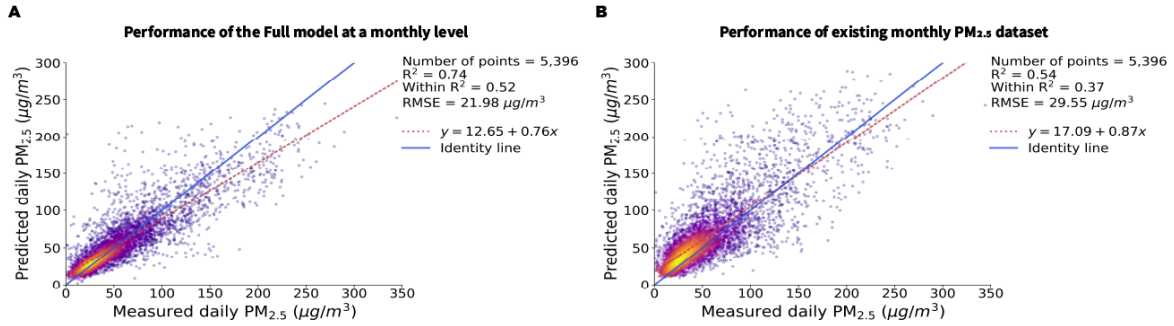


Figure S7: Panel A: monthly performance of the Full model using PM<sub>2.5</sub> daily observations and predictions aggregated at a monthly level from July 10, 2018, to December 31, 2021. Panel B: performance evaluation of the existing PM<sub>2.5</sub> monthly dataset (14), from July 2018, to December 2021, representing their latest available dataset, using the same monthly PM<sub>2.5</sub> observations as employed in the assessment of the Full model.

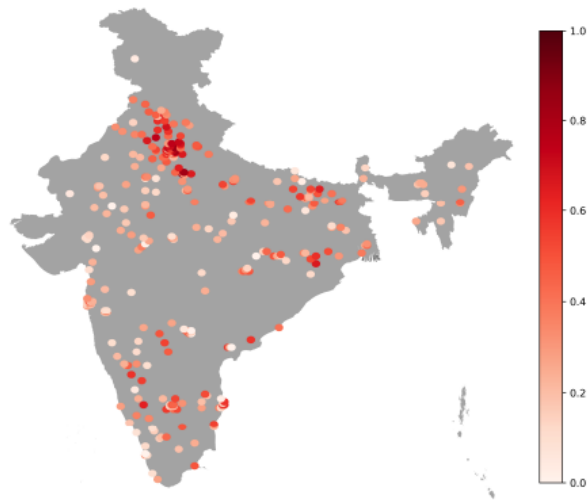


Figure S8: Out-of-sample within  $R^2$  per 10 km by 10 km grid cell, calculated over PM<sub>2.5</sub> observations using PM<sub>2.5</sub> predictions derived from the AOD model for each location with at least 5 observations (points on map).

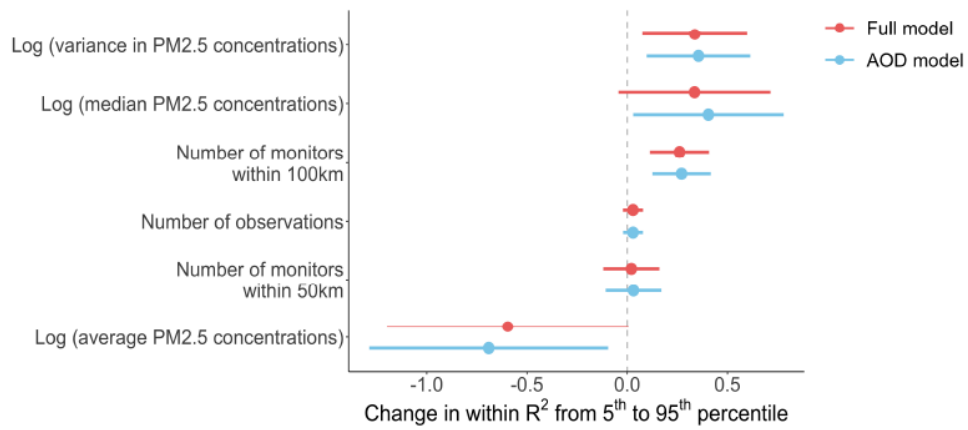


Figure S9: Linear regression results relating monitor-specific performance (within  $R^2$ ) shown in (Fig 1B)) to location characteristics. Predictive power of each characteristic is calculated as the estimated change in monitor within  $R^2$  when each characteristic is increased from the 5th to 95th percentile of its distribution. Points show central estimates, and line segments show 95% confidence intervals.

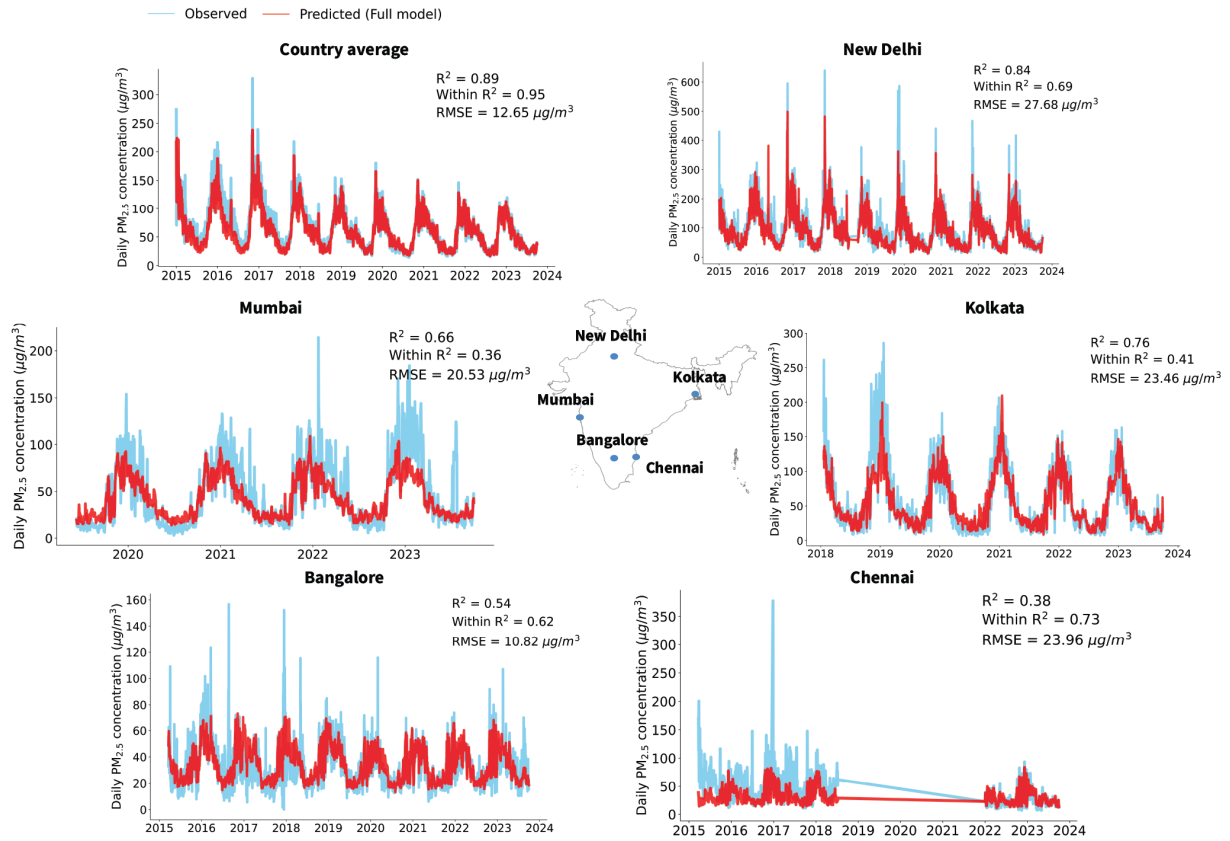


Figure S10: Temporal performance of the AOD model across five mega-cities. Different y-axis and x-axis scales are used in the figures to accommodate variations in PM<sub>2.5</sub> concentrations and monitor availability across the cities. No monitor data is available for Chennai from July 2018 to December 2022.

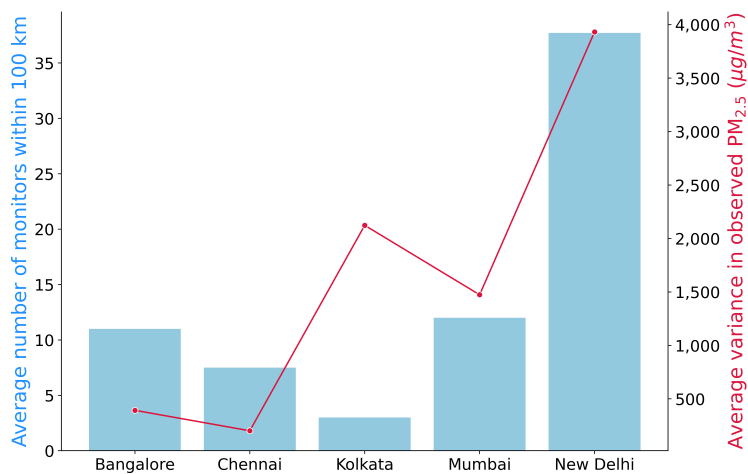


Figure S11: Average number of air quality monitors within 100 km and average variance in observed PM<sub>2.5</sub> concentrations in each city.



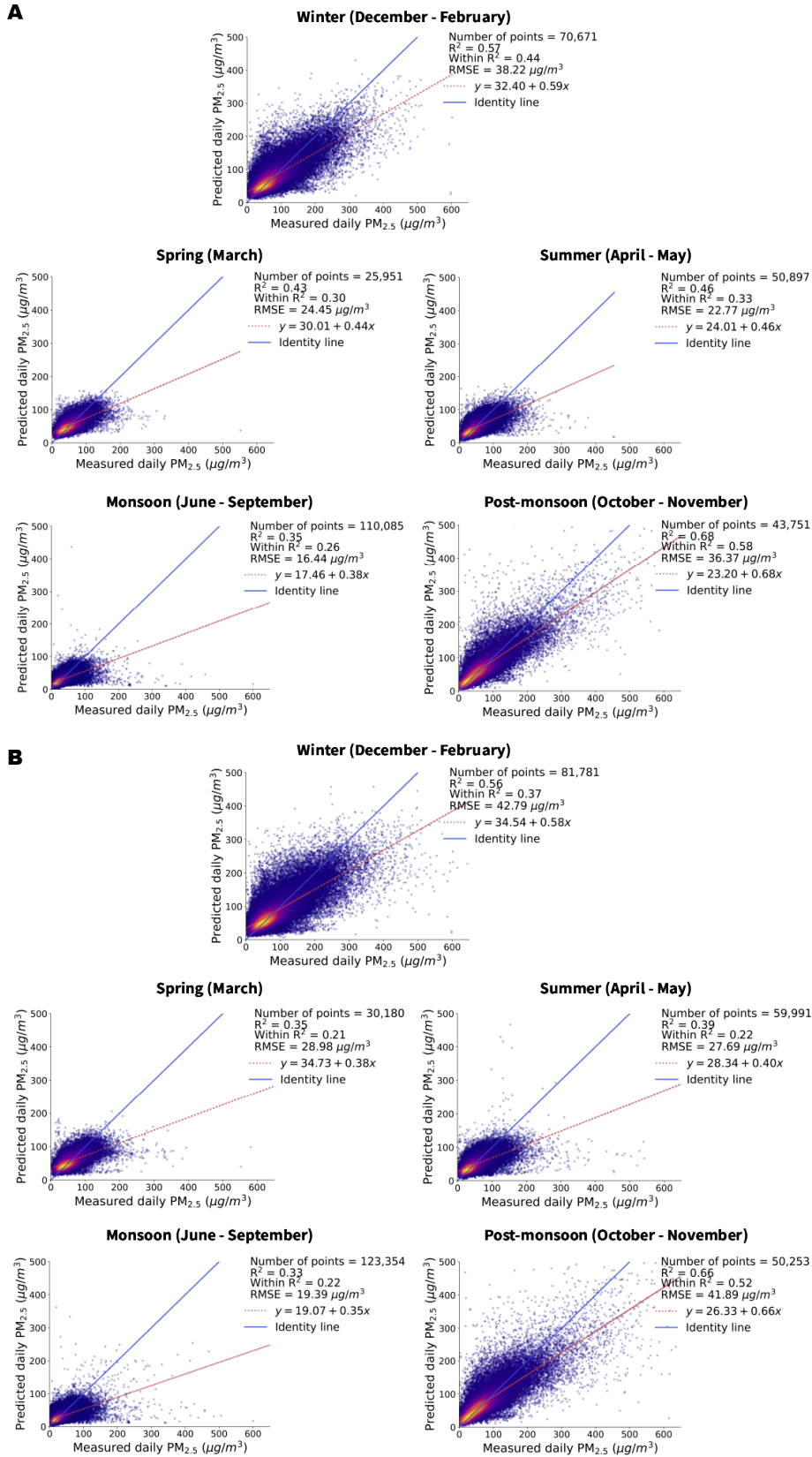


Figure S12: Panel A: seasonal performances of the Full model. Panel B: seasonal performances of the AOD model.

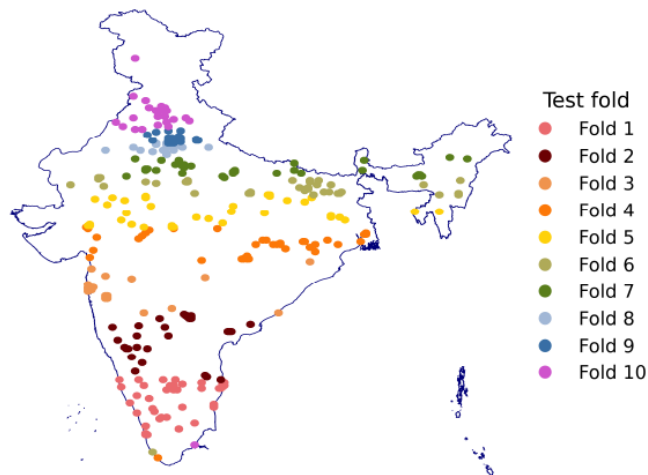


Figure S13: Locations of test data based on latitude included in each of the 10 test folds.

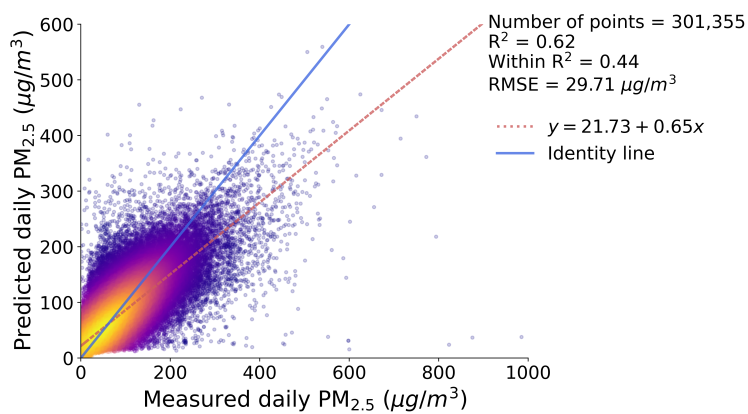


Figure S14: Performance evaluation of the Full model using larger blocks of test data based on latitude.

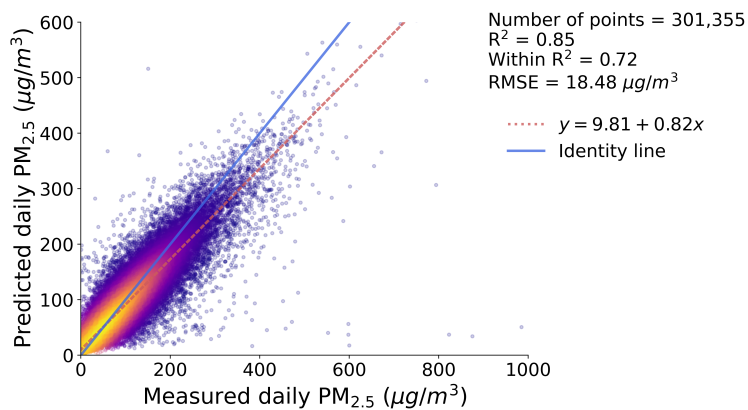


Figure S15: Performance evaluation of the Full model using random 10-fold CV.

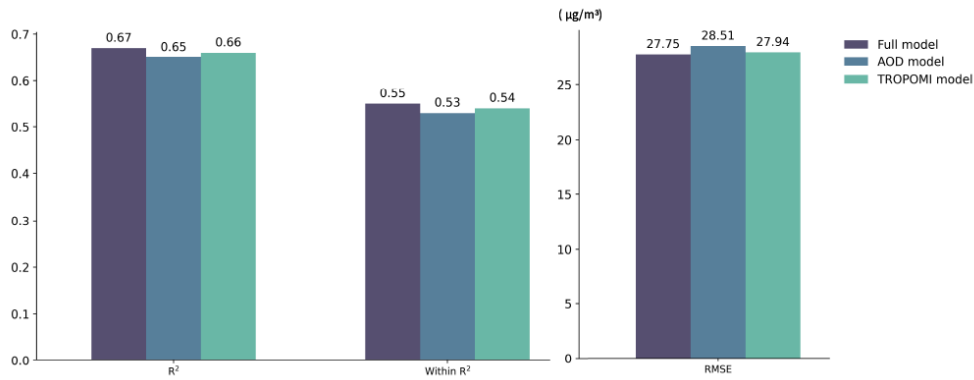


Figure S16: Out-of-sample performances of the Full, AOD, and TROPOMI models using the same sets of training and test data from July 10, 2018, to September 30, 2023.

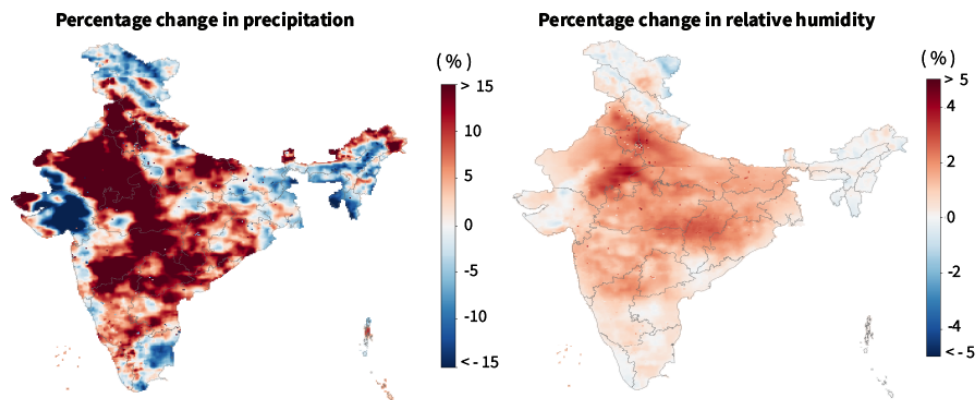


Figure S17: Percentage change in precipitation (left panel) and relative humidity (right panel) calculated for each 10 km grid from 2005-2015 to 2016-2022 using daily total precipitation, temperature, and dewpoint temperature obtained from ECMWF ERA5 data. Temperature and dewpoint temperature are utilized to compute relative humidity for each 10 km grid.

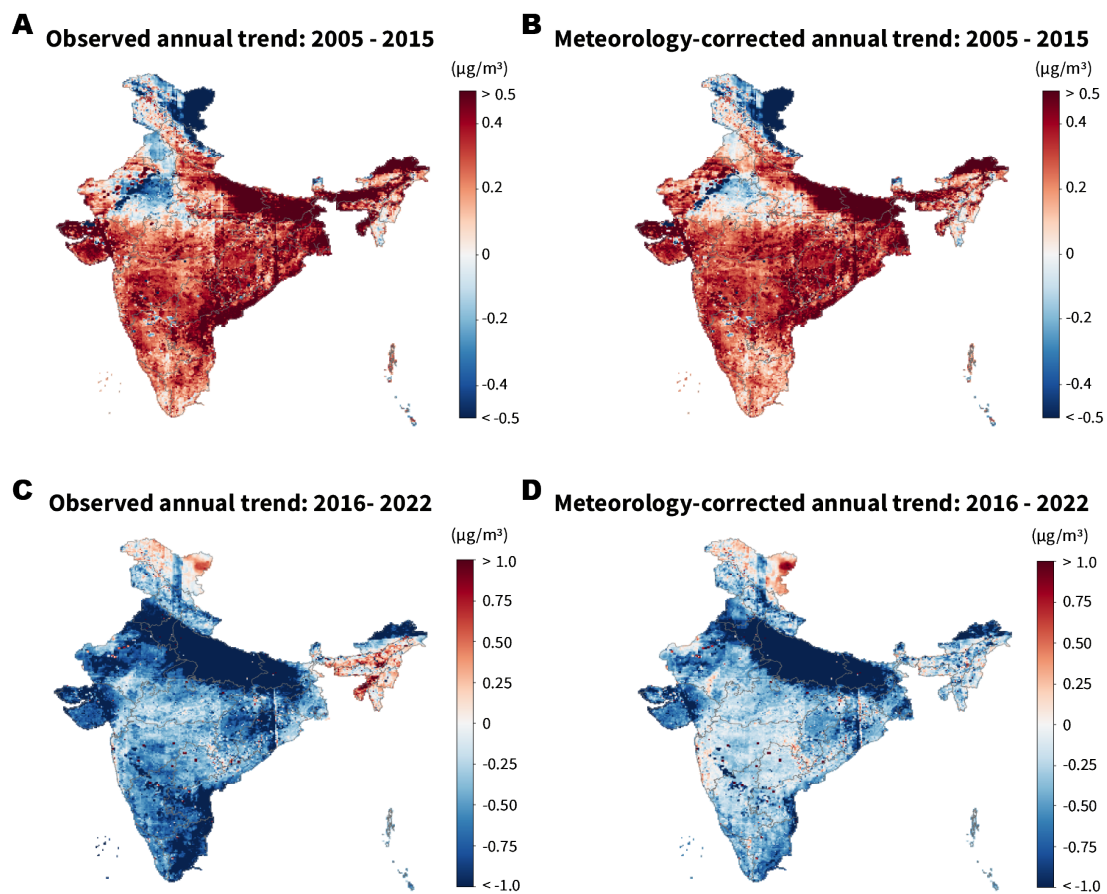


Figure S18: Panel A: average annual trend of observed PM<sub>2.5</sub> from 2005 to 2015. Panel B: average annual trend of PM<sub>2.5</sub> concentrations after controlling for local meteorology from 2005 to 2015. Panel C: average annual trend of observed PM<sub>2.5</sub> from 2016 to 2022. Panel D: average annual trend of PM<sub>2.5</sub> concentrations after controlling for local meteorology from 2016 to 2022.

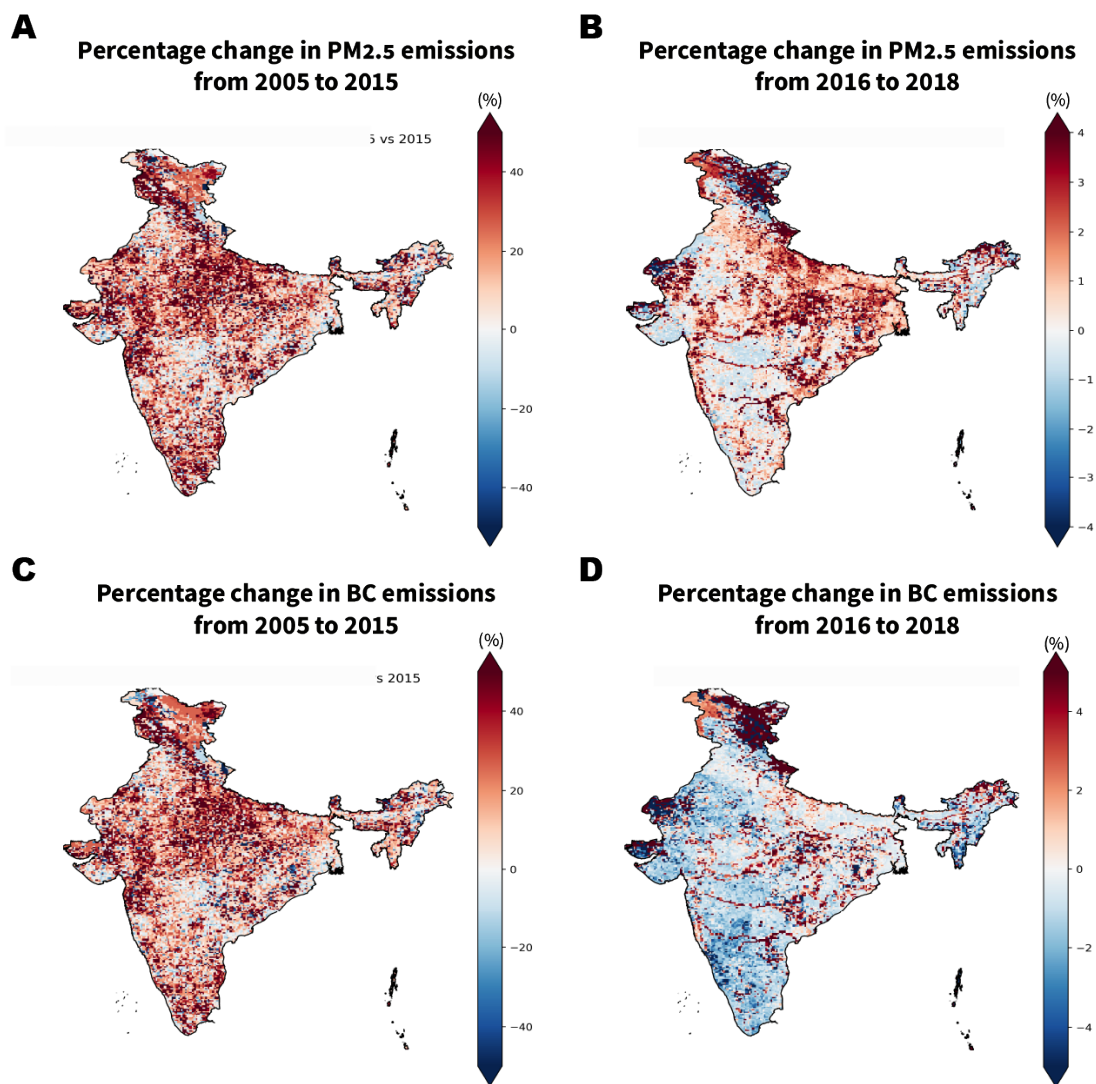


Figure S19: Panel A: percentage change in PM<sub>2.5</sub> emissions from 2005 to 2015. Panel B: percentage change in PM<sub>2.5</sub> emissions from 2016 to 2018. Panel C: percentage change in BC emissions from 2005 to 2015. Panel D: percentage change in BC emissions from 2016 to 2018.

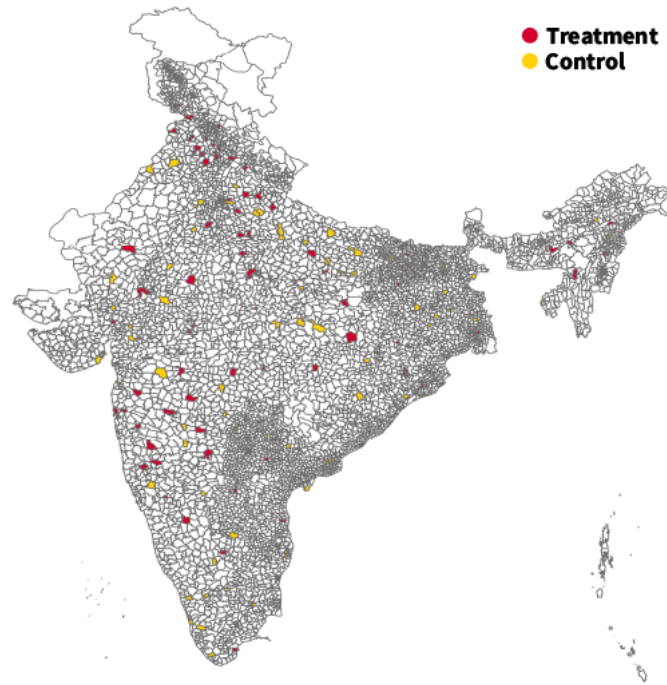


Figure S20: 88 treatment subdistricts consisting of NCAP’s 102 non-attainment cities whose clean action plans were approved in 2020, along with 74 control subdistricts selected using a propensity score method. To account for spillover effects, control subdistricts are not adjacent to treatment subdistricts and located outside a 50 km radius from them.

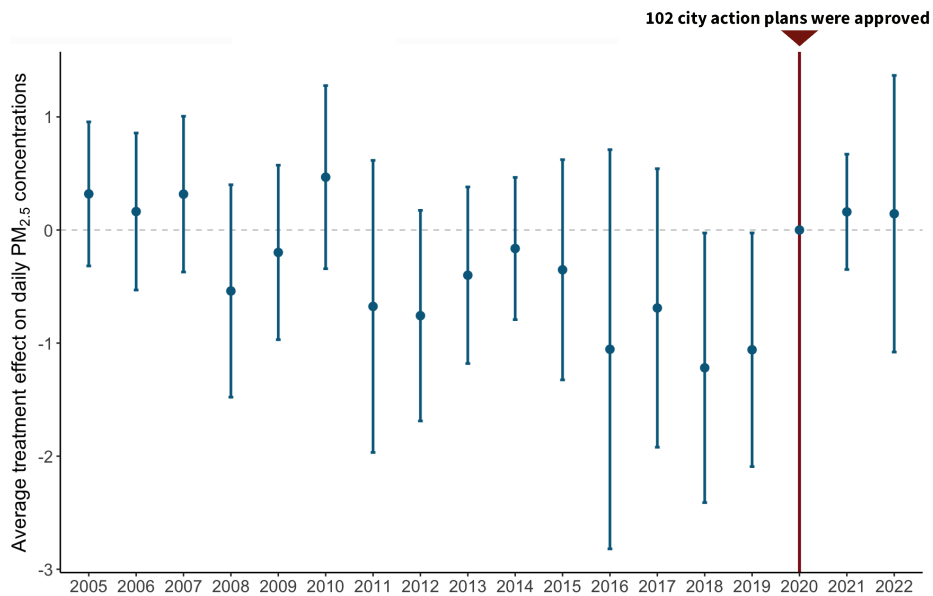


Figure S21: Average changes in daily PM<sub>2.5</sub> concentrations post the implementation of NCAP do not show a statistically significant difference between treatment and control subdistricts, highlighting that there is no evidence that NCAP contributed to declines in the PM<sub>2.5</sub> concentrations.

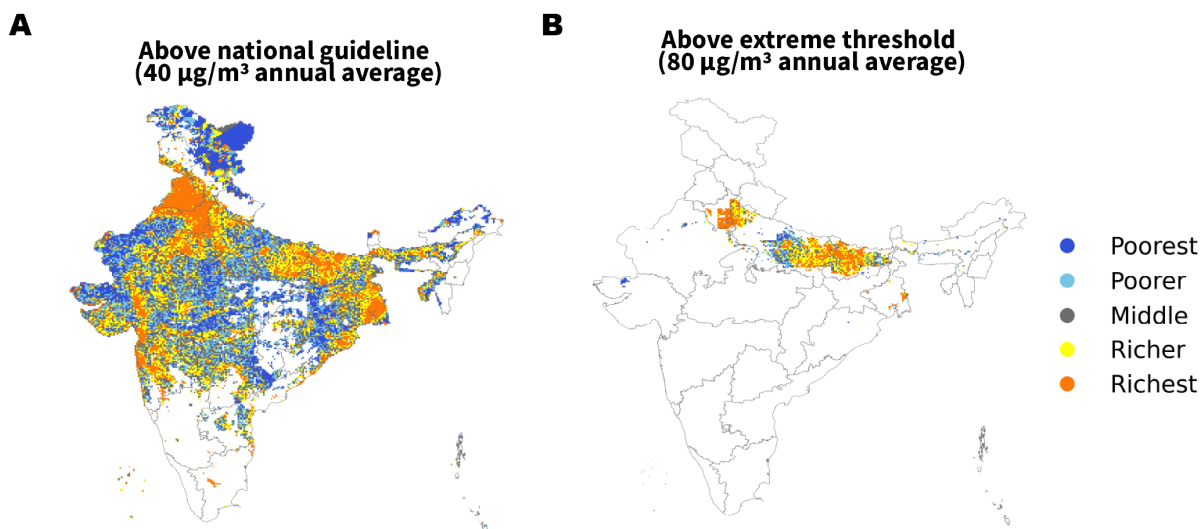


Figure S22: Panel A: locations that were exposed to average concentrations of  $\text{PM}_{2.5}$  exceeding the national guideline of  $40 \mu\text{g}/\text{m}^3$  per wealth category during 2015-2019. Panel B locations with  $\text{PM}_{2.5}$  average concentrations above the extreme threshold of  $80 \mu\text{g}/\text{m}^3$  per wealth category during the same period.

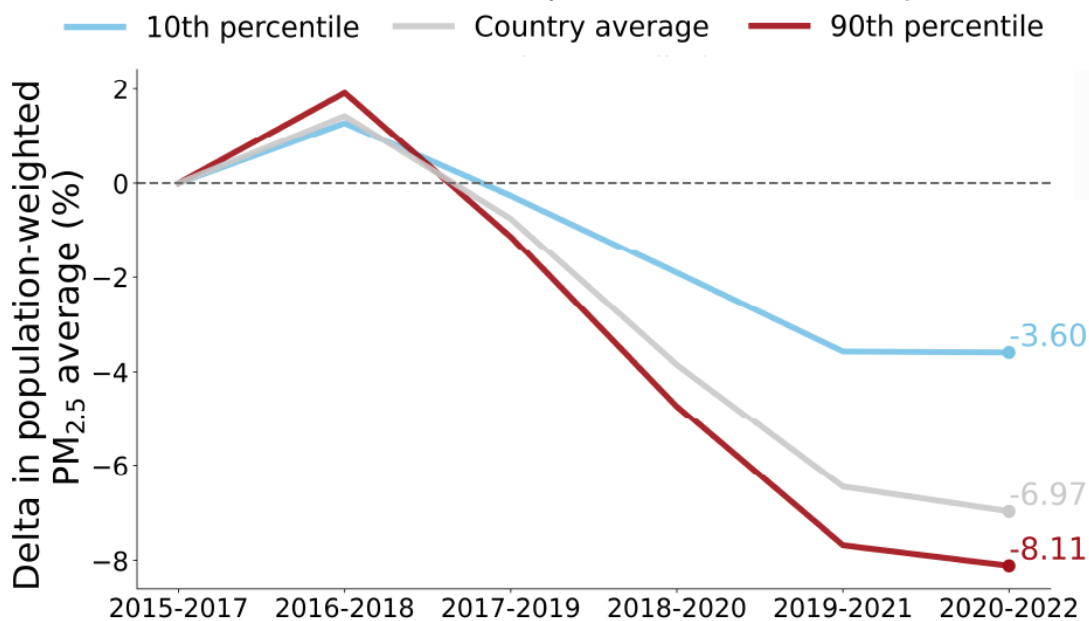


Figure S23: Percentage changes in 3-year population-weighted averages relative to the 2015-17 average. The x-axis label represents the running means of years from 2015-2017 to 2020-2022.

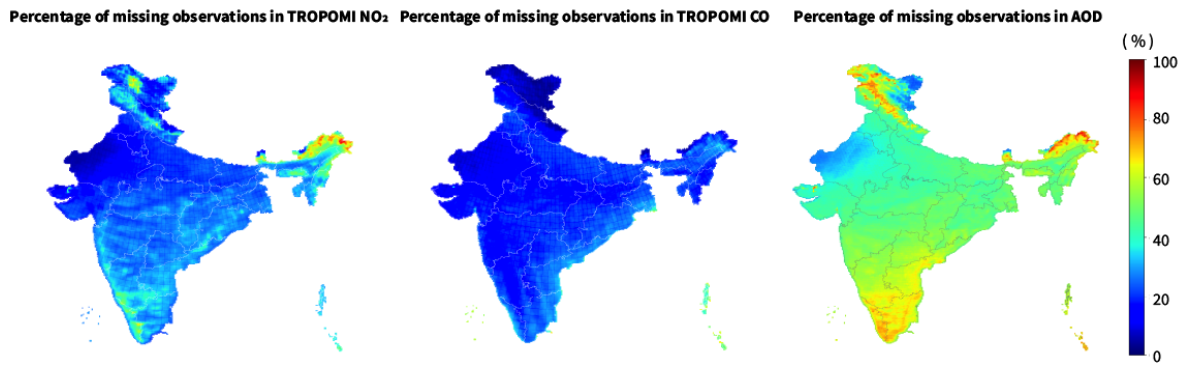


Figure S24: Percentage of daily observations missing in TROPOMI NO<sub>2</sub>, TROPOMI CO, and MODIS AOD data per 10 km grid from July 10, 2018, to September 30, 2023.

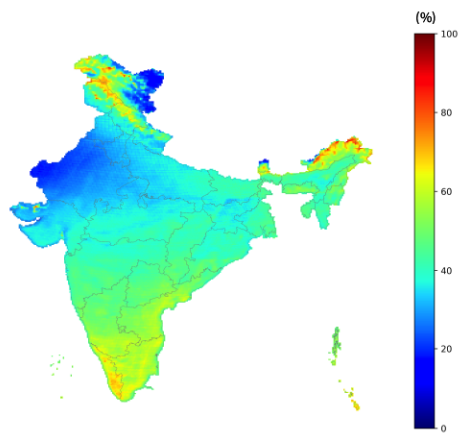


Figure S25: Percentage of daily observations missing in MODIS AOD data per 10 km grid from January 1, 2013, to September 30, 2023.