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1 Two decades of kilometer-scale daily PM<sub>2.5</sub> from satellite observations  
2 and machine learning reveal geographically diverging exposure in Ghana

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12 **Abstract**

13 Exposure to fine particulate matter (PM<sub>2.5</sub>) is a major contributor to global burden of disease, yet  
14 air quality data remain sparse in many low- and middle-income countries, limiting nationwide  
15 monitoring and effective policy development. We address this gap by developing a high-resolution  
16 gridded (1 km × 1 km) dataset for daily surface PM<sub>2.5</sub> concentrations in Ghana from 2005 to 2025  
17 by training multiple machine learning (ML) models built on ground-based monitoring, satellite  
18 observations, and reanalysis products for atmospheric composition and meteorological parameters.  
19 Estimates from these models were evaluated with measurements from reference-grade monitors  
20 and a large network of calibrated low-cost sensors deployed across Ghana. XGBoost showed the  
21 strongest performance among all ML algorithms and best captured spatial and temporal variability  
22 in PM<sub>2.5</sub> levels. SHapley Additive exPlanations (SHAP) analysis for model predictors indicates  
23 that both meteorological variables and aerosol optical properties are key contributors to model  
24 performance. The long-term gridded PM<sub>2.5</sub> dataset reveals a unique north-south exposure disparity  
25 in Ghana, with northern regions of the country experiencing substantially higher PM<sub>2.5</sub>  
26 concentrations compared to the South, that may be widening over the 21-year period by over 0.2  
27 µg m<sup>-3</sup> yr<sup>-1</sup>. This study provides the first long-term high-resolution PM<sub>2.5</sub> exposure levels for Ghana  
28 and presents a scalable framework for generating air quality information in data-sparse regions to  
29 support air pollution relevant health impact assessment and evidence-based mitigation policies.

30 **Keywords**

31 Air quality, fine particulate matter (PM<sub>2.5</sub>), satellite-remote sensing, low-cost sensors, geospatial  
32 machine learning, sub-Saharan Africa, exposure disparity

## 33 1 INTRODUCTION

34 Air pollution is the second leading risk factor of deaths globally after high blood pressure (Health  
35 Effects Institute, 2024). In 2021, exposure to air pollution was associated with around 1.1 million  
36 premature annual deaths in Africa (Amooli et al., 2025; Fisher et al., 2021). This risk is dominated  
37 by exposure to fine particulate matter (PM<sub>2.5</sub>), which is associated with ischemic heart disease,  
38 stroke, respiratory and neurodegenerative diseases (Hayes et al., 2020; Shi et al., 2020). This  
39 burden is particularly severe in low- and middle-income countries (LMICs), including many  
40 African nations, where ambient PM<sub>2.5</sub> concentrations frequently exceed World Health  
41 Organization (WHO) guidelines. However, long-term air quality observations in these regions  
42 remain sparse, constraining accurate exposure assessment, epidemiological analysis, and  
43 consequently, evidence-based mitigation policies.

44 The high capital cost and technical skills needed for establishing and operating a network of  
45 regulatory instruments present a significant barrier for continuous air quality monitoring. African  
46 countries contain an average of only ~3 sensors per 100 million people compared with 379 in  
47 Europe (Gualtieri et al., 2024). Beta attenuation monitors (BAMs) deployed by the US Department  
48 of State at the US Embassies have been one of the major sources of reliable open access PM<sub>2.5</sub>  
49 measurements around the world. Multi-year hourly PM<sub>2.5</sub> levels from these monitors have  
50 provided important insights into PM<sub>2.5</sub> trends across the African continent, including the first long-  
51 term observations in some countries (Anand et al., 2024; Li et al., 2024). However, these  
52 deployments are typically limited to just one to two monitors per country capital and hence are  
53 unable to capture spatial variability of PM<sub>2.5</sub> even within the cities. Recent efforts to address these  
54 monitoring gaps include using networks of consumer-grade air sensors for city-scale monitoring  
55 of particulate matter through fixed-duration campaigns (Owusu-Tawiah et al., 2025; Raheja et al.,  
56 2022; Westervelt et al., 2023). However, these sensors are primarily suited for point-location  
57 measurements and therefore deploying dense networks over large geographic regions remains  
58 prohibitively expensive, limiting their use for long-term nationwide air quality monitoring.

59 Satellite-based air quality products have emerged as a promising solution for long-term  
60 spatiotemporal monitoring in data-sparse regions. These products often provide near-global spatial  
61 coverage, with several satellite instruments offering multi-decadal records of daily observations  
62 for air pollution parameters extending back to the early 2000s. Aerosol optical depth (AOD)  
63 retrieved from satellites has been widely used for estimating ground level PM<sub>2.5</sub>. Liu et al. (2004)  
64 used a scaling factor calculated as the ratio of surface PM<sub>2.5</sub> mass concentration to satellite-  
65 measured column-integrated AOD to estimate spatial PM<sub>2.5</sub> concentrations across the United  
66 States (Liu et al., 2004). Other studies derived PM<sub>2.5</sub>-to-AOD ratios simulated by the GEOS-Chem  
67 chemical transport model and applied to satellite-based AOD to generate global maps of annual  
68 (Hammer et al., 2020; Van Donkelaar et al., 2016) and monthly (Van Donkelaar et al., 2021) PM<sub>2.5</sub>  
69 concentrations. However, the relationship between PM<sub>2.5</sub> and AOD varies substantially across the  
70 globe, largely due to geographical differences in vertical distribution of aerosol loading (Van  
71 Donkelaar et al., 2006). As a result, the accuracy of these estimates heavily relies on the availability  
72 of reliable ground-based measurements for constraining models and capturing these regional  
73 variations between columnar AOD and surface PM<sub>2.5</sub>. Several studies have therefore emphasized  
74 the urgent need for expanding surface monitoring infrastructure in the Global South, particularly  
75 in Africa, to improve satellite-based PM<sub>2.5</sub> estimates (Anand et al., 2024; Bachwenkizi et al., 2021;

76 Di et al., 2016; Hammer et al., 2020; Philip et al., 2014; Van Donkelaar et al., 2016; Vohra et al.,  
77 2021; Weagle et al., 2018).

78 Recent advances in African air quality monitoring provide an opportunity for regionally-refined  
79 satellite-based surface PM<sub>2.5</sub> datasets trained and tested on local measurements. Zhang et al. (2021)  
80 used satellite AOD, meteorological features, land use variables and socioeconomic parameters to  
81 develop daily 1 km × 1 km PM<sub>2.5</sub> concentration maps for 2014–2018 in South Africa, validated by  
82 measurements from a network of 20 surface monitors in South Africa. Amooli et al. (2024)  
83 developed an AOD to PM<sub>2.5</sub> conversion framework to generate 1 km × 1 km spatial PM<sub>2.5</sub> for  
84 Ouagadougou, the capital of Burkina Faso, using surface measurements at the US Embassy in the  
85 capital. Furthermore, the spatial resolutions of existing global PM<sub>2.5</sub> products based on Modern-  
86 Era Retrospective analysis for Research and Applications, Version 2 (MERRA-2) data remain too  
87 coarse (>50 km) to capture intra-city PM<sub>2.5</sub> variations across most cities in Africa (Sayeed et al.,  
88 2022).

89 Gaseous pollutants, such as nitrogen dioxide (NO<sub>2</sub>), sulfur dioxide (SO<sub>2</sub>) and volatile organic  
90 compounds, act as precursors to PM<sub>2.5</sub> formation (Pye et al., 2022) and can help improve the  
91 accuracy of surface-level PM<sub>2.5</sub> estimates (Zheng et al., 2023). In this study, we propose a multi-  
92 model machine learning approach to estimate 21 years (2005–2025) of daily PM<sub>2.5</sub> concentrations  
93 for Ghana at 1 km × 1 km spatial resolution. In addition to AOD, this modeling approach  
94 incorporates satellite-derived information on gaseous precursors to PM<sub>2.5</sub> formation and other  
95 aerosol indicators. We also include meteorological parameters to account for weather-induced  
96 variability in PM<sub>2.5</sub> concentrations. A large network of ~98 well-calibrated surface PM<sub>2.5</sub> monitors  
97 (~275 monitors per 100 million people) were deployed across Ghana, and the measurements were  
98 used to capture the geographic distribution of PM<sub>2.5</sub> concentration in Ghana. These additions  
99 collectively mitigate large uncertainties in satellite-derived surface PM<sub>2.5</sub>, conventionally observed  
100 due to a limited representation of input variables as well as from the lack of surface measurements  
101 of PM<sub>2.5</sub> in regions like sub-Saharan Africa. We further augment missing PM<sub>2.5</sub> information due  
102 to occasional unavailability of satellite-derived predictors by developing a MERRA-2-based PM<sub>2.5</sub>  
103 model. To our knowledge, this is among the first efforts to generate long-term gap-filled PM<sub>2.5</sub>  
104 estimates at 1 km × 1 km grid resolution trained and evaluated using robust local observations in  
105 Africa, while also providing insights into spatiotemporal trends of PM<sub>2.5</sub> across Ghana.

## 106 **2 MATERIALS AND METHODS**

### 107 **2.1 Study area**

108 Ghana is located in West Africa and lies between latitudes ~ 4.5°N and 11.5°N and longitudes  
109 3.5°W and 1.5°E, covering an area of about 238,535 km<sup>2</sup>. The country is characterized by diverse  
110 geographic features, including coastal plains along the Gulf of Guinea, forested regions in the  
111 south, and savanna landscapes in the north. Major urban centers include Accra, Kumasi, Tamale,  
112 Sekondi-Takoradi, Cape Coast, Sunyani and the industrial port of Tema, representing important  
113 population and economic hubs with diverse emission characteristics.

114 Ghana has a typical tropical climate with distinct wet and dry seasons. Southern Ghana typically  
115 exhibits bimodal rainfall patterns due to its proximity to the Gulf of Guinea and the progression of  
116 the Intertropical Convergence Zone (ITCZ), with major and minor wet seasons occurring between

117 April–June and September–October, respectively. In contrast, Northern Ghana has a unimodal wet  
118 season between April and October, peaking during August–September. Ghana experiences a major  
119 meteorological phenomenon during mid-November to mid-March, known as the Harmattan, which  
120 overlaps with the dry season. During this period, dry northeasterly winds transport large amounts  
121 of mineral dust from the Sahara Desert across West African countries, including Ghana. This  
122 results in extremely high concentrations of particulate matter across the country, leading to  
123 severely degraded air quality and reduced visibility, with impacts more pronounced in northern  
124 Ghana than in the south.

## 125 **2.2 Surface PM<sub>2.5</sub> measurements**

126 Ground-level measurements for PM<sub>2.5</sub> were collected from a network of 98 ground sensors spread  
127 across Ghana. The network consisted of 96 low-cost sensors, including 10 Airnote v2.0 (Blues  
128 Inc., Boston, MA), 21 Clarity Node-S Generation II (Clarity Movement Co., Berkeley, CA), 15  
129 PurpleAir PA-II SD monitors (PurpleAir Inc., Draper, UT), 6 MODULAIR™ (QuantAQ Inc.,  
130 Somerville, MA), and 44 MODULAIR™-PM monitors (QuantAQ Inc., Somerville, MA). PM<sub>2.5</sub>  
131 measurements from two Beta Attenuation Monitors (BAMs; BAM-1020, Met One Instruments  
132 Inc., Grants Pass, Oregon) were also obtained, one deployed in Techiman and another located at  
133 the U.S. Embassy in Accra, Ghana. BAM is certified by the U.S. EPA as a Federal Equivalent  
134 Method and hence served as the reference monitor for calibrating raw PM<sub>2.5</sub> measurements from  
135 the low-cost sensors in the network. Table S1 of the Supporting Materials details the technical  
136 specifications of each monitor type and Fig. S1 illustrates the deployment locations of all sensors  
137 used in this study. PM<sub>2.5</sub> measurements collected from this sensor network between August 2018  
138 and October 2024 were used as ground-truth for model development. Fig. S2 shows deployment  
139 periods of each surface instrument included in the study.

140 These sensors were strategically deployed across Ghana, with a significantly higher density in  
141 urban areas where population is largely concentrated, to capture high spatial variability in PM<sub>2.5</sub>  
142 concentrations arising from diverse sources, including vehicular emissions, open burning, cooking  
143 activities and industrial emissions. Air quality monitors were also deployed in suburban and rural  
144 regions of Ghana, where local air quality is strongly influenced by emissions from solid biomass  
145 fuel use for cooking, agricultural residue burning and dust from unpaved roads. This sensor  
146 network architecture helps inform the model about the spatial distribution of PM<sub>2.5</sub> in Ghana during  
147 the Harmattan period by using measurements from northern, middle and southern belts of Ghana.  
148 Fig. S1 depicts the locations of every monitor against a population count map retrieved from the  
149 LandScan project at Oak Ridge National Laboratory (Lebakula et al., 2025). Additional  
150 deployment details are available in Tables S2–S3.

151 Raw PM<sub>2.5</sub> measurements from each low-cost sensor, available at sub-hourly time resolution, were  
152 aggregated to hourly concentrations and subjected to custom-designed calibration models unique  
153 to each sensor type to derive QA/QC-corrected exposure levels. The calibration procedures are  
154 further explained in Supplementary Text S1.

## 155 2.3 Satellite and reanalysis datasets

### 156 2.3.1 MODIS Level 2 aerosol optical depth

157 Moderate Resolution Imaging Spectroradiometer (MODIS) is an optical sensor aboard NASA's  
158 sun-synchronous low-Earth orbit (LEO) Terra (launched in 1999) and Aqua (launched in 2002)  
159 satellites, passing over the equator at 10:30 am and 1:30 pm local time, respectively. In this study,  
160 we used MODIS AOD at 550 nm derived through the Multi-Angle Implementation of Atmospheric  
161 Correction (MAIAC) algorithm (Lyapustin et al., 2018), specifically from the MCD19A2 product.  
162 It is a Level 2 (L2) product, produced by combining measurements from MODIS instruments  
163 aboard both Aqua and Terra satellites, that provides daily global coverage in multiple spectral  
164 bands and has been widely used for monitoring land, ocean, and atmospheric parameters for over  
165 two decades. The MAIAC algorithm combines time series analysis and multi-angle radiative  
166 transfer modeling to produce atmospherically corrected surface reflectance and high-quality AOD  
167 retrievals at a  $1 \text{ km} \times 1 \text{ km}$  spatial resolution beginning from February 2000. The enhanced spatial  
168 resolution, improved cloud screening, and robust performance over complex surfaces make  
169 MCD19A2 particularly valuable for estimating ground-level  $\text{PM}_{2.5}$  in data-sparse regions, such as  
170 Ghana.

171 The AOD from the MCD19A2 product is available in sinusoidal projection tiles. Tiles h17v07,  
172 h17v08, h18v07, and h18v08 were combined to gather AOD information for Ghana. For each day,  
173 the *Optical\_Depth\_055* product and corresponding *AOD\_QA* layers were extracted and used for  
174 further analysis. The *AOD\_QA* layer contains 16-bit unsigned integers, with bits 8-11 embedding  
175 the QA information. QA-based filtering was applied to retain *Optical\_Depth\_055* retrievals with  
176 QA flags {0000, 0011, 1011}, classified as best-quality observations over land for research  
177 applications, while excluding pixels affected by cloud contamination, snow, sun glint, coastline  
178 artifacts, or missing retrieval conditions, to ensure AOD retrievals considered suitable for scientific  
179 analysis in air quality applications. The AOD values were then calibrated using the scale factor  
180 and offset from the product attributes to obtain physically meaningful retrievals. All valid orbit-  
181 level retrievals within each day were aggregated for each cell, and measurements for duplicate grid  
182 cells were averaged to generate daily mean AOD values.

### 183 2.3.2 OMI Level 2 air quality products

184 Ozone Monitoring Instrument (OMI) has been continuously measuring air quality parameters  
185 onboard NASA's Aura spacecraft since July 2004. Similar to Terra and Aqua, Aura is a sun-  
186 synchronous LEO satellite and crosses the equator at 1:45 PM local time. We used OMI L2  
187 Collection 3 measurements for ultraviolet aerosol index (UVAI) and column number density for  
188 ozone ( $\text{O}_3$ ), tropospheric  $\text{NO}_2$ ,  $\text{SO}_2$  and formaldehyde (HCHO). These air quality features are  
189 available at a spatial resolution of  $13 \text{ km} \times 24 \text{ km}$ . UVAI is a unitless quantity derived from top-  
190 of-atmosphere radiances at 354 nm and 388 nm, which allows distinction between UV-absorbing  
191 aerosols (e.g., dust, biomass burning smoke, haze) and non-absorbing particles such as clouds  
192 (Torres et al., 2020). HCHO and  $\text{NO}_2$  are measured in molecules  $\text{cm}^{-2}$ , whereas  $\text{O}_3$  and  $\text{SO}_2$  are  
193 expressed in Dobson Unit (DU), where  $1 \text{ DU} = 2.69 \times 10^{16} \text{ molecules cm}^{-2}$ . The Level 2 OMI  
194 data occasionally contain multiple retrievals for a given grid cell within a single day due to multiple  
195 orbital overpasses. These intra-day measurements were aggregated to generate a single daily-  
196 averaged retrieval for each grid cell.

197 Quality control was applied individually to each OMI product to account for differences in  
198 retrieval sensitivity, signal strength, and known instrumental artifacts. For the UV Aerosol Index,  
199 pixels were retained only when  $CloudFraction \leq 0.3$ ,  $SolarZenithAngle \leq 75^\circ$ , and  
200  $XTrackQualityFlags = 0$ . The  $XTrackQualityFlags$  filter is particularly useful in excluding cross-  
201 track (row) anomalies and geometries associated with increased radiative transfer uncertainty.  
202 HCHO is more sensitive to cloud contamination and retrieval noise, and hence strict filtering  
203 criteria were applied by requiring  $AMFCloudFraction \leq 0.3$ ,  $SolarZenithAngle \leq 75^\circ$ ,  
204  $XTrackQualityFlags = 0$ , and  $MainDataQualityFlag = 0$  to ensure successful spectral fitting and  
205 reliable air mass factor calculations. Total column ozone, a comparatively high signal-to-noise  
206 product, was screened using  $RadiativeCloudFraction \leq 0.3$ ,  $SolarZenithAngle \leq 85^\circ$ , and  
207  $QualityFlags \leq 1$ , thereby retaining only good or glint-corrected retrievals. For tropospheric  $NO_2$ ,  
208 pixels were restricted to  $CloudFraction \leq 0.3$ ,  $SolarZenithAngle \leq 75^\circ$ , and  $VcdQualityFlags = 0$   
209 to remove cloud-affected scenes and retrievals with known fitting or cross-track issues.  $SO_2$   
210 retrievals, which are particularly sensitive to instrumental artifacts and weak signal conditions,  
211 were subjected to the most stringent screening:  $CloudRadiationFraction \leq 0.3$ ,  $SolarZenithAngle$   
212  $\leq 65^\circ$ ,  $Flag\_RowAnomaly \neq 1$ ,  $Flag\_SAA \neq 1$ , and  $AlgorithmFlag\_SnowIce \neq 2$ , excluding scenes  
213 affected by the OMI row anomaly, South Atlantic Anomaly interference, or uncertain cloud  
214 detection over snow and ice. The  $ColumnAmountSO2\_PBL$  dataset from the OMI L2  $SO_2$  product  
215 was specifically selected because it represents  $SO_2$  VCD within the planetary boundary layer and  
216 is commonly used as an indicator of anthropogenic  $SO_2$  emissions (Li et al., 2013). These species-  
217 specific filtering criteria are consistent with OMI Collection 3 product documentation and widely  
218 adopted validation studies, ensuring that only physically meaningful and robust retrievals were  
219 included in the analysis. More details on quality screening of OMI and other satellite-derived  
220 products are summarized in Table S4.

### 221 **2.3.3 TROPOMI Level 2 air quality features**

222 TROPospheric Monitoring Instrument (TROPOMI) is onboard the Copernicus Sentinel-5  
223 Precursor (S5P), another sun-synchronous LEO satellite that passes over the equator at 1:30 PM  
224 local time and was launched in October 2017. We used L2 Version 2 UVAI (unitless) and column  
225 number density measurements (in  $mol\ m^{-2}$ ) for  $O_3$ , tropospheric  $NO_2$ ,  $SO_2$ , and tropospheric  
226 HCHO, which are similar to the parameters used from OMI and are consistently available from  
227 May 7, 2018. In addition, carbon monoxide (CO) retrievals from TROPOMI were also used as it  
228 is one of the primary combustion products from any combustion activity and hence a strong tracer  
229 for combustion-related emission sources in Ghana (Griffin et al., 2024). The spatial resolution for  
230  $O_3$ ,  $NO_2$ ,  $SO_2$ , HCHO and UVAI products was  $7\ km \times 3.5\ km$  (across  $\times$  along track) until August  
231 5, 2019 and improved to  $5.5\ km \times 3.5\ km$  from August 6, 2019 onward due to reduced across-  
232 track detector binning. This corresponds to approximately a 16-fold improvement in spatial  
233 resolution compared to OMI. In contrast, CO retrievals from the short wavelength infrared channel  
234 were provided at  $7\ km \times 7\ km$  (across  $\times$  along track) prior to August 6, 2019 and at  $7\ km \times 5.5\ km$   
235 thereafter. TROPOMI provides tropospheric column density for HCHO which differs from Level  
236 2 total column HCHO density retrievals from OMI. In most cases, particularly over polluted areas,  
237 the amount of HCHO in the stratosphere is negligible and almost all HCHO resides in the  
238 troposphere due to its short lifetime (Ayazpour et al., 2025; De Smedt et al., 2008). Therefore,  
239 tropospheric HCHO is typically approximated as total column HCHO. The reprocessed dataset  
240 (RPRO) covers April 30, 2018 until July 25, 2022 and is fully compatible with the current offline  
241 dataset (OFFL), which covers July 26, 2022 onwards. TROPOMI provides UVAI at three

242 wavelength pairs: 335/367 nm, 340/380 nm, and 354/388 nm but only UVAI calculated at 354 nm  
243 and 388 nm wavelengths was used as it is an indicator of dust and biomass burning smoke (Torres  
244 et al., 2020). Following the quality screening criteria provided in the official product  
245 documentation, only pixels with  $qa\_value \geq 0.5$  were retained for each TROPOMI product to  
246 ensure high-quality retrievals and exclude observations affected by clouds or snow/ice (Van  
247 Geffen et al., 2022).

#### 248 **2.3.4 ERA5-Land reanalysis hourly dataset**

249 ERA5-Land is a fifth-generation reanalysis of global climate dataset from European Centre for  
250 Medium-Range Weather Forecasts (ECMWF), covering the period 1940-present at hourly  
251 temporal resolution (Muñoz-Sabater et al., 2021). The dataset is available at  $0.1^\circ \times 0.1^\circ$  grid  
252 resolution for dewpoint temperature at 2 m above Earth’s surface ( $T_d$ ), surface temperature at 2 m  
253 above surface ( $T$ ), total precipitation (TP), and wind velocities at 10 m above the ground in east-  
254 west direction (U) and north-south direction (V). These variables have proven to be important  
255 meteorological drivers of ambient pollution dynamics (Westervelt et al., 2025). Surface  
256 temperatures and relative humidity are derived at 2 m whereas wind speeds at 10 m above ground,  
257 following the guidelines from World Meteorological Organization for surface measurements of  
258 meteorological variables (World Meteorological Organization (WMO), 2008). Relative humidity  
259 (RH, in %) is not inherently available from the ERA5-Land dataset and hence was calculated using  
260 the Magnus-Tetens equation (Amooli et al., 2024) as:

$$261 \quad RH(\%) = 100 \times \exp\left(\frac{17.625 \cdot T_d}{243.12 + T_d} - \frac{17.625 \cdot T_{2m}}{243.12 + T_{2m}}\right) \dots (1)$$

262 where  $RH$  is the relative humidity (in %) at 2 m above ground,  $T_d$  and  $T_{2m}$  are 2 m ambient and  
263 dewpoint temperatures (in  $^\circ\text{C}$ ) from ERA5-Land dataset.

#### 264 **2.3.5 MERRA-2 reanalysis product**

265 MERRA-2 reanalysis dataset, developed by NASA’s Global Modeling and Assimilation Office  
266 (GMAO), is a global atmospheric reanalysis that integrates the GEOS-5 Earth system model with  
267 a three-dimensional variational data assimilation system. MERRA-2 assimilates a comprehensive  
268 suite of meteorological observations, including satellite radiances, radiosondes, aircraft, and  
269 surface measurements, and uniquely incorporates space-based AOD observations, enabling  
270 improved representation of aerosol variability and aerosol–radiation interactions. The reanalysis  
271 is produced at a horizontal resolution of  $0.5^\circ \times 0.625^\circ$  with 72 vertical levels extending to 0.01  
272 hPa and provides hourly to monthly-averaged meteorological and aerosol diagnostics from 1980  
273 to present. In this study, we used MERRA-2 aerosol mass and optical properties from aerosol  
274 diagnostics product, M2T1NXAER, as predictors in the model development framework. The  
275 features include hourly total extinction aerosol optical thickness at 550 nm (TOTEXTTAU) and  
276 surface concentrations of black carbon (BCSMASS), organic carbon (OCSMASS), sulfate  
277 (SO4SMASS), sulfur dioxide (SO2SMASS), fine-mode dust (DUSMASS25) and fine-mode sea  
278 salt (SSSMASS25). These variables collectively represent major anthropogenic and natural  
279 aerosol components and provide physically consistent, spatially complete information on aerosol  
280 loading and composition.

## 281 2.4 Data preprocessing

### 282 2.4.1 Regridding input features

283 The ERA5-Land and MERRA-2 datasets are available in Coordinated Universal Time (UTC),  
284 which aligns with the local time in Ghana. Hourly datasets were aggregated to daily resolution as  
285 24-hour averages for each feature retrieved from ERA5-Land and MERRA-2, except the ERA5  
286 total precipitation product, which reports cumulative hourly values since the start of the day. The  
287 00:00 UTC precipitation value was used as the total precipitation for the previous day.

288 ERA5-Land dataset often suffers from missing parameter retrievals for coastal grids containing  
289 even a small ocean fraction. This presents a limitation in deriving  $PM_{2.5}$  for coastal locations,  
290 particularly for Ghana, where major urban centers, such as Accra, Tema, and Sekondi, are located  
291 along the coastline. Missing feature values in these coastal grid cells were imputed using nearest-  
292 neighbor spatial interpolation from adjacent land grid cells.

293 The QA-corrected daily AOD from the MCD19A2 product was transformed from the sinusoidal  
294 to the World Geodetic System 1984 (WGS84; EPSG:4326) projection using the tile-specific  
295 projection information embedded in each file. The daily AOD fields were further regridded to a  
296 spatial resolution of  $0.01^\circ \times 0.01^\circ$  over Ghana, covering a rectangular domain bounded by  $4.5^\circ N$   
297 to  $11.5^\circ N$  and  $3.5^\circ W$  to  $1.5^\circ E$ , using area-weighted averaging.

298 To harmonize the spatial resolution of all input datasets with the 1 km MODIS MAIAC grid, we  
299 performed downscaling of coarser-resolution input features. The ERA5 meteorological variables  
300 ( $0.1^\circ$  resolution) and MERRA-2 features were downscaled to  $0.01^\circ$  grid resolution ( $\sim 1$  km) using  
301 bilinear interpolation, which provides a smooth transition between grid points while preserving  
302 spatial gradients. Satellite-derived atmospheric composition variables from OMI and TROPOMI  
303 were regridded to a spatial resolution of  $0.01^\circ \times 0.01^\circ$  using area-weighted averaging. This method  
304 accounts for the varying overlap between original satellite pixels and the target 1 km grid cells,  
305 ensuring that spatial integrity is maintained during the resampling process. These approaches  
306 enable the integration of diverse atmospheric and environmental data at a common high-resolution  
307 scale, suitable for fine-scale air pollution modeling.

### 308 2.4.2 Data preparation

309 Since surface monitors provide  $PM_{2.5}$  measurements at discrete geographic locations, the training  
310 dataset was constructed by spatially matching  $PM_{2.5}$  observations from each monitor with predictor  
311 values from the  $0.01^\circ \times 0.01^\circ$  grid cells containing the monitor. For each sensor-day observation,  
312 the corresponding daily values of MODIS AOD, OMI/TROPOMI gas-phase retrievals, ERA5-  
313 Land meteorological parameters, and MERRA-2 aerosol diagnostics were extracted from  
314 corresponding grid cells. This collocation was performed for each observation date across the full  
315 sensor deployment period (August 2018 – October 2024), yielding a tabular dataset with each row  
316 representing a unique sensor-day pair and contains the measured  $PM_{2.5}$  concentration alongside  
317 the corresponding values of all predictor variables. Sensor-day records for which any satellite-  
318 derived predictor was unavailable (e.g., due to cloud cover or instrument downtime) were excluded  
319 from the training set for the respective model. Collinearity among predictors was not explicitly  
320 removed, as the machine learning algorithms used in this study are generally robust to correlated

321 inputs. Retaining correlated variables allows the models to capture potentially informative  
322 relationships among atmospheric and satellite-derived features without imposing arbitrary feature  
323 exclusion.

### 324 **2.4.3 Cyclic features**

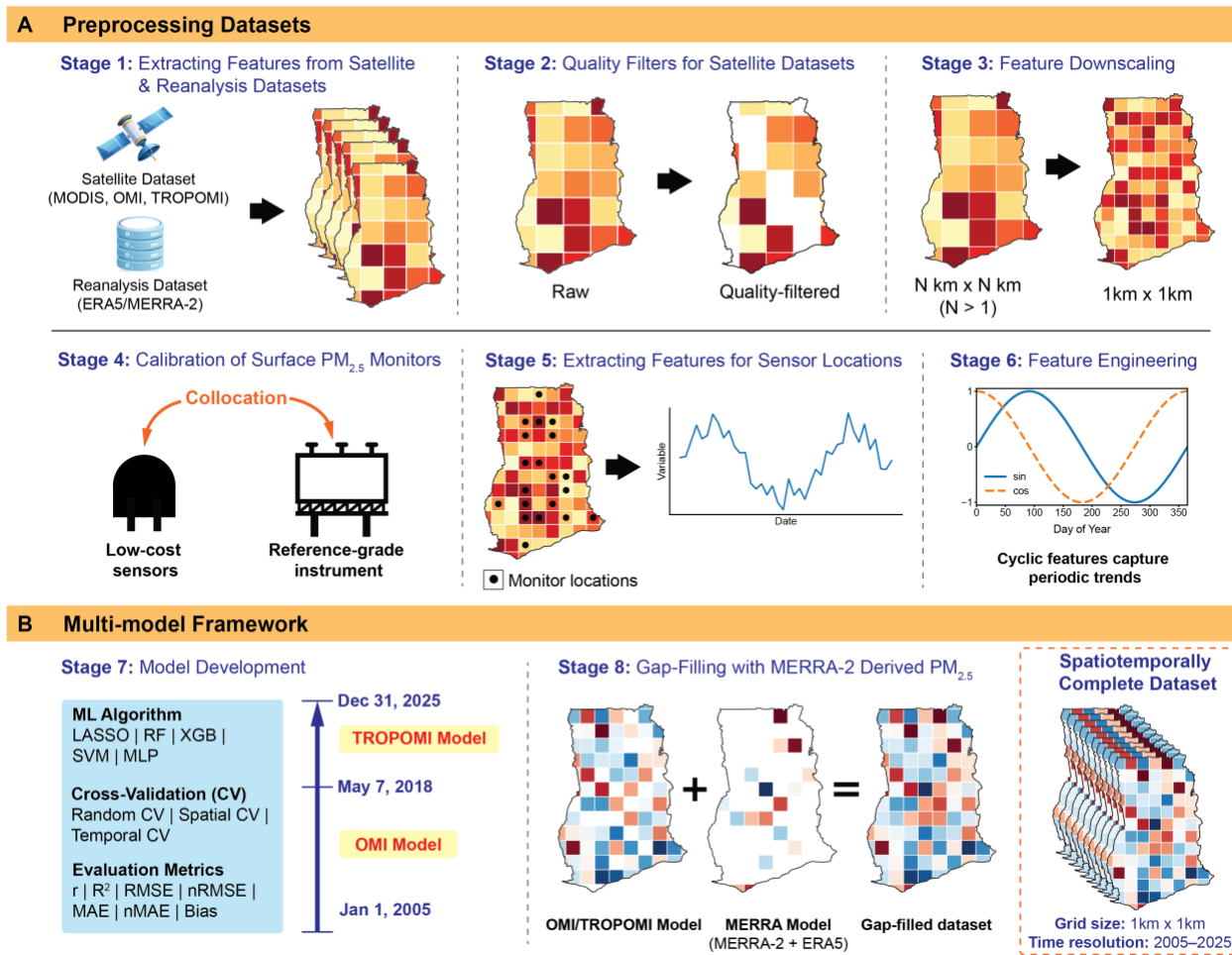
325 PM<sub>2.5</sub> concentrations in Ghana exhibit pronounced seasonal variability, particularly during the  
326 Harmattan period, when long-range transport of Saharan dust substantially elevates particulate  
327 levels. To account for this cyclical behavior and enable the model to capture recurring seasonal  
328 patterns in PM<sub>2.5</sub> variability, the cosine and sine transformation of the day of year (DOY) for a  
329 year of N days was computed below and were included as predictor variables.

$$330 \quad DOY_{COS} = \text{Cosine} \left( \frac{2 * \pi * DOY}{N} \right) \dots (2)$$

$$331 \quad DOY_{SIN} = \text{Sine} \left( \frac{2 * \pi * DOY}{N} \right) \dots (3)$$

### 332 **2.4.4 Standardization**

333 Prior to model training, all continuous predictor variables were standardized using z-score  
334 normalization by subtracting the training set mean ( $\mu$ ) and dividing by the standard deviation ( $\sigma$ )  
335 as  $(x - \mu)/\sigma$ , which is required for gradient-based algorithms, including multi-layer perceptron  
336 (MLP), least absolute shrinkage and selection operator (LASSO), and support vector machine  
337 (SVM), to ensure consistent regularization-based penalties. Tree-based models, including random  
338 forest (RF) and XGBoost, are invariant to monotonic feature transformations and thus did not  
339 require scaling, but the same preprocessed dataset was used for all algorithms to ensure a consistent  
340 comparison. Fill values and physically implausible retrievals were replaced with NaN and  
341 excluded from training prior to normalization.



342

343 **Fig. 1. Overview of the modeling framework for generating high-resolution  $\text{PM}_{2.5}$  estimates.**

344 The workflow demonstrates a three-model framework which developed machine learning  
 345 algorithms by integrating satellite and reanalysis products with ground-based  $\text{PM}_{2.5}$  measurements  
 346 through preprocessing, feature engineering, and model training to produce continuous  
 347 spatiotemporal  $\text{PM}_{2.5}$  concentrations at  $1 \text{ km} \times 1 \text{ km}$  spatial resolution from 2005–2025. Air quality  
 348 input predictors were obtained from satellite instruments, along with meteorological variables  
 349 from ERA5. Satellite observations were quality screened and spatially harmonized prior to model  
 350 development. Ground-based  $\text{PM}_{2.5}$  measurements from reference monitors and calibrated low-cost  
 351 sensors were used for training and validation of multiple machine learning algorithms under  
 352 random, spatial, and temporal cross-validation schemes. Two satellite-based models were  
 353 developed using predictors derived from OMI (OMI Model; January 1, 2005–May 6, 2018) and  
 354 TROPOMI (TROPOMI Model; May 7, 2018–December 31, 2025). Missing satellite retrievals  
 355 were supplemented using a gap-filling model (MERRA Model) developed using aerosol  
 356 diagnostics dataset from MERRA-2 and meteorological variables from ERA5 to produce a  
 357 spatiotemporally complete daily  $\text{PM}_{2.5}$  dataset between 2005–2025 across Ghana.

## 358 2.5 Development of a spatiotemporal $\text{PM}_{2.5}$ model

359 We developed a multi-model framework by integrating satellite retrievals, reanalysis products, and  
 360 ground-based measurements to ensure complete coverage of  $\text{PM}_{2.5}$  for the entire geographical

361 region of Ghana and all days from 2005 to 2025. The framework consists of three complementary  
362 machine learning models with varying input features: TROPOMI model, OMI model and  
363 MERRA-2 model. All three models incorporate ground-level PM<sub>2.5</sub> measurements and  
364 meteorological variables from ERA5 dataset. TROPOMI model derives PM<sub>2.5</sub> beginning May 7,  
365 2018 by integrating AOD from MODIS and air quality parameters (UVAI, CO, HCHO, NO<sub>2</sub>, O<sub>3</sub>,  
366 and SO<sub>2</sub>) from TROPOMI instrument. OMI model also uses MODIS AOD but utilizes only UVAI,  
367 HCHO, NO<sub>2</sub>, O<sub>3</sub>, and SO<sub>2</sub> retrievals from OMI to estimate PM<sub>2.5</sub> from January 1, 2005 to May 6,  
368 2018, before TROPOMI measurements became available.

369 Since both models rely heavily on satellite-derived observations, occasional cloud cover and  
370 instrument-specific limitations, including row anomalies in OMI (Torres et al., 2018) and retrieval  
371 uncertainties in TROPOMI (Veefkind et al., 2012), make it difficult to obtain high-quality satellite  
372 retrievals, leading to missing PM<sub>2.5</sub> estimates from OMI and TROPOMI models in certain regions  
373 and time periods. The MERRA-2-based model was developed to mitigate these PM<sub>2.5</sub> gaps. It  
374 utilizes air quality features from MERRA-2 reanalysis dataset, including BCSMASS, OCSMASS,  
375 SO<sub>4</sub>SMASS, SO<sub>2</sub>SMASS, DUSMASS25, SSSMASS25, and TOTEXTTAU. Although MERRA-  
376 2 also contains meteorological variables, those features were rather extracted from the ERA5-Land  
377 reanalysis dataset due to availability of hourly variables at approximately 31 times finer grid sizes  
378 (0.1° × 0.1°) which is more suitable for developing a high spatial resolution PM<sub>2.5</sub> model. The  
379 MERRA-2-based model therefore serves as a fallback framework for periods or locations where  
380 satellite retrievals from OMI or TROPOMI are unavailable, ensuring continuous spatiotemporal  
381 PM<sub>2.5</sub> coverage across Ghana from 2005–2025.

382 For each model type, geospatial machine learning algorithms, including LASSO, SVM, RF,  
383 XGBoost and MLP, were developed and evaluated to determine the optimal modeling approach  
384 for estimating daily PM<sub>2.5</sub> concentrations. Model performance was evaluated using cross-  
385 validation to ensure robust assessment and reduce the risk of overfitting. The probability  
386 distribution of PM<sub>2.5</sub> measurements from surface monitors exhibits a right-skewed distribution  
387 with a long tail towards higher concentrations (Fig. S4). Therefore, the dataset was first divided  
388 into training (80%) and testing (20%) subsets using stratified sampling based on PM<sub>2.5</sub>  
389 concentration ranges. PM<sub>2.5</sub> values were grouped into discrete bins using thresholds of 35, 60, 100,  
390 150, and 200 µg m<sup>-3</sup>, to ensure that low, moderate, and high pollution conditions were  
391 proportionally represented in both subsets. This stratification improves model robustness by  
392 ensuring representation of episodic high-PM events, which is particularly important in  
393 geographical regions such as Ghana where the Harmattan season periodically leads to elevated  
394 particulate matter levels. Model hyperparameters were optimized using repeated five-fold cross-  
395 validation (5 folds repeated 5 times) applied to the training dataset, with the mean root mean square  
396 error (RMSE) across validation folds used as the loss function to identify the best-performing  
397 model configuration with minimal risk of overfitting. Model performance was assessed using  $r$ ,  
398  $R^2$ , RMSE, MAE and mean bias. Mean-normalized RMSE (nRMSE) and MAE (nMAE) were  
399 computed by dividing the errors by the mean observed PM<sub>2.5</sub> concentrations, enabling comparison  
400 of model performance across environments with varying pollution levels.

401 After determining the optimal hyperparameters, the model configuration was fixed and further  
402 evaluated using structured cross-validation to assess its ability to generalize across Ghana and  
403 time. Spatial and temporal cross-validation were used to reduce the influence of spatial and  
404 temporal autocorrelation and provide a more realistic estimate of model generalization.

405 Spatial cross-validation was conducted using a leave-one-location-out cross-validation approach,  
406 where data from one monitoring location were withheld as the test set while the model was trained  
407 using data from all remaining locations. This procedure was repeated until each location had served  
408 once as the test set. Locations with limited data coverage (Somanya and Sekondi) were excluded  
409 from the spatial cross-validation analysis because the sparse observations at these sites were  
410 insufficient for robust evaluation of model performance. This approach quantifies the model’s  
411 ability to generalize to regions of Ghana where surface measurements are not available.

412 Temporal cross-validation was performed using the leave-one-year-out cross-validation approach,  
413 where observations from a single year were excluded from training and used exclusively for testing  
414 for each year from 2018 to 2024 of the surface measurement campaign. In each fold, the model  
415 was retrained using the fixed hyperparameters and evaluated on the held-out subset. This approach  
416 assesses the model’s ability to generalize across interannual variability in meteorology, emissions,  
417 and episodic pollution events, increasing confidence in the robustness of PM<sub>2.5</sub> estimates for  
418 periods outside the training data. This step is particularly important because model training is  
419 primarily based on observations from 2018–2024, while PM<sub>2.5</sub> concentrations are estimated  
420 retrospectively back to 2005, requiring the model to perform reliably during time periods not  
421 directly represented in the training dataset.

422 The optimized model from each model type (TROPOMI, OMI, and MERRA) was subsequently  
423 applied to the complete gridded predictor dataset to generate spatially continuous daily PM<sub>2.5</sub>  
424 estimates across the spatiotemporal study domain. For each grid cell  $s$  and day  $t$ , predicted  
425 concentrations were obtained as  $\widehat{PM}_{2.5,s,t} = f(X_{s,t})$ , where  $X_{s,t}$  denotes the vector of satellite,  
426 meteorological, and cyclic predictors. The final output consisted of daily PM<sub>2.5</sub> maps at  $0.01^\circ \times$   
427  $0.01^\circ$  spatial resolution ranging from 2005 to 2025, enabling high-resolution spatiotemporal  
428 exposure assessment for subsequent analyses.

## 429 **2.6 City-level aggregated PM<sub>2.5</sub>**

430 Daily PM<sub>2.5</sub> concentrations were estimated for key locations across Ghana using an area-weighted  
431 spatial averaging approach applied to gridded satellite-derived air quality data. Geographic  
432 coordinates for 20 locations across Ghana were compiled and verified through manual inspection.  
433 These locations include the regional capitals: Bolgatanga (Upper East), Nalerigu (North East), Wa  
434 (Upper West), Tamale (Northern), Damongo (Savannah), Dambai (Oti), Techiman (Bono East),  
435 Sunyani (Bono), Goaso (Ahafo), Kumasi (Ashanti), Ho (Volta), Sefwi Wiawso (Western North),  
436 Koforidua (Eastern), Cape Coast (Central), Sekondi-Takoradi (Western), and Accra (Greater  
437 Accra). Additional locations of interest include Navrongo (Upper East), Kintampo (Bono East),  
438 Somanya (Eastern), and Tema (Greater Accra). To represent urban exposure while accounting for  
439 spatial heterogeneity in city size, circular buffers ranging from 1–9 km were defined around each  
440 city center based on visual assessment of the urban extent. Buffers were constructed in a projected  
441 coordinate system (UTM Zone 30N, EPSG:32630) to ensure accurate distance calculations in  
442 meters and subsequently transformed back to geographic coordinates (EPSG:4326) for  
443 compatibility with the PM<sub>2.5</sub> raster grid. For each city, daily PM<sub>2.5</sub> values were calculated using an  
444 area-weighted mean of all grid cells intersecting the buffer. Fractional overlap between each grid  
445 cell and the buffer polygon was computed to determine pixel-level weights, allowing partial grid  
446 cells along buffer boundaries to contribute proportionally to the average. This approach reduces  
447 bias introduced by binary inclusion or exclusion of edge pixels and provides a spatially

448 representative estimate of population-relevant exposure. The resulting dataset consists of daily  
 449 PM<sub>2.5</sub> time series for each city over the study period, enabling consistent comparisons across  
 450 northern and southern regions of Ghana.

## 451 2.7 Long-term PM<sub>2.5</sub> trend detection

452 To quantify the direction, magnitude, and statistical significance of long-term changes in surface  
 453 PM<sub>2.5</sub> over Ghana, we applied the Mann-Kendall (MK) monotonic trend test and Sen's slope  
 454 estimator to monthly mean PM<sub>2.5</sub> concentrations derived from the multi-model output between  
 455 2005 and 2025.

### 456 2.7.1 Mann-Kendall test

457 The MK test is a rank-based, non-parametric statistical procedure for detecting monotonic trends  
 458 in time series data (Kendall, 1975; Mann, 1945). It makes no assumption about the underlying  
 459 distribution of the data, making it particularly well-suited for environmental datasets, such as  
 460 PM<sub>2.5</sub>, which routinely exhibit non-normal distributions, heteroscedasticity, and intermittent  
 461 extreme values. The test statistic  $S$  is computed as:

$$462 \quad S = \sum_{i=1}^{n-1} \sum_{j=i+1}^n \text{sgn}(x_j - x_i) \dots (4)$$

463 Where  $x_i$  and  $x_j$  are observations at times  $i$  and  $j$  (with  $j > i$ ),  $n$  is the total number of observations,  
 464 and  $\text{sgn}(\cdot)$  is the sign function returning +1, 0, or -1 for positive, zero, or negative differences,  
 465 respectively. Under the null hypothesis of no monotonic trend ( $H_0$ : the data are independently and  
 466 identically distributed with no temporal ordering), the standardized test statistic  $Z$  is given by:

$$467 \quad Z = \begin{cases} \frac{S - 1}{\sqrt{\text{Var}(S)}} & \text{if } S > 0 \\ 0 & \text{if } S = 0 \dots (5) \\ \frac{S + 1}{\sqrt{\text{Var}(S)}} & \text{if } S < 0 \end{cases}$$

468 The variance of  $S$  is adjusted for tied ranks in the data. A statistically significant positive (negative)  
 469  $Z$  indicates a monotonically increasing (decreasing) trend. We adopt a two-tailed significance  
 470 threshold of  $\alpha = 0.05$ , corresponding to  $|Z| > 1.96$ .

### 471 2.7.2 Seasonal Mann-Kendall Test

472 Monthly means were computed by averaging daily gridded PM<sub>2.5</sub> concentrations within each  
 473 calendar month at every 1 km grid cell, yielding a time series of 252 monthly observations per grid  
 474 cell. Monthly resolution was preferred compared to annual aggregation because it provides  
 475 substantially more statistical value for trend detection in Ghana while retaining seasonally resolved  
 476 trend information, such as elevated concentrations during the Harmattan period.

477 Since PM<sub>2.5</sub> in Ghana exhibits strong and recurring seasonal cycles, applying the standard MK test  
 478 directly to the raw monthly time series can conflate underlying long-term trends with seasonal  
 479 periodicity, inflating or suppressing the apparent trend signal. To address this, we employ the  
 480 Seasonal Mann-Kendall (SMK) test from Hirsch and Slack (1984), which is specifically designed  
 481 for time series with seasonal structure.(Hirsch and Slack, 1984) In the SMK framework, the  $S$   
 482 statistic is computed independently for each of the twelve calendar months, comparing, for  
 483 example, all January values across 2005-2025, all February values across these years, and so on.  
 484 The resulting within-season statistics are then summed to yield an overall test statistic:

$$485 \quad S_{\text{total}} = \sum_{g=1}^{12} S_g \dots (6)$$

486 where  $S_g$  is the MK statistic computed for calendar month  $g$ . The variance of  $S_{\text{total}}$  accounts for  
 487 any covariance between months arising from shared seasonal events (e.g., Harmattan onset), and  
 488 the final  $Z$  statistic is derived from this pooled estimate.

### 489 2.7.3 Sen's slope estimator

490 The magnitude of any statistically significant trend was quantified using Sen's slope estimator,  
 491 also known as the Theil-Sen estimator (Sen, 1968), which provides a robust, non-parametric  
 492 measure of the rate of change. Sen's slope  $\beta$  is defined as the median of all pairwise slopes between  
 493 observations:

$$494 \quad \beta = \text{median} \left( \frac{x_j - x_i}{j - i} \right), \quad \forall j > i \dots (7)$$

495  
 496 where  $x_i$  and  $x_j$  are the monthly mean PM<sub>2.5</sub> values for the same calendar month in years  $i$  and  $j$ .  
 497 The resulting slope  $\beta$  is expressed in units of  $\mu\text{g m}^{-3} \text{yr}^{-1}$  and represents the central tendency of  
 498 the trend rate, resistant to the influence of outliers or single anomalous events such as exceptional  
 499 dust episodes.

500 The SMK test and Sen's slope were applied independently to the monthly mean PM<sub>2.5</sub> time series  
 501 at each 1 km grid cell across Ghana to produce spatially resolved maps of trend magnitude and  
 502 significance. Additionally, spatially averaged time series were computed for the national domain  
 503 and for distinct sub-regions of Ghana (northern, middle, and southern belts) to characterize large-  
 504 scale changes in PM<sub>2.5</sub> burden over the 21-year study period.

## 505 3 RESULTS AND DISCUSSION

### 506 3.1 Surface PM<sub>2.5</sub> measurements

507 The daily PM<sub>2.5</sub> concentrations collected by surface monitors during the measurement campaign  
 508 (2018–2024) exhibit a mean concentration of  $31.8 \mu\text{g m}^{-3}$ , indicating consistently elevated  
 509 pollution levels across Ghana. The distribution of observed daily PM<sub>2.5</sub> concentrations is strongly  
 510 right-skewed, characterized by a long tail toward higher concentration values, as shown in Fig. S6

511 of the Supplementary Materials. The 95<sup>th</sup> percentile PM<sub>2.5</sub> levels reached 81.1  $\mu\text{g m}^{-3}$ , 2.6 times  
 512 the mean concentration which highlights the frequent occurrence of episodic high-pollution events.  
 513 The large proportion of Harmattan-season observations at higher PM<sub>2.5</sub> concentrations suggests  
 514 that these extreme values are strongly associated with Harmattan dust transport during the dry  
 515 season. The skewed distribution indicates that while most observations fall within moderate  
 516 concentration ranges, a smaller proportion of high-concentration events contributes  
 517 disproportionately to population exposure.

518 Fig. S7 shows violin distributions of measured daily PM<sub>2.5</sub> concentrations across all cities with  
 519 surface monitoring locations, revealing pronounced spatial heterogeneity in mean daily PM<sub>2.5</sub>. The  
 520 embedded box plots indicate that median concentrations at all locations range between 15–40  $\mu\text{g m}^{-3}$ ,  
 521 consistently exceeding the WHO 24-hour guideline of 15  $\mu\text{g m}^{-3}$ .

### 522 3.2 Model performance

523 Model performance was rigorously evaluated across five machine learning algorithms, LASSO,  
 524 RF, SVM, XGBoost and MLP, for three model configurations TROPOMI model, OMI model, and  
 525 MERRA model, under random, temporal and spatial cross-validation (CV) schemes. MLP, also  
 526 known as a fully connected neural network, is a neural network. The MLP model architecture used  
 527 in this study is shown in Fig. S8 in the Supplementary Materials, whereas optimized  
 528 hyperparameter information for each ML algorithm and mode configuration is summarized in  
 529 Tables S5 to S7. Model performances metrics, including Pearson correlation coefficient (r),  
 530 coefficient of determination ( $R^2$ ), root-mean square error (RMSE) and mean-normalized RMSE  
 531 (nRMSE), for all machine learning algorithms evaluated for the three model configurations are  
 532 summarized in Table 1. Mean bias, mean absolute error (MAE), and mean-normalized MAE  
 533 (nMAE) are presented in Tables S8–S10 as additional error metrics.

534 **Table 1. Performance comparison of machine learning algorithms TROPOMI, OMI, and**  
 535 **MERRA models.** Model performance is evaluated under random, temporal, and spatial cross-  
 536 validation using the coefficient of determination ( $R^2$ ), Pearson correlation coefficient (r), bias, root  
 537 mean squared error (RMSE), and normalized RMSE (nRMSE). Error metrics (RMSE and bias)  
 538 are reported in  $\mu\text{g m}^{-3}$ . Additional error metrics are listed in Tables S8 to S10. Models selected for  
 539 generating PM<sub>2.5</sub> concentration maps are indicated with an asterisk (\*).

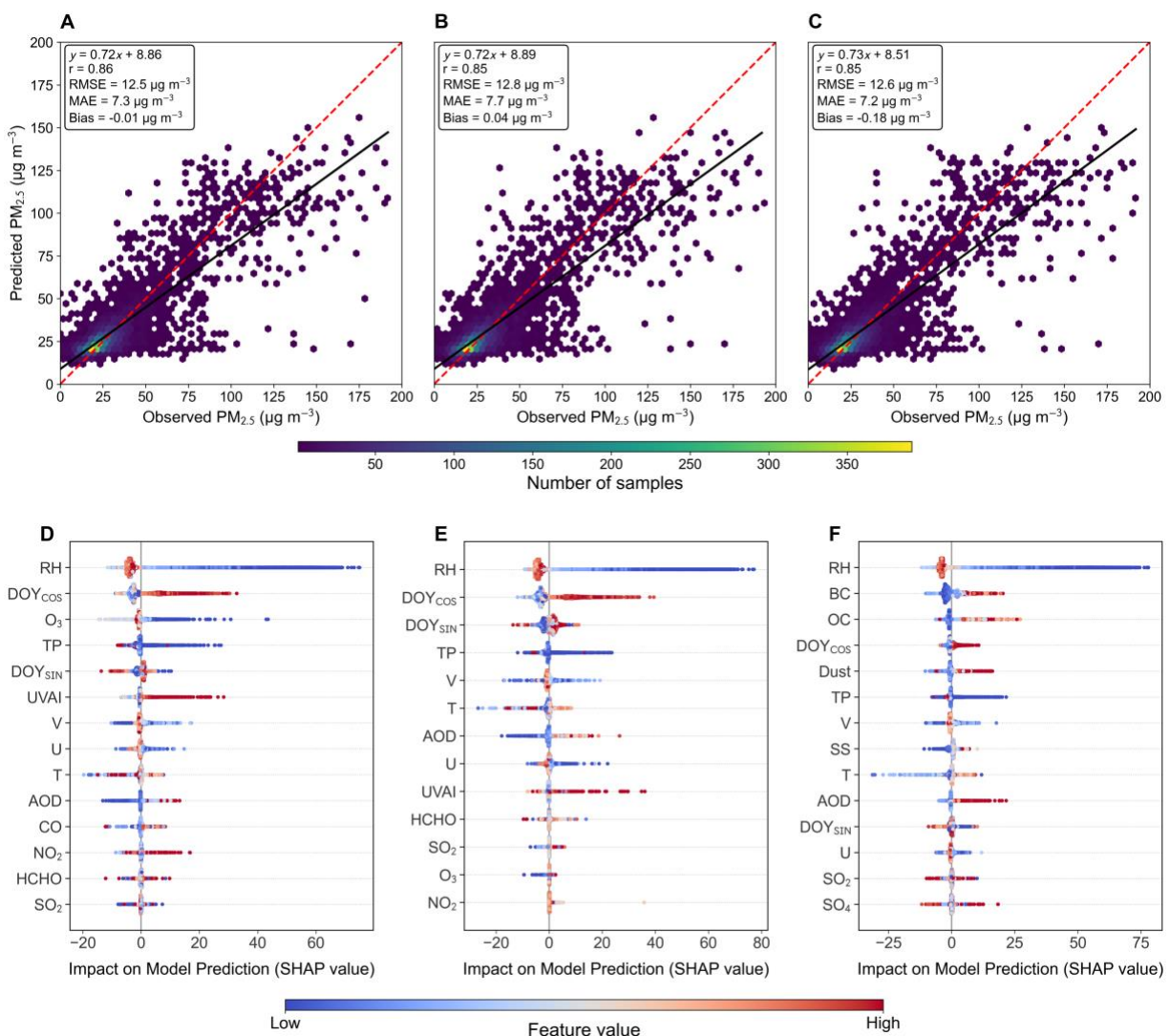
Algorithms	Random CV			Temporal CV			Spatial CV		
	$R^2$ (r)	RMSE (nRMSE)	Bias	$R^2$ (r)	RMSE (nRMSE)	Bias	$R^2$ (r)	RMSE (nRMSE)	Bias
<i>TROPOMI Model</i>									
LASSO	0.60 (0.78)	15.0 (0.48)	-0.17	0.56 (0.75)	15.7 (0.50)	0.33	0.45 (0.68)	19.8 (0.57)	2.96
SVM	0.69 (0.83)	13.2 (0.42)	-1.55	0.55 (0.77)	15.8 (0.50)	-0.36	0.52 (0.75)	19.4 (0.55)	-5.91

RF	0.71 (0.85)	12.7 (0.40)	0.09	0.55 (0.77)	15.9 (0.50)	0.77	0.56 (0.76)	18.4 (0.53)	-1.52
<b>XGB*</b>	0.72 (0.86)	12.5 (0.40)	-0.01	0.57 (0.78)	15.6 (0.49)	0.66	0.57 (0.76)	17.3 (0.49)	-0.93
MLP	0.71 (0.84)	12.8 (0.40)	-0.12	0.54 (0.76)	16.0 (0.51)	-1.56	0.42 (0.66)	18.7 (0.54)	-2.75
<i><b>OMI Model</b></i>									
LASSO	0.58 (0.76)	15.4 (0.49)	-0.19	0.53 (0.73)	16.3 (0.52)	0.35	0.38 (0.64)	21.7 (0.59)	-4.19
SVM	0.68 (0.83)	13.4 (0.42)	-1.79	0.52 (0.75)	16.4 (0.52)	-1.31	0.48 (0.73)	20.2 (0.57)	-5.55
RF	0.71 (0.84)	13.0 (0.41)	-0.16	0.53 (0.75)	16.3 (0.52)	1.46	0.48 (0.75)	19.3 (0.55)	-1.53
<b>XGB*</b>	0.72 (0.85)	12.8 (0.41)	0.04	0.56 (0.76)	15.6 (0.50)	0.75	0.54 (0.75)	18.9 (0.54)	-1.01
MLP	0.70 (0.83)	13.3 (0.42)	-1.19	0.50 (0.71)	16.6 (0.53)	1.78	0.46 (0.70)	19.8 (0.57)	-1.83
<i><b>MERRA Model</b></i>									
LASSO	0.63 (0.79)	14.5 (0.46)	-0.19	0.58 (0.75)	15.2 (0.48)	-0.06	0.41 (0.67)	18.2 (0.57)	-4.63
SVM	0.70 (0.83)	12.7 (0.41)	-1.68	0.55 (0.76)	15.9 (0.50)	-1.17	0.37 (0.66)	19.0 (0.60)	5.60
RF	0.71 (0.84)	12.6 (0.40)	0.38	0.51 (0.75)	16.6 (0.53)	1.37	0.47 (0.70)	17.2 (0.54)	2.16
<b>XGB*</b>	0.72 (0.85)	12.6 (0.40)	-0.18	0.59 (0.78)	15.2 (0.48)	0.03	0.49 (0.73)	16.8 (0.50)	0.39
MLP	0.70 (0.83)	12.9 (0.42)	1.03	0.55 (0.74)	16.0 (0.51)	1.92	0.44 (0.69)	18.0 (0.55)	-1.54

540

541 Under random cross-validation, XGBoost achieved the best and consistent performance across all  
542 three model configurations, with  $R^2$  of 0.72 for TROPOMI, OMI, and MERRA models and RMSE

543 values of 12.5, 12.8, and 12.6  $\mu\text{g m}^{-3}$ , respectively. Linear models (LASSO) consistently showed  
 544 the weakest performance, with  $R^2$  approximately 0.09–0.12 lower than XGBoost and substantially  
 545 higher RMSE across all cross-validation schemes, reflecting their limited ability to capture the  
 546 non-linear relationships of surface  $\text{PM}_{2.5}$  with meteorological and satellite-derived aerosol  
 547 predictors in the heterogeneous emission environments in Ghana. Support vector machines (SVM)  
 548 showed competitive random-CV performance but degraded substantially under spatial and  
 549 temporal cross-validation. Among the non-linear approaches, tree-based ensemble methods RF  
 550 and XGBoost performed comparably under random cross-validation, but XGBoost exhibited  
 551 consistently lower bias ( $-0.01$ ,  $0.04$ , and  $-0.18 \mu\text{g m}^{-3}$  for TROPOMI, OMI and MERRA models,  
 552 respectively) and comparable or lower normalized errors across all CV schemes. The regression  
 553 slopes in Figs. 2A–C (0.72–0.73) further indicate that XGB predictions closely track observed  
 554 variability in  $\text{PM}_{2.5}$ , with only minor underestimation at higher  $\text{PM}_{2.5}$  concentrations.



555  
 556 **Fig. 2. Model performance and interpretability of XGBoost  $\text{PM}_{2.5}$  prediction models. (A–C)**  
 557 Scatterplots show the relationship between daily observed  $\text{PM}_{2.5}$  concentrations from surface  
 558 monitors and corresponding model estimates for TROPOMI, OMI, and MERRA-2 models. Each

559 point represents paired daily observations from surface monitors and corresponding model  
560 estimates. The red dashed line denotes the 1:1 reference line, while the solid black line represents  
561 the fitted linear regression. All models demonstrate strong agreement between observations and  
562 predictions ( $r \approx 0.84\text{--}0.85$ ) with slopes close to unity, indicating good representation of variability  
563 and slight underestimation at higher  $\text{PM}_{2.5}$  concentrations. Color shading indicates the density of  
564 samples. **(D–F)** SHAP summary plots show the relative importance and direction of feature  
565 contributions to  $\text{PM}_{2.5}$  predictions for TROPOMI, OMI, and MERRA-2 models. Each point  
566 represents an individual sample, colored according to the feature value (blue = low, red = high).  
567 The horizontal spread of points indicates the magnitude and direction of each feature’s influence  
568 on model output, with features ordered by overall importance.

569 Consistent with performances of spatiotemporal models in other studies, model performance  
570 declined under temporal and spatial cross-validation relative to random cross-validation across all  
571 algorithm and model types, which reflects additional challenge of generalizing to time periods or  
572 geographic locations unfamiliar to the models (Chi et al., 2022). Under temporal cross-validation,  
573 XGBoost shows  $R^2$  values of 0.57, 0.56, and 0.59 TROPOMI, OMI, and MERRA models, with  
574 RMSE increasing to 15.6, 15.6, and 15.2  $\mu\text{g m}^{-3}$ , respectively. Under spatial cross-validation,  
575 arguably the most operationally relevant metric for applying the model to unmonitored grid cells  
576 across Ghana, XGBoost  $R^2$  values were 0.57, 0.54, and 0.49 for TROPOMI, OMI, and MERRA  
577 models. The MLP model showed the largest degradation under spatial cross-validation, with  $R^2$   
578 falling to 0.45, 0.38, and 0.41 for TROPOMI, OMI, and MERRA models, indicating greater  
579 susceptibility to overfitting to sensor-specific concentration patterns when the training sample is  
580 limited to a sparse monitoring network. In addition to more robust spatial and temporal  
581 generalization of XGBoost, better interpretability through SHAP-based analysis provides insight  
582 into the relative contributions of meteorological and aerosol predictors, further supporting its  
583 suitability for generating spatially continuous daily  $\text{PM}_{2.5}$  maps across unmonitored regions and  
584 time periods of Ghana. Therefore, XGBoost was selected as the final model for further analysis.  
585 Figs. 2A–C further illustrates the agreement between observed and predicted  $\text{PM}_{2.5}$  concentrations  
586 for the best-performing model (XGBoost) under random CV, showing strong clustering along the  
587 1:1 line and limited systematic bias across the concentration range.

588 Among the three model schemes, the MERRA model achieved the lowest spatial and temporal  
589 cross-validation RMSE values of 15.2  $\mu\text{g m}^{-3}$  and 16.8  $\mu\text{g m}^{-3}$ , respectively. However, spatially  
590 cross-validated  $R^2$  for the MERRA model (0.49) is slightly lower than that of the other models  
591 (0.54–0.57), suggesting that the lower absolute RMSE is partly attributable to the reduced  $\text{PM}_{2.5}$   
592 dynamic range in this gap-filling dataset due to inherently larger grid sizes of MERRA-2 dataset.  
593 The higher temporally cross-validated  $R^2$  of 0.59 indicates improved representation of diel  
594 variability in aerosol-related predictors from MERRA-2 compared to only once-a-day  
595 measurements from TROPOMI and OMI. Nonetheless, the MERRA model provides physically  
596 consistent estimates during periods of satellite data unavailability, ensuring complete  
597 spatiotemporal coverage in the final merged product.

598 SHAP analysis plots in Figs. 2D–2F provide insights into the influence of predictor variables on  
599  $\text{PM}_{2.5}$  concentrations. The results indicate that meteorological and seasonal predictors dominate  
600  $\text{PM}_{2.5}$  variability in Ghana. Relative humidity exhibited the strongest influence on model  
601 predictions, with low-humidity conditions strongly associated with elevated  $\text{PM}_{2.5}$  concentrations.  
602 This asymmetric and wide range of SHAP response under dry conditions suggests that low-RH

603 acts as a strong indicator of Harmattan season and contributes positively to ambient PM<sub>2.5</sub>  
604 concentrations due to enhanced dust transport and low rainfall during the dry season. In contrast,  
605 high-humidity conditions during the wet seasons correspond predominantly to negative SHAP  
606 values, consistent with efficient wet scavenging and reduced aerosol loading under increased  
607 precipitation and enhanced aerosol removal.

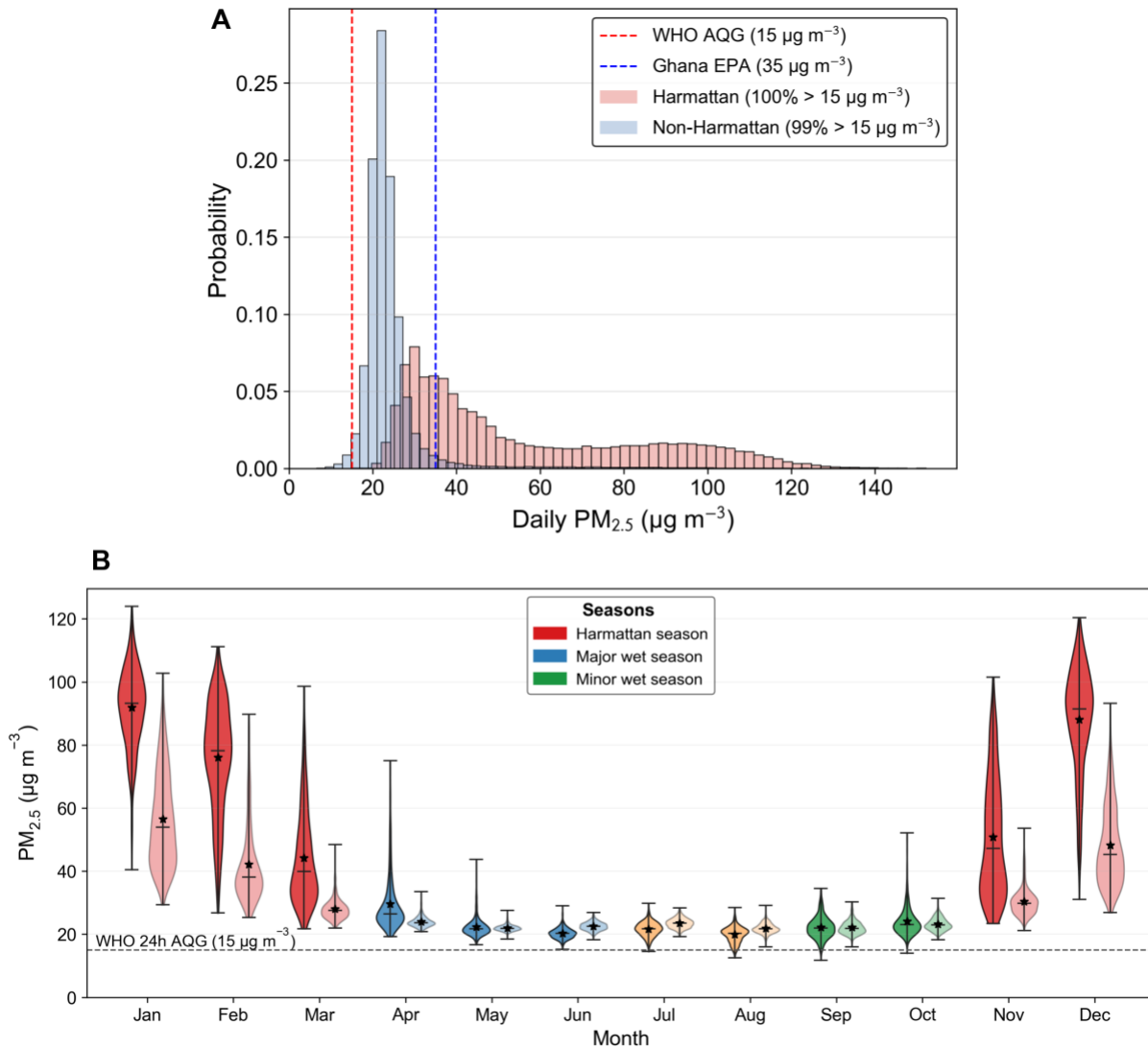
608 The seasonal cyclic features represented by the sine and cosine transformations of DOY exhibited  
609 large absolute SHAP values distributed across both positive and negative directions, indicating  
610 that the model successfully captured the strong annual periodicity of PM<sub>2.5</sub> concentrations in  
611 Ghana. Wind speeds (U and V) and precipitation variables from ERA5-Land also contributed  
612 through their influence on dust transport intensity and wet deposition of aerosols, respectively.  
613 These results are broadly consistent with findings from a recent satellite-based PM<sub>2.5</sub> study in West  
614 Africa by Westervelt et al. (2025), which identified meteorological drivers and dust transport as  
615 major determinants of surface aerosol loading.

616 Satellite-derived aerosol indicators, particularly the ultraviolet aerosol index (UVAI), showed  
617 consistently positive SHAP contributions in both the TROPOMI and OMI models, indicating  
618 higher predicted PM<sub>2.5</sub> concentrations under elevated absorbing aerosol conditions. These  
619 conditions are commonly associated with mineral dust transport during the Harmattan season and  
620 episodic biomass burning, highlighting the importance of absorbing aerosol loading in shaping  
621 surface PM<sub>2.5</sub> variability. For the MERRA-2-based model, PM composition variables, particularly  
622 BC, OC, and dust, exhibited increased importance alongside meteorological predictors. The strong  
623 contributions from dust-related variables are consistent with the influence of Harmattan-driven  
624 mineral dust transport, while BC and OC likely reflect contributions from combustion-related  
625 aerosol sources, including biomass burning and other anthropogenic emissions. SHAP values for  
626 AOD suggest that the column-integrated aerosol loading represented by AOD in the MERRA-2  
627 reanalysis provides valuable predictive information, particularly during high-PM<sub>2.5</sub> episodes,  
628 similar to the role of UVAI in OMI- and TROPOMI-based models. Despite differences in predictor  
629 types among the three model configurations, the overall consistency in dominant drivers provides  
630 confidence that the modeling framework captures physically meaningful controls on PM<sub>2.5</sub>  
631 variability across seasons and regions.

### 632 **3.3 Daily PM<sub>2.5</sub> distribution and long-term spatiotemporal trends**

633 The daily PM<sub>2.5</sub> concentrations derived at 1 km × 1 km resolution for 2005–2025 were used for  
634 assessing spatial and long-term patterns. Daily PM<sub>2.5</sub> concentration distributions for Harmattan  
635 and non-Harmattan periods are illustrated in Fig. 3A. During the non-Harmattan season, the  
636 distribution is relatively narrow, with a mode around 20 µg m<sup>-3</sup> and limited probability mass  
637 extending beyond 60 µg m<sup>-3</sup>, reflecting more consistent and moderate pollution conditions driven  
638 primarily by local and regional anthropogenic sources. In contrast, the Harmattan season  
639 distribution is characteristically broader, with a mode ~69 µg m<sup>-3</sup> and a pronounced long tail  
640 extending to around 150 µg m<sup>-3</sup>, strongly indicating episodic high-concentration events associated  
641 with large amounts of mineral dust transport from the Sahara and Sahel to the West African regions  
642 during this season. Notably, both distributions are almost entirely (>99%) situated above the WHO  
643 24-hour air quality guideline of 15 µg m<sup>-3</sup> (dashed red line), underscoring that PM<sub>2.5</sub> exceedances  
644 in Ghana are not confined to the dust season but represent a year-round public health concern due  
645 to high exposure to ambient particulate matter. The wider spread and heavier tail of the Harmattan-

646 season distribution imply that PM<sub>2.5</sub> exposure among the Ghanaian population during this period  
 647 is not only substantially more variable, but also consistently higher, with extreme pollution  
 648 episodes frequently reaching concentration 2-10 times higher than the WHO 24-hour guideline.



649

650 **Fig. 3. Seasonal distribution and monthly variability of daily PM<sub>2.5</sub> concentrations across**  
 651 **Ghana (2005–2025).** (a) Probability distributions of daily PM<sub>2.5</sub> concentrations during Harmattan  
 652 (red) and non-Harmattan (blue) periods. Harmattan conditions exhibit a broader distribution and  
 653 substantially higher concentrations, reflecting the influence of long-range Saharan dust transport.  
 654 The red and blue dashed vertical lines indicate the 24-hour air quality guidelines from WHO (15  
 655 µg m<sup>-3</sup>) and Ghana EPA (35 µg m<sup>-3</sup>), highlighting extremely high number of exceedances,  
 656 particularly during Harmattan months. (b) Monthly climatology of daily PM<sub>2.5</sub> concentrations for  
 657 Harmattan, major wet, and minor wet seasons across Northern (dark shading) and Southern (light  
 658 shading) Ghana. Violin plots illustrate the distribution of daily values for each month. The central  
 659 horizontal line within each violin denotes the median, and the star marker indicates the monthly  
 660 mean concentration. The dashed horizontal line represents the WHO 24-hour air quality guideline

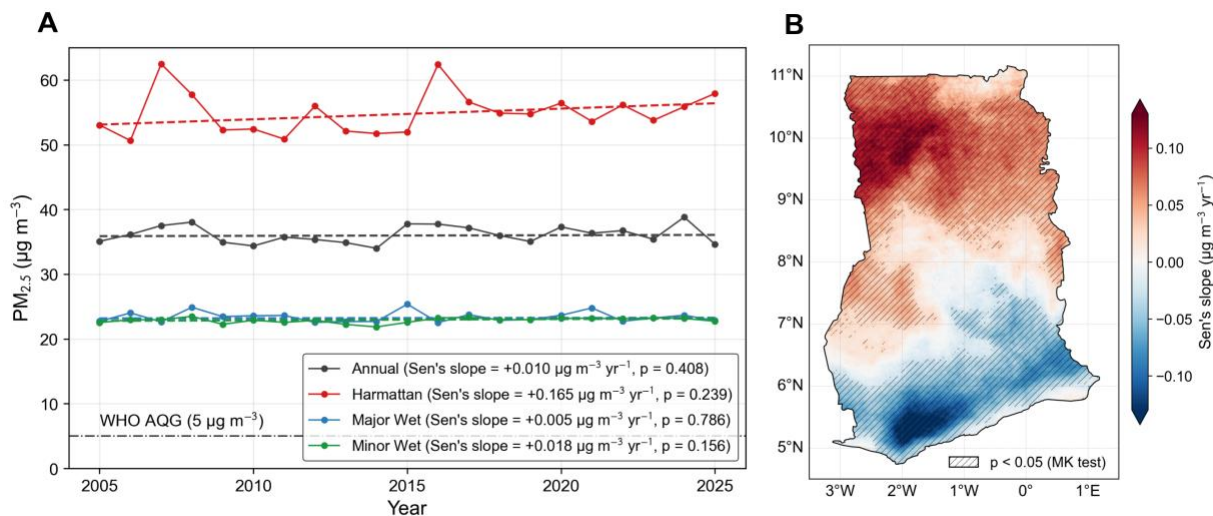
661 (15  $\mu\text{g m}^{-3}$ ), showing that exceedances occur predominantly during Harmattan months, particularly  
662 in Northern Ghana.

663 These distributions for 20 locations, including regional capitals, in Ghana are shown in Fig. S9.  
664 The northern region shows a strong characteristic normal  $\text{PM}_{2.5}$  distribution with a wider spread  
665 during the Harmattan period, opposed to a narrow distribution and much lower means for southern  
666 regions of Ghana. Big cities of Ghana, such as Accra, Cape Coast, Sekondi–Takoradi and Kumasi,  
667 are mostly located in the South, and show a much narrower  $\text{PM}_{2.5}$  distribution with less pronounced  
668 seasonal variability during this period. On the other hand, rural or sub-urban locations exhibit  
669 relatively broader  $\text{PM}_{2.5}$  distribution, potentially reflecting greater variability in emission sources,  
670 including episodic agricultural burning and savannah fires during the dry season (Ofosu et al.,  
671 2013).

672 Fire activity is an additional important contributor to seasonal  $\text{PM}_{2.5}$  variability in Ghana,  
673 particularly across northern and transitional ecological zones dominated by savanna land cover  
674 (Fig. S10). Analysis of the MODIS MCD12Q1 Version 6.1 land cover observations in 2024  
675 indicates that savannas (42%), grasslands (27%), and woody savannas (10%) together account for  
676 nearly 80% of Ghana’s land area, with these ecosystems concentrated primarily in northern Ghana  
677 and the forest–savanna transition belt (Fig. S11). Fire detections derived from MODIS active fire  
678 products for 2005–2025 show that fire occurrence is spatially concentrated in these savanna-  
679 dominated regions, consistent with widespread seasonal biomass burning associated with  
680 agricultural land management, pasture renewal, and accidental wildfires. Annual mean fire  
681 frequency and fire radiative power (FRP), a proxy for combustion intensity, both show pronounced  
682 seasonal dependence, with substantially higher activity and FRP during the dry Harmattan period  
683 (December–February) relative to the wetter non-Harmattan months. The elevated frequency and  
684 intensity of fires during the Harmattan season likely enhance regional aerosol loading through  
685 increased emissions of carbonaceous particulate matter, contributing to the broader and more right-  
686 skewed  $\text{PM}_{2.5}$  distribution observed during this period. Frequent fire occurrences concentrated  
687 predominantly within the savanna regions of Ghana suggest that biomass burning may contribute  
688 to seasonal aerosol loading and  $\text{PM}_{2.5}$  variability in Ghana, complementing the dominant influence  
689 of long-range Saharan dust transport during Harmattan conditions (Ofosu et al., 2013).

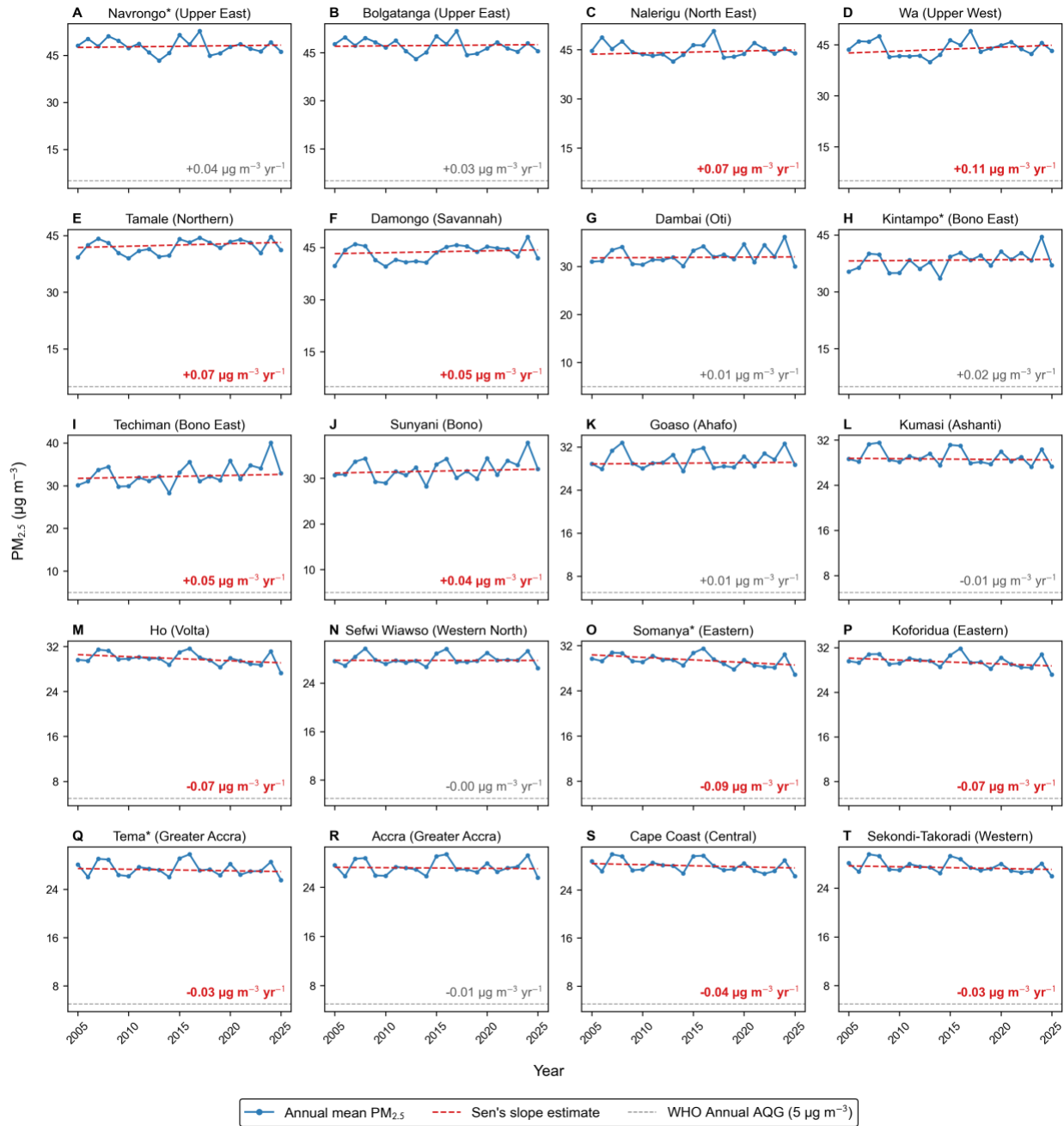
690 This north-south difference in  $\text{PM}_{2.5}$  exposure levels is even more evident from monthly  $\text{PM}_{2.5}$   
691 climatology averaged over the 21-year study period (2005–2025), disaggregated by northern and  
692 southern regions of Ghana shown in Fig. 3B. Ghana is classified into northern and southern  
693 subregions, defined as regions above and below 8° N latitude, respectively (Klutse et al., 2020). A  
694 pronounced seasonal cycle can be observed for monthly mean  $\text{PM}_{2.5}$  in Ghana, with peak  
695 concentrations occurring from November through March, coinciding with the Harmattan season.  
696 Typically, North experiences 1.63 to 1.83 times higher PM exposure compared to the South.  $\text{PM}_{2.5}$   
697 concentrations decline sharply with the onset of the major wet season in April–June, driven by  
698 enhanced wet removal of atmospheric particles and suppressed dust transport. A secondary  
699 minimum occurs during the minor wet season (September–October), separated from the major wet  
700 season by a brief mid-season dry period in July–August during which  $\text{PM}_{2.5}$  concentrations exhibit  
701 a modest secondary peak. This bimodal wet season structure is more pronounced in southern  
702 Ghana, consistent with the bimodal rainfall regime associated with the seasonal northward and  
703 southward migration of the Intertropical Convergence Zone (ITCZ) over the coastal and forested  
704 zones of West Africa (Nicholson, 2018). In contrast, northern Ghana exhibits a unimodal rainfall

705 pattern peaking in August and September (Fig. S12), during which monthly mean PM<sub>2.5</sub>  
 706 concentrations (19.9  $\mu\text{g m}^{-3}$ ) are lower than those observed in the south (21.0  $\mu\text{g m}^{-3}$ ).



707  
 708 **Fig. 4. Long-term spatiotemporal trends in ambient PM<sub>2.5</sub> concentrations across Ghana from**  
 709 **2005 to 2025.** (a) Ghana-wide annual and seasonal mean PM<sub>2.5</sub> concentrations, including  
 710 Harmattan, Major Wet, and Minor Wet seasons. Dashed lines represent trends estimated using the  
 711 Theil–Sen slope estimator, with corresponding slope magnitudes and Mann–Kendall test p-values  
 712 reported in the legend. The horizontal dash–dotted line indicates the WHO annual air quality  
 713 guideline (AQG) of 5  $\mu\text{g m}^{-3}$ . (b) Spatial distribution of pixel-wise PM<sub>2.5</sub> trends estimated using  
 714 the Theil–Sen slope estimator. Stippling denotes statistically significant trends ( $p < 0.05$ ) based on  
 715 the Mann–Kendall test. Positive values indicate increasing PM<sub>2.5</sub> concentrations over time, while  
 716 negative values indicate decreasing trends.

717 Fig. 4A shows the seasonal and annual evolution of PM<sub>2.5</sub> in Ghana over the 21-year period.  
 718 Annual mean PM<sub>2.5</sub> has remained relatively stable over the study period with the Sen's slope  
 719 estimate indicating an average annual increase of only 0.01  $\mu\text{g m}^{-3}$  every year and seasonal slopes  
 720 varying between 0.005–0.165  $\mu\text{g m}^{-3} \text{ yr}^{-1}$ . Mann-Kendall's test suggests that these seasonal and  
 721 yearly trends observed in Ghana are not statistically significant ( $p \geq 0.05$ ). However, spatial map of  
 722 Sen's slope estimate for Ghana at 1 km  $\times$  1 km grid resolution in Fig. 4B indicates the majority of  
 723 Ghana has experienced a statistically significant trend. The Sen's slope estimates and  $p$  values  
 724 calculated from Mann–Kendall test is summarized in Table S11.



725

726 **Fig. 5. Temporal trends in annual mean PM<sub>2.5</sub> concentrations across Ghanaian regional**  
 727 **capitals from 2005–2025.** (A–T) Each panel shows the yearly average PM<sub>2.5</sub> concentration (blue  
 728 line with markers), Sen’s slope trend (red dashed line), and the WHO annual PM<sub>2.5</sub> guideline of 5  
 729 µg m<sup>-3</sup> (grey dashed line) for regional capitals and non-regional capitals (\*). The estimated Sen’s  
 730 slope (µg m<sup>-3</sup> yr<sup>-1</sup>) is shown in the lower-right corner of each panel. Slope values with statistically  
 731 significant trends ( $p \leq 0.05$ ) assessed by Mann–Kendall test is indicated in bold red values. Results  
 732 indicate generally stable or modestly increasing PM<sub>2.5</sub> levels across most cities, with several  
 733 statistically significant increasing trends observed, while annual mean concentrations in all cities  
 734 consistently exceed the WHO AQG threshold throughout the study period.

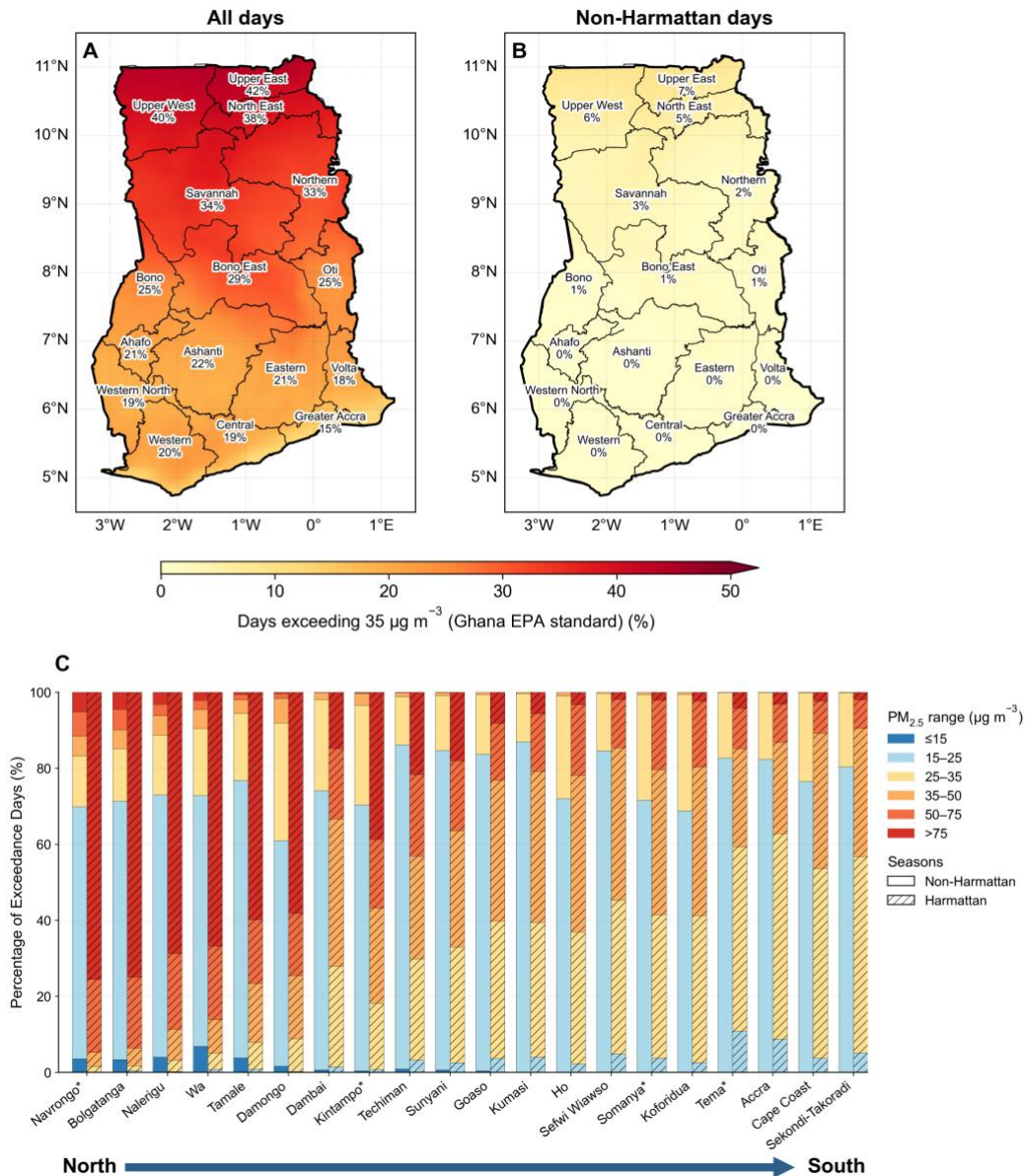
735 City scale trend plots in Fig. 5 show, increasing PM<sub>2.5</sub> trends (0.05–0.11  $\mu\text{g m}^{-3} \text{yr}^{-1}$ ) over the 21-  
736 year study period in some suburban and rural regions such as Nalerigu, Wa, and Damongo in  
737 northern Ghana. Another region exhibiting increasing PM<sub>2.5</sub> trends is Bono and Bono East, where  
738 cities such as Sunyani and Techiman may reflect increasing emissions associated with  
739 anthropogenic activities in these rapidly developing urban centers in addition to contributions from  
740 Harmattan-related PM<sub>2.5</sub> episodes. In contrast, several southern cities exhibit statistically  
741 significant declining trends, with the strongest decreases observed in Somanya ( $-0.090 \mu\text{g m}^{-3} \text{yr}^{-1}$ ,  $p < 0.001$ ),  
742 Ho ( $-0.073 \mu\text{g m}^{-3} \text{yr}^{-1}$ ,  $p < 0.001$ ), and Koforidua ( $-0.070 \mu\text{g m}^{-3} \text{yr}^{-1}$ ,  $p < 0.001$ ),  
743 followed by Cape Coast ( $-0.036 \mu\text{g m}^{-3} \text{yr}^{-1}$ ), Sekondi-Takoradi ( $-0.029 \mu\text{g m}^{-3} \text{yr}^{-1}$ ), and Tema  
744 ( $-0.025 \mu\text{g m}^{-3} \text{yr}^{-1}$ ), which may be associated with improvements in vehicular fuel quality (Ayetor  
745 et al., 2025) and gradual adoption of cleaner household energy sources, potentially reflecting more  
746 effective policy implementation in the urbanized southern coastal regions of Ghana. Although  
747 these trends are statistically significant, their cumulative magnitudes over the 21-year period  
748 (approximately 0.5–1.9  $\mu\text{g m}^{-3}$ ) are modest relative to the model's daily spatial prediction  
749 uncertainty, which ranges from approximately 7–21  $\mu\text{g m}^{-3}$  RMSE across southern monitoring  
750 sites (Fig. S13). However, the trend detection analysis was performed on annual means, for which  
751 unsystematic daily errors are largely attenuated through temporal averaging, reducing the effective  
752 uncertainty to only a fraction of the daily RMSE. Thus, the Mann-Kendall test can reliably identify  
753 these directional changes. A map of leave-one-site-out cross-validation RMSE at the monitoring  
754 locations is provided in Fig. S13 to contextualize model uncertainty in estimating daily PM<sub>2.5</sub>  
755 across Ghana.

756 Although long-term changes in PM<sub>2.5</sub> across Ghanaian suburban and urban locations are modest,  
757 typically ranging between  $-0.2 \mu\text{g m}^{-3} \text{yr}^{-1}$  to  $0.2 \mu\text{g m}^{-3} \text{yr}^{-1}$ , these trends reveal a clear and  
758 widening north-south divergence in PM<sub>2.5</sub> exposure over the 21-year record. Positive trends in  
759 northern Ghana contrast with stable or declining trends in the south, reflecting differences in  
760 dominant drivers of PM<sub>2.5</sub> variability across regions. In northern Ghana, increases are consistent  
761 with the continued influence of Harmattan dust, regionally transported aerosols, and Savannah  
762 fires, whereas several southern urban centers show stable or declining concentrations indicative of  
763 gradual changes in local emission patterns. At the national scale, the strong influence of  
764 interannual variability in Harmattan dust loading limits the detectability of statistically significant  
765 Ghana-wide trends, making it challenging to isolate long-term emission-driven changes from  
766 meteorological variability.

767 Trends during the non-Harmattan season are broadly consistent with the Seasonal Mann–Kendall  
768 results (Table S11) and provide additional insight into long-term PM<sub>2.5</sub> variability after accounting  
769 for the dominant seasonal dust cycle. Several southern cities, including Somanya, Ho, Koforidua,  
770 Tema, Cape Coast, and Sekondi-Takoradi, exhibit statistically significant declines in non-  
771 Harmattan PM<sub>2.5</sub> concentrations, suggesting gradual improvements in baseline air quality  
772 conditions outside the dust season. In contrast, northern cities such as Wa and Tamale show  
773 persistent positive non-Harmattan trends, indicating that factors beyond long-range dust transport,  
774 including local emissions and biomass burning, contribute to sustained PM<sub>2.5</sub> exposure levels  
775 throughout the year. The spatial heterogeneity in non-Harmattan trends reinforces the north-south  
776 divergence observed in annual trends and highlights the combined influence of regional  
777 atmospheric processes and evolving local emission sources on long-term PM<sub>2.5</sub> variability across  
778 Ghana.

779 **3.4 Exceedances for air quality guidelines**

780 The spatial distribution of exceedance frequency relative to Ghana EPA 24-hour guideline ( $35 \mu\text{g}$   
781  $\text{m}^{-3}$ ) is shown in Fig. 6. During 2005–2025 (Fig. 6A), exceedances occur on approximately 15–  
782 20% of days across southern Ghana and 35–42% of days in northern regions. Restricting the  
783 analysis to non-Harmattan periods (Fig. 6B) substantially lowers exceedance frequency, with little  
784 to no exceedance in the south and fewer than 10% of days exceeding the guideline in northern  
785 Ghana. In contrast, exceedance frequencies relative to the WHO 24-hour guideline ( $15 \mu\text{g m}^{-3}$ ),  
786 shown in Fig. S14, remain above 90% across entire Ghana, including the southern coastal zone,  
787 reflecting the widespread and year-round burden of ambient  $\text{PM}_{2.5}$  across Ghana in both Harmattan  
788 and non-Harmattan periods. Persistently high exceedance frequencies in major urban centers such  
789 as Accra, Kumasi, and Tema indicate that anthropogenic emissions contribute to sustained  $\text{PM}_{2.5}$   
790 exposure beyond the Harmattan season, highlighting the year-round burden of ambient air  
791 pollution across Ghana.



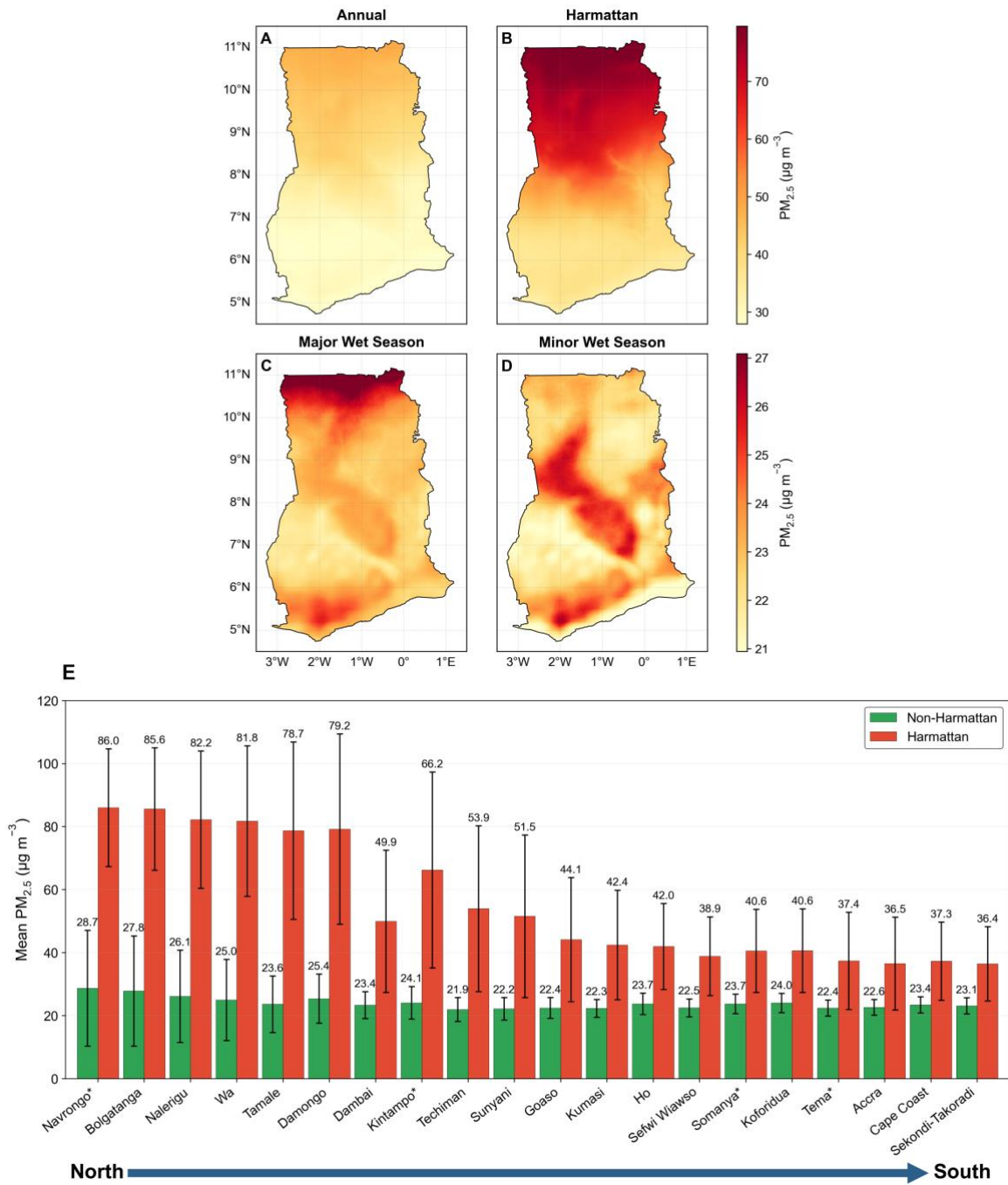
792

793 **Fig. 6. Ghana-wide exceedance frequency of daily PM<sub>2.5</sub> air quality guidelines.** Spatial  
 794 distribution of the percentage of days exceeding the Ghana EPA PM<sub>2.5</sub> guideline (35 μg m<sup>-3</sup>) across  
 795 Ghana during 2005–2025. Panels show exceedance frequency calculated using (A) all available  
 796 days and (B) non-Harmattan days. Values represent the proportion of days (%) at each grid cell  
 797 where daily PM<sub>2.5</sub> concentrations exceeded 35 μg m<sup>-3</sup>. Plot C shows the percentage of days  
 798 exceeding daily PM<sub>2.5</sub> for each concentration range (≤15, 15–25, 25–35, 35–50, 50–75, and >75  
 799 μg m<sup>-3</sup>) across WHO and Ghana EPA air quality guideline categories during non-Harmattan and  
 800 Harmattan seasons for locations across Ghana (2005–2025). Cities are ordered geographically  
 801 from north (left) to south (right) with non-regional capitals indicated by an asterisk (\*).

802 The city-level frequency distribution of daily PM<sub>2.5</sub> concentration ranges, shown in the bar plot in  
 803 Fig. 6C, further quantifies the north-south contrast. The bars are categorized using 24-hour

804 thresholds of the WHO Air Quality Guideline ( $15 \mu\text{g m}^{-3}$ ), WHO Interim Target 4 ( $25 \mu\text{g m}^{-3}$ ),  
805 Ghana EPA standard ( $35 \mu\text{g m}^{-3}$ ), WHO Interim Target 2 ( $50 \mu\text{g m}^{-3}$ ), and WHO Interim Target 1  
806 ( $75 \mu\text{g m}^{-3}$ ). During the Harmattan season, northern cities record substantially higher proportions  
807 of days exceeding 50 and  $75 \mu\text{g m}^{-3}$  compared to southern cities, while even during non-Harmattan  
808 conditions, fewer than 10% of days in any Ghanaian city fall below the WHO 24-hour guideline  
809 of  $15 \mu\text{g m}^{-3}$ . The annual frequency corresponding to each concentration range for these cities is  
810 shown in Fig. S15. Collectively, these exceedance patterns demonstrate that ambient  $\text{PM}_{2.5}$  in  
811 Ghana constitutes a pervasive and chronic public health threat that cannot be attributed solely to  
812 episodic Harmattan dust events, underscoring the need for sustained emission reduction efforts  
813 across all seasons.

814 3.5 Long-term mean PM<sub>2.5</sub> in Ghana

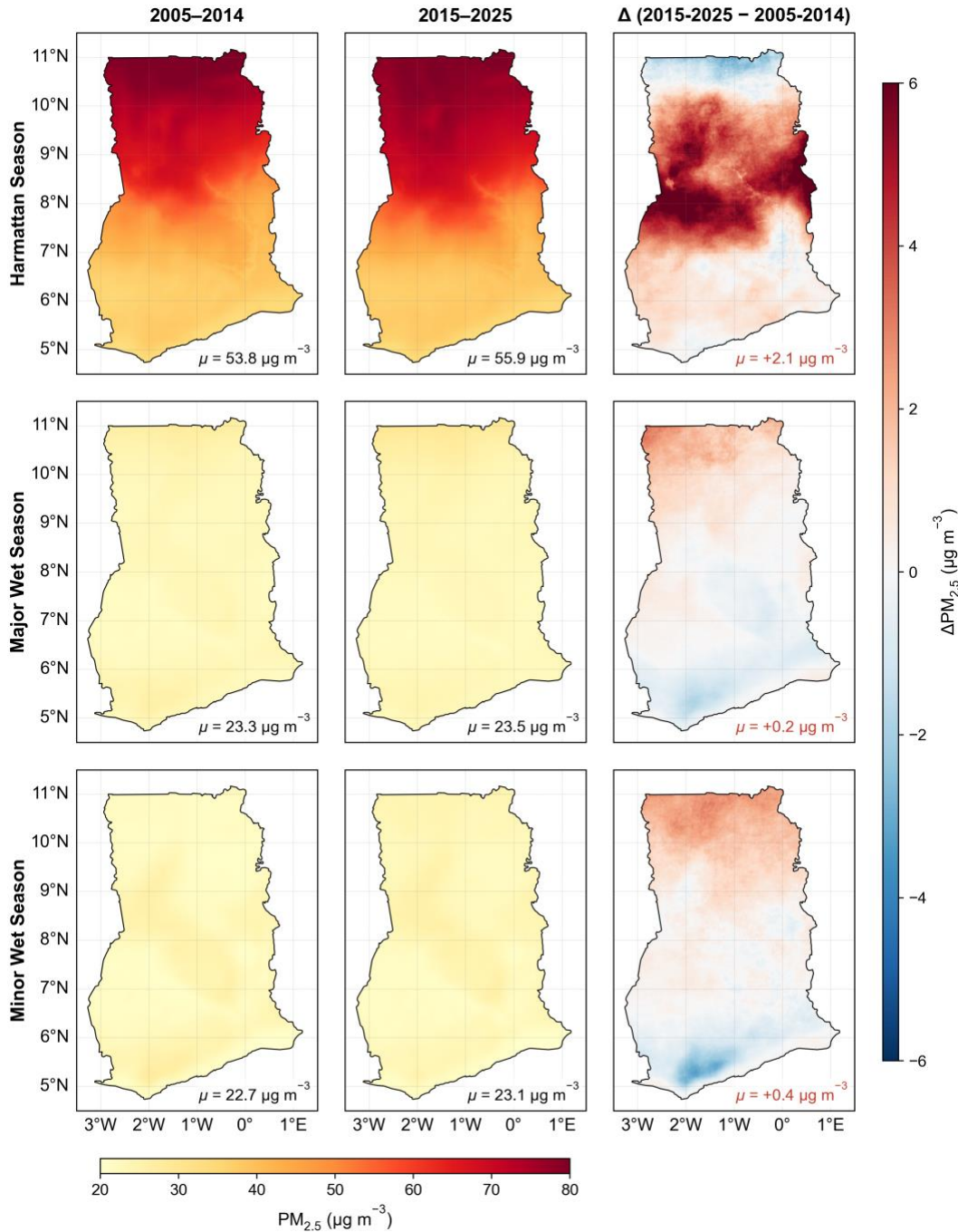


815

816 **Fig. 7. Spatial distribution of long-term means of PM<sub>2.5</sub> concentrations across Ghana.** Maps  
 817 A–D show average PM<sub>2.5</sub> concentrations during 2005–2025 for annual conditions, Harmattan  
 818 season, major wet season, and minor wet season, respectively. Color scales are adjusted separately  
 819 for each row to highlight spatial variability with high (annual and Harmattan) and low PM<sub>2.5</sub>  
 820 concentrations (wet seasons). E shows the mean daily PM<sub>2.5</sub> for regional capitals and additional

821 locations (indicated with \*) in Ghana during Non-Harmattan and Harmattan seasons between  
822 2005–2025. The mean PM<sub>2.5</sub> concentrations (in  $\mu\text{g m}^{-3}$ ) are shown above the error bars.

823 The 21-year mean spatial distribution of PM<sub>2.5</sub> across Ghana (Fig. 7) reveals a pronounced north–  
824 south gradient that is most extreme during the Harmattan season (Fig. 7B) and attenuates  
825 substantially during the wet seasons (Figs 7C–D). The 21-year annual mean (Fig. 7A) shows  
826 consistently elevated concentrations in the Upper East, Upper West, and Northern/Savanna regions  
827 compared to the forested south, reflecting the dominant influence of Harmattan-transported  
828 mineral dust from Saharan and Sahelian regions that disproportionately affects northern Ghana  
829 due to its open savanna landscape and exposure to dry northeasterly winds. During the major wet  
830 season (Fig. 7C), PM<sub>2.5</sub> concentrations are markedly reduced nationally and the north-south  
831 gradient attenuates substantially, driven by enhanced wet deposition and suppressed dust transport,  
832 while southern coastal cities retain discernible PM<sub>2.5</sub> elevations relative to the rural background,  
833 reflecting persistent contributions from traffic, open waste burning, and industrial activity. The  
834 minor wet season (Fig. 7D) shows intermediate concentrations with partial recovery of the north–  
835 south gradient. The city-level seasonal comparison (Fig. 7E) directly quantifies these contrasts:  
836 northern and central cities consistently experience substantially higher Harmattan-season PM<sub>2.5</sub>  
837 than their non-Harmattan levels, while southern coastal cities show comparatively muted  
838 seasonality, consistent with the dominance of year-round anthropogenic rather than dust-driven  
839 sources at those locations.



840

841 **Fig. 8. Multi-year changes in seasonal mean PM<sub>2.5</sub> concentrations across Ghana.** Maps show  
 842 spatial patterns of mean PM<sub>2.5</sub> concentrations for the Harmattan, major wet, and minor wet seasons  
 843 for two multi-year periods (2005–2014 and 2015–2025), along with the corresponding differences  
 844 ( $\Delta = 2015\text{--}2025$  minus  $2005\text{--}2014$ ).  $\mu$  denotes the spatial mean concentration for each panel. The  
 845 color scale for the mean concentration levels is in the bottom, while the diverging color scale on  
 846 the right highlights the magnitude of change.

847 Decadal-scale changes in seasonal PM<sub>2.5</sub> concentrations, comparing the first (2005–2014) and  
848 second (2015–2025) halves of the study period, are shown in Fig. 8. During the Harmattan season,  
849 difference maps reveal a net increase in PM<sub>2.5</sub> across much of northern Ghana in the more recent  
850 decade, consistent with the upward Sen’s slope estimates for Wa, Tamale, Nalerigu, and Damongo  
851 (Table S11) and potentially reflecting intensified dust mobilization from Sahelian source regions  
852 under changing land use and climate conditions. In contrast, wet season PM<sub>2.5</sub> shows modest  
853 decreases in parts of southern Ghana between the two periods, particularly in and around major  
854 urban centers, corroborating the declining trends observed in Fig. 5 and Table S11 for cities such  
855 as Somanya, Ho, and Koforidua. Spatially, the most notable inter-decadal increases are  
856 concentrated along the northern belt, while decreases are evident in the southeastern coastal zone.  
857 Period-by-period comparisons at finer temporal resolution, comparing 2005–2009 with 2010–  
858 2014, 2010–2014 with 2015–2019, and 2015–2019 with 2020–2025, are provided in Figs. S16–  
859 S18. Together, these analyses document a diverging trajectory of PM<sub>2.5</sub> exposure in Ghana over  
860 the 21-year record, with a widening disparity between a dust-burdened north experiencing  
861 increasing concentrations and a south showing modest but measurable improvements.

## 862 4 CONCLUSIONS

863 In this study, we developed three complementary XGBoost models to achieve complete  
864 spatiotemporal coverage, with OMI-based (2005–May 6, 2018) and TROPOMI-based (May 7,  
865 2018 to 2025) models as the primary sources of PM<sub>2.5</sub> estimates, while a MERRA-2-based  
866 XGBoost model fills gaps observed in TROPOMI- and OMI-based models arising from  
867 unavailability of satellite-derived predictors due to low-quality retrievals. Spatial cross-validation  
868 performance ( $r \approx 0.73$ – $0.76$ ) indicates that the XGBoost models capture dominant spatial patterns  
869 across Ghana, though prediction uncertainty may be higher in areas far from monitoring sites and  
870 have distinct emission environment, particularly in the sparsely monitored northern and central  
871 regions (Fig. S13). SHAP analysis revealed that meteorological features (primarily relative  
872 humidity), seasonal cyclic encoding, and aerosol optical indicators (UVAI and AOD) are the  
873 dominant predictors of PM<sub>2.5</sub> in Ghana, consistent with its PM<sub>2.5</sub> climatology being strongly  
874 influenced by Harmattan dust transport and wet-season aerosol scavenging.

875 The 21-year PM<sub>2.5</sub> record reveals a persistent north–south exposure disparity, with Northern Ghana  
876 consistently experiencing substantially higher PM<sub>2.5</sub> concentrations than the south, where mean  
877 exposure levels are typically 1.6–1.8 times greater during peak Harmattan months. This disparity  
878 is further reinforced by contrasting long-term trends. Several northern cities, including Wa,  
879 Tamale, and Nalerigu, exhibit statistically significant increases in PM<sub>2.5</sub> ( $0.05$ – $0.11 \mu\text{g m}^{-3} \text{ yr}^{-1}$ ),  
880 while multiple southern urban centers, such as Somanya, Ho, and Koforidua, show measurable  
881 declines ( $-0.025$  to  $-0.090 \mu\text{g m}^{-3} \text{ yr}^{-1}$ ). These opposing trajectories suggest that national averages  
882 obtained using measurements from a few point locations obscure important regional dynamics,  
883 with air quality improving modestly in parts of the south while deteriorating in the north.

884 These findings carry direct implications for public-health and air-quality policy in Ghana. WHO  
885 24-hour guideline suggests that the mean daily PM<sub>2.5</sub> concentrations should not exceed  $15 \mu\text{g m}^{-3}$   
886 for more than 3–4 days in a year. With mean observed concentrations of  $31.8 \mu\text{g m}^{-3}$ , and more  
887 than 99% of days during the 21-year period consistently exceeding the WHO guideline, ambient  
888 PM<sub>2.5</sub> in Ghana constitutes a pervasive and chronic exposure burden that extends well beyond the  
889 Harmattan season. Even during non-Harmattan months, major urban centers such as Accra,

890 Kumasi, and Tema exceed the higher PM<sub>2.5</sub> threshold from Ghana EPA 24-hour guideline (35 µg  
891 m<sup>-3</sup>) on more than half of days during 2005–2025, indicating that combustion-driven sources  
892 contribute substantially to year-round high PM<sub>2.5</sub> exposure in these urban areas. The statistically  
893 significant declines detected in several southern urban centers are broadly consistent with phased  
894 improvements in vehicle fuel standards, evolving industrial emission controls, and incremental  
895 adoption of cleaner household energy, demonstrating that targeted policy levers can yield  
896 improvements in local air quality. However, these observed changes remain modest, suggesting  
897 the need for more effective policy implementation. In contrast, rising concentrations across parts  
898 of northern Ghana, where long-range mineral dust interacts with expanding biomass burning,  
899 agricultural activity, and urbanization, highlight the limits of local-only mitigation when  
900 transboundary and regional drivers dominate, and underscore the need for coordinated West  
901 African strategies and adaptive measures to reduce dry-season exposure.

902 Although the multi-model framework provides the most spatially and temporally complete high-  
903 resolution daily PM<sub>2.5</sub> dataset currently available for Ghana, characterization of diel variability is  
904 constrained by the scarcity of sub-daily observations needed to better constrain emission sources.  
905 Current satellite retrievals over Africa rely primarily on polar-orbiting instruments with once-daily  
906 overpasses. The absence of geostationary atmospheric composition satellites over the African  
907 continent, such as TEMPO, Sentinel-4, and GEMS in North America, Europe, and East Asia limits  
908 the ability to capture short-term pollution dynamics and separate anthropogenic signals from  
909 meteorological variability.

910 Translating this dataset into public-health and environmental-management outcomes will require  
911 several further steps. Linking the 1 km × 1 km daily PM<sub>2.5</sub> fields with Ghanaian health surveillance,  
912 hospitalization, and demographic and health survey data can quantify the mortality and morbidity  
913 burden attributable to long- and short-term exposures to PM<sub>2.5</sub>, and support policy evaluation for  
914 more effective implementation and monitoring. Extending the modeling framework from Ghana  
915 to other African countries, where calibrated low-cost sensor networks are increasingly available,  
916 can be extended to generate a continental-scale high-resolution PM<sub>2.5</sub> record that helps close the  
917 data gaps currently constraining African representation in global air-quality and climate–health  
918 assessments. Together, these steps would transform the retrospective dataset presented here into a  
919 sustained decision-support resource for air-quality governance across data-sparse regions.

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923 **CRedit authorship contribution statement**

924 Conceptualization: AA, DMW. Investigation: AA, DMW. Methodology: AA, JAA, DMW.  
925 Software: DMW. Formal analysis: AA. Data curation: AA, DMW. Validation: AA, DMW.  
926 Visualization: AA. Supervision: AA, DMW. Project administration: AA, DMW. Funding  
927 acquisition: DMW. Writing—original draft: AA. Writing—review & editing: AA, JAA, SA, ENT,  
928 NABK, CA, BE, MKA, DMW.

929 **Declaration of competing interest**

930 The authors declare that they have no known competing financial interests or personal  
931 relationships that could have appeared to influence the work reported in this paper.

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934 **Data and materials availability**

935 The daily PM<sub>2.5</sub> gridded dataset generated from this study is publicly available on the Zenodo  
936 repository at: <https://zenodo.org/records/19636051>. Additional data produced and used during  
937 analysis can be found in the main text and supporting materials.

938

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1129 <https://doi.org/10.1029/2022MS003099>

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1132 **Supplementary Materials**

1133           This PDF file includes:

1134           Supplementary Text S1 to S2

1135           Figures S1 to S18

1136           Tables S1 to S11

## Supplementary Materials

### **Two decades of kilometer-scale daily PM<sub>2.5</sub> from satellite observations and machine learning reveal geographically diverging exposure in Ghana**

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### **Summary of the Supplementary Materials**

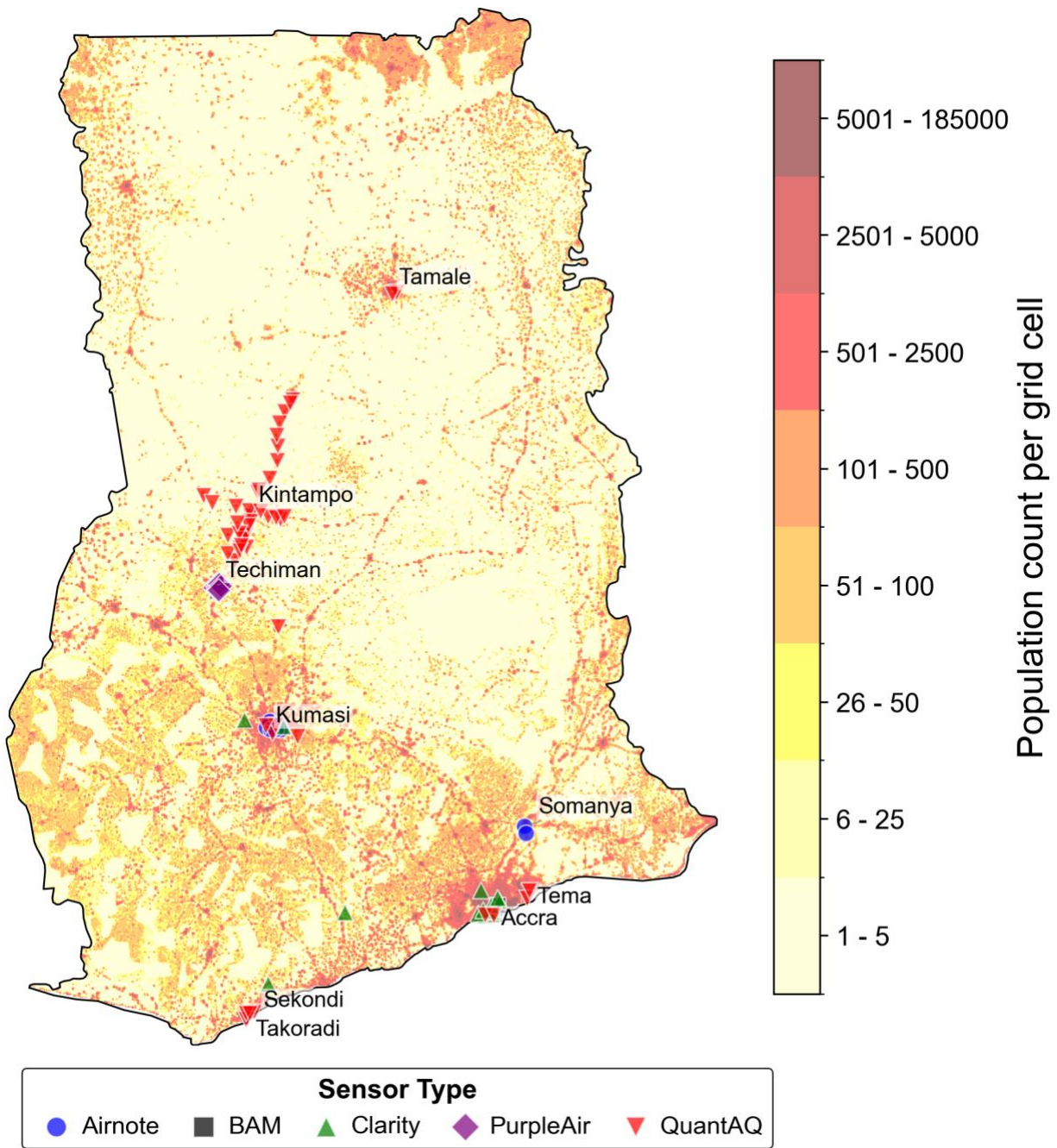
Number of pages: 34

Figures: S1 – S18

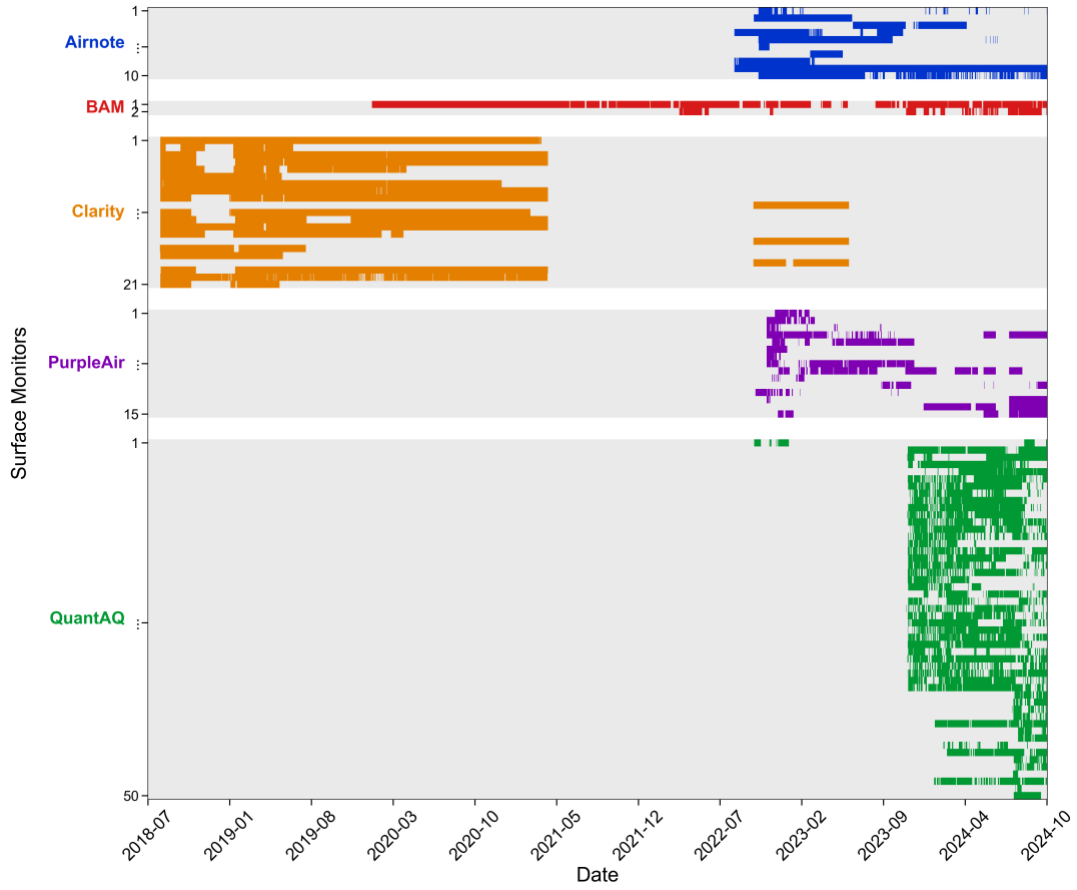
Tables: S1 – S11

**Table S1.** Technical specifications of particulate monitors deployed in Ghana.

<b>Sensor type</b>	<b>Relevant measurement variables</b>	<b>Measurement frequency</b>	<b>Operating PM<sub>2.5</sub> range (µg m<sup>-3</sup>)</b>
Airnote v2.0	PM <sub>2.5</sub> , T, RH	1 min	0–1000
BAM	PM <sub>2.5</sub> , T, RH	1 hr	0–10,000
Clarity Node-S (Generation II)	PM <sub>2.5</sub> , T, RH	1 min	0–1000
PA-II SD	PM <sub>2.5</sub> , T, RH	2 min	0–1000
MODULAIR-PM	PM <sub>2.5</sub> , T, RH	1 min	0–2000



**Fig. S1.** Map of sensor locations. A map of Ghana showing the locations and counts of all surface PM<sub>2.5</sub> monitors and the population density for 2024 from the LandScan Program at Oak Ridge National Lab (Lebakula et al., 2025). The darker regions indicate higher population density.



**Fig. S2.** Availability of PM<sub>2.5</sub> measurements from 98 surface monitors used as ground truth for building the GRASP model. Y-axis represents different sensor types and their corresponding numbers.

**Table S2.** Summary of sensor locations, location types and corresponding counts in Ghana.

<b>Location</b>	<b>Location type</b>	<b>Sensor count</b>
Accra	Urban	20
Kintampo	Rural	35
Kumasi	Urban	13
Nyamibechire	Peri-urban	1
Sekondi	Urban	1
Somanya	Peri-urban	3
Supomu Dunkwa	Peri-urban	1
Takoradi	Urban	3
Tamale	Urban	3
Techiman	Peri-urban	16
Tema	Urban	2

**Table S3.** Summary of total and location-aggregated counts for each sensor types used in Ghana.

<b>Sensor types</b>	<b>Count</b>	<b>Locations</b>
Airnote	10	Kumasi (7) Somanya (3)
BAM	2	Accra (1) Techiman (1)
Clarity	21	Accra (17) Kumasi (2) Nyamibechire (1) Supomu Dunkwa (1)
PurpleAir	15	Techiman (15)
QuantAQ	50	Accra (2) Kintampo (35) Kumasi (4) Sekondi (1) Takoradi (3) Tamale (3) Tema (2)

## Text S1. Calibration of PM<sub>2.5</sub> measurements from low-cost sensors

Each of the low-cost sensors (Airnote, Clarity, PurpleAir and QuantAQ) in the surface monitoring network were calibrated collocating either with a BAM or other BAM-corrected low-cost monitors in the network. The corrected PM<sub>2.5</sub> concentrations were calculated as follow.

$$PM_{2.5, corrected} = f(PM_{2.5, raw}, T, RH) \dots (S1)$$

where PM<sub>2.5, raw</sub>, T and RH denote raw measurements of PM<sub>2.5</sub>, temperature (in °C) and relative humidity (in %) collected by a low-cost sensor. PM<sub>2.5, corrected</sub> represents the quality-assured/quality-controlled (QA/QC) PM<sub>2.5</sub> concentrations retrieved from calibration algorithms.

### *Airnote*

Airnote sensors were mainly deployed in Kumasi (urban) and Somanya (peri-urban). The raw PM<sub>2.5</sub> concentrations, temperature and relative humidity measured by Airnote sensors were used to develop a multiple linear regression (MLR) model to estimate corrected PM<sub>2.5</sub> levels (Owusu-Tawiah et al., 2025).

$$PM_{2.5, corrected} = -6.6 + 0.57*PM_{2.5, raw} + 0.28*T + 0.03*RH \dots (S2)$$

### *Clarity*

Majority of Clarity sensors are located in Accra and a few in Kumasi, Supomu Dunkwa and Nyamibechire. A MLR algorithm was developed using BAM located at Accra US Embassy as reference monitor (Raheja et al., 2023).

$$PM_{2.5, corrected} = 54.60 + 0.4*PM_{2.5, raw} - 0.76*T - 0.35*RH \dots (S3)$$

### *PurpleAir*

All PA-II SD sensors were deployed in Techiman. Each PurpleAir sensor consists of two identical Laser Particle Counters (PMS-5003, PlanTower, Beijing, China) to measure PM<sub>2.5</sub>, named channel A and B. A BME280 sensor (Bosch, Gerlingen, Germany) measures pressure, temperature and humidity sensor which can be used for calibrating the raw PM<sub>2.5</sub> measurements. We applied three filtering criteria to include only valid data points in the analysis: (1) a PM<sub>2.5</sub> threshold for both channels to 1000 µg m<sup>3</sup> and discarded datapoints with PM<sub>2.5</sub> higher than the threshold to comply with the manufacturer recommendation, (2) keep only rows with valid entry in temperature, humidity and PM<sub>2.5</sub> concentrations from the two channels, and (3) retains rows where relative percentage difference in PM<sub>2.5</sub>, defined as  $2*100*|PM_{2.5,A} - PM_{2.5,B}| / (PM_{2.5,A} + PM_{2.5,B})$ , is  $\leq 25\%$ , which neglects datapoints with large differences in PM<sub>2.5</sub> readings from the two channels mostly attributed to ageing of either or both laser particle counters or their clogging due to accumulation of dust or insects.

The PM<sub>2.5</sub> measurements from Techiman BAM was used as ground-truth for calibrating PurpleAir-based PM<sub>2.5</sub> concentrations. A PurpleAir sensor within the Techiman network located at ~500m to the BAM was selected for model development due to its closest proximity to the BAM. An eXtreme Gradient Boosting (XGB) model was developed to derive corrected PM<sub>2.5</sub>.

$$\text{PM}_{2.5, \text{XGB-corrected}} = f(\text{Mean PM}_{2.5, \text{raw}}, T, \text{RH}) \dots(\text{S4})$$

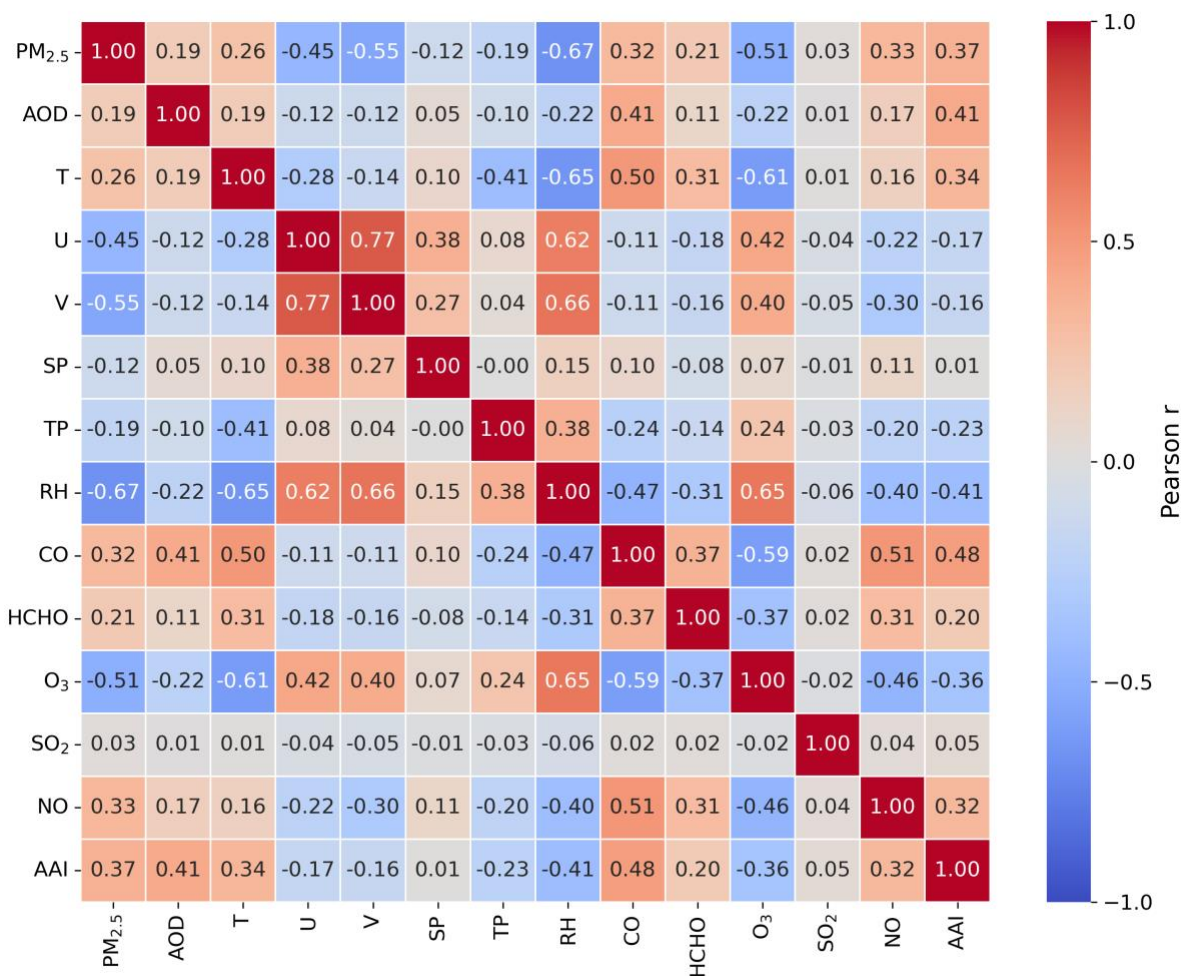
where,  $\text{Mean PM}_{2.5, \text{raw}}$ , is the average of raw  $\text{PM}_{2.5}$  measurements from channels A and B.  $\text{PM}_{2.5, \text{XGB-corrected}}$  represents model corrected  $\text{PM}_{2.5}$ . This model is further used to corrected  $\text{PM}_{2.5}$  outputs from the rest of the PurpleAir sensors in the Techiman network.

### ***QuantAQ***

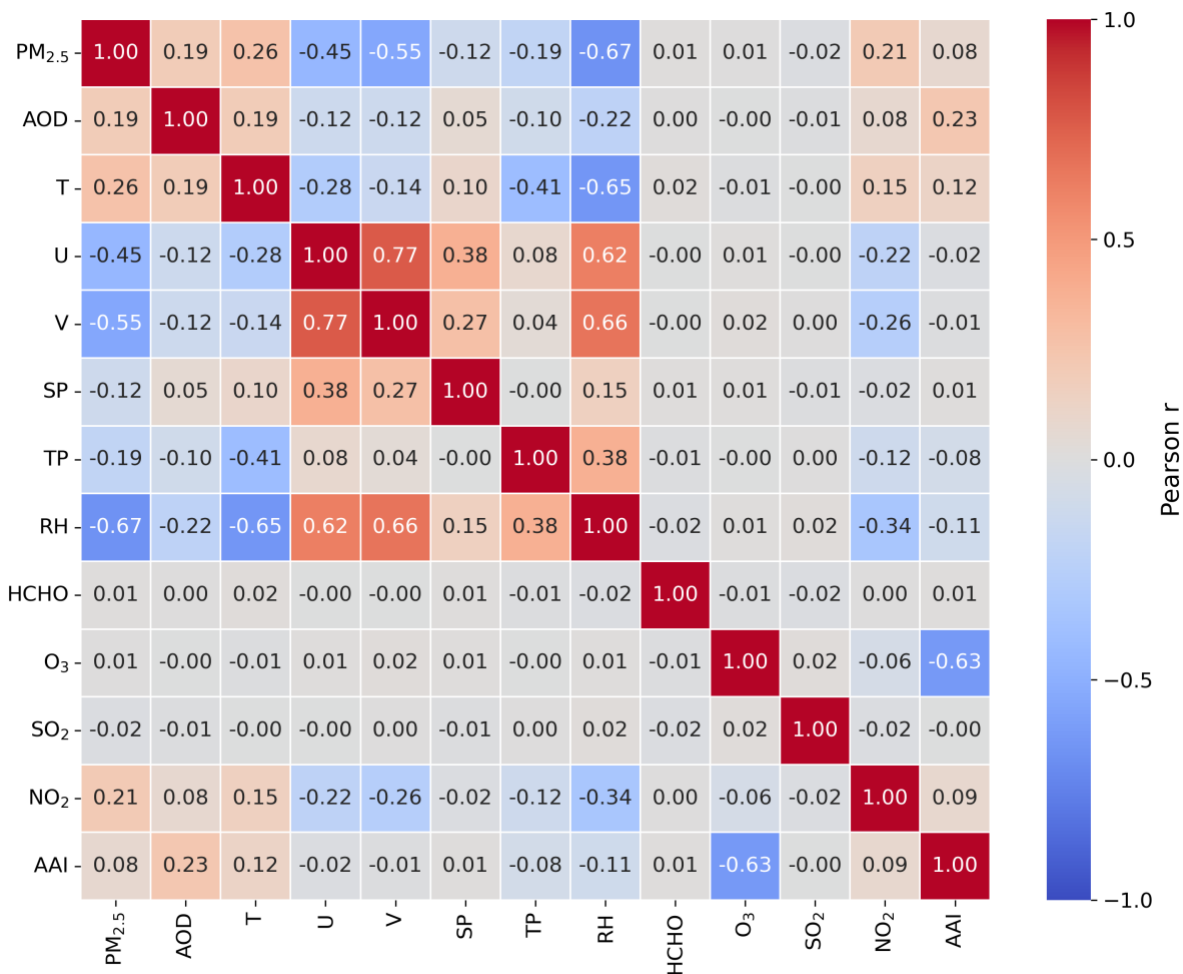
MODULAIR-PM reports manufacturer-built correction applied  $\text{PM}_{2.5}$  that accounts for particle density, aspiration efficiency, and hygroscopic growth. These measurements have shown strong correlation to reference measurements and perform comparable to regression and machine learning algorithms developed for operating in specifically for deployed environment (Raheja et al., 2023). Therefore, the  $\text{PM}_{2.5}$  measurements reported by the MODULAIR monitors in the network were directly used for model development.

**Table S4.** Quality screening of satellite-derived products. A summary of satellite-derived products and corresponding QA criteria to generate high-quality retrievals.

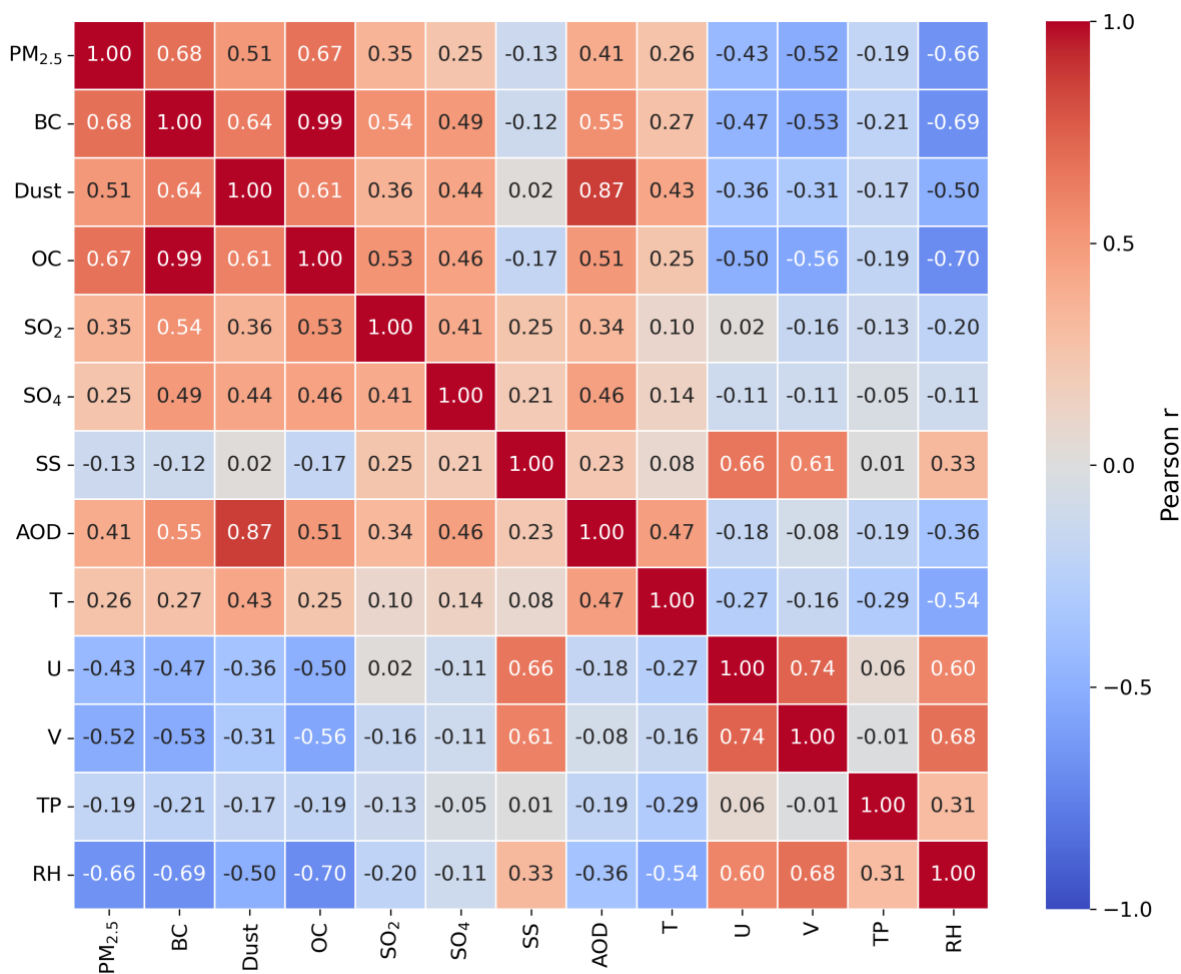
Parameters	Satellite product ( <i>ID</i> )	QA layers and criteria
AOD at 550 nm (MODIS MAIAC)	AOD ( <i>MCD19A2</i> )	$AOD\_QA \in \{0000, 0011, 1011\}$ ( <i>AOD_QA</i> is a 16-bit unsigned integer.)
OMI (Level 2, Collection 3)	UVAI ( <i>OMAERUV v003</i> )	$CloudFraction \leq 0.3$ $SolarZenithAngle \leq 75^\circ$ $XTrackQualityFlags = 0$
	HCHO ( <i>OMHCHO v003</i> )	$AMFCloudFraction \leq 0.3$ $SolarZenithAngle \leq 75^\circ$ $XTrackQualityFlags = 0$ $MainDataQualityFlag = 0$
	O <sub>3</sub> ( <i>OMTO3 v003</i> )	$RadiativeCloudFraction \leq 0.3$ $SolarZenithAngle \leq 85^\circ$ $QualityFlags \leq 1$
	NO <sub>2</sub> ( <i>OMNO2 v003</i> )	$CloudFraction \leq 0.3$ $SolarZenithAngle \leq 75^\circ$ $VcdQualityFlags = 0$
	SO <sub>2</sub> ( <i>OMSO2 v003</i> )	$CloudRadiationFraction \leq 0.3$ $SolarZenithAngle \leq 65^\circ$ $Flag\_RowAnomaly \neq 1$ $Flag\_SAA \neq 1$ $AlgorithmFlag\_SnowIce \neq 2$
TROPOMI (Level 2, Collection 2)	UVAI ( <i>S5P_L2__AER_AI_HiR v2</i> )	$qa\_value \geq 0.5$
	HCHO ( <i>S5P_L2__HCHO__HiR v2</i> )	
	O <sub>3</sub> ( <i>S5P_L2__O3_TOT_HiR v2</i> )	
	NO <sub>2</sub> ( <i>S5P_L2__NO2__HiR v2</i> )	
	SO <sub>2</sub> ( <i>S5P_L2__SO2__HiR v2</i> )	



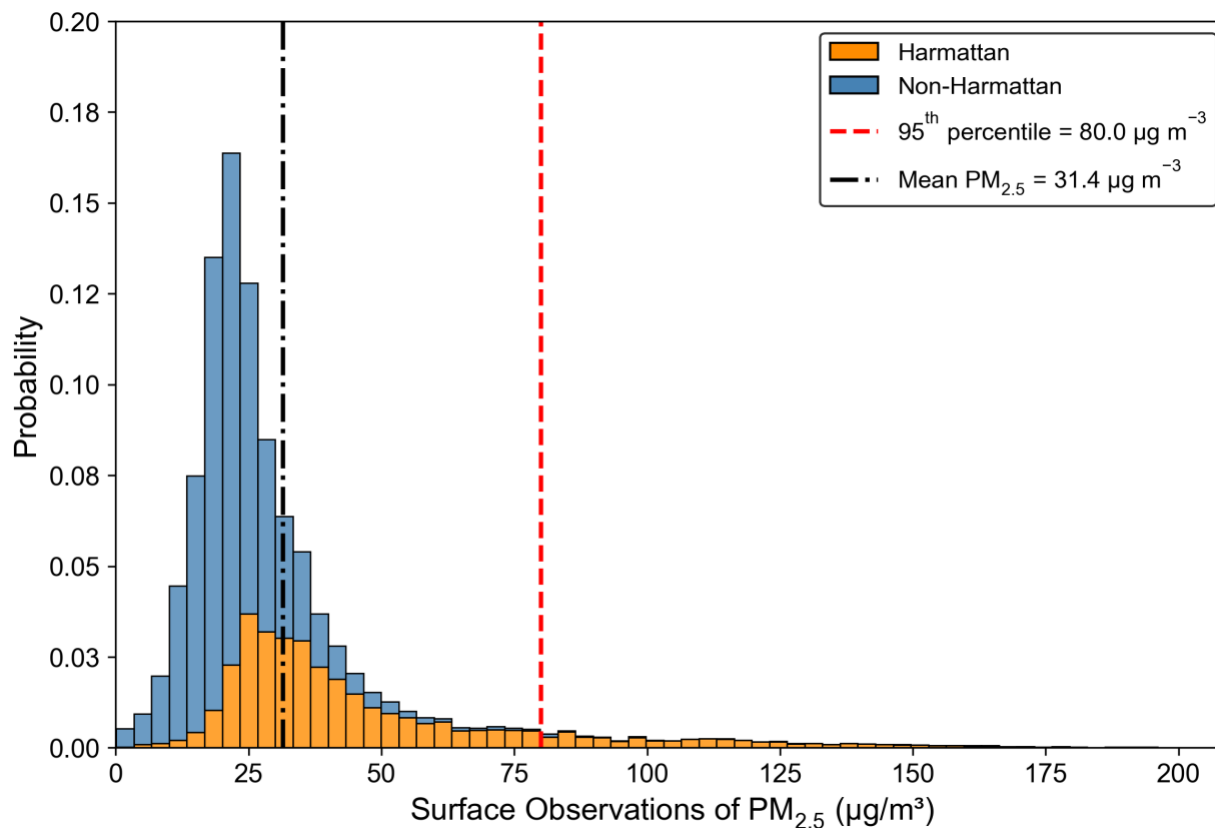
**Fig. S3.** Correlation between training features used in TROPOMI model. Pearson correlation matrix of PM<sub>2.5</sub>, meteorological (ERA5), and satellite-derived (MODIS and TROPOMI) variables used in the modeling framework. Color intensity represents the strength and direction of linear correlations, ranging from -1 (strong negative correlation) to +1 (strong positive correlation).



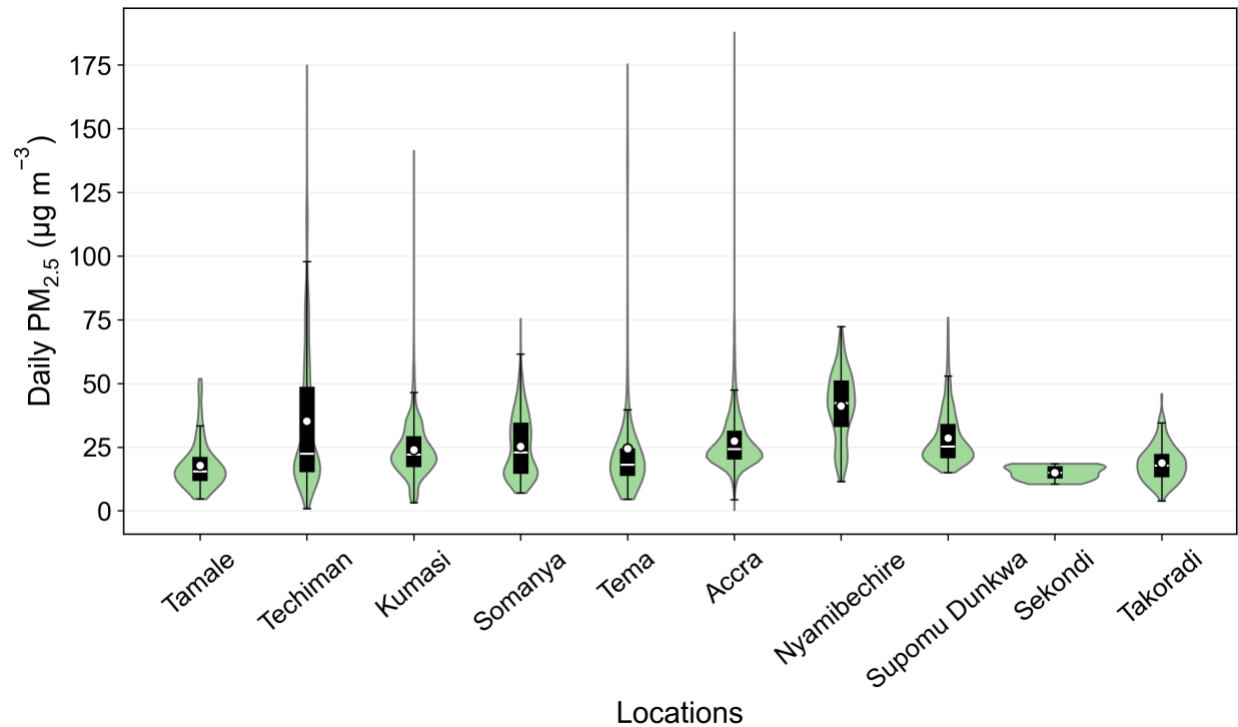
**Fig. S4.** Correlation between training features used in OMI model. Pearson correlation matrix of PM<sub>2.5</sub>, meteorological (ERA5), and satellite-derived (MODIS and OMI) variables used in the modeling framework. Color intensity represents the strength and direction of linear correlations, ranging from -1 (strong negative correlation) to +1 (strong positive correlation).



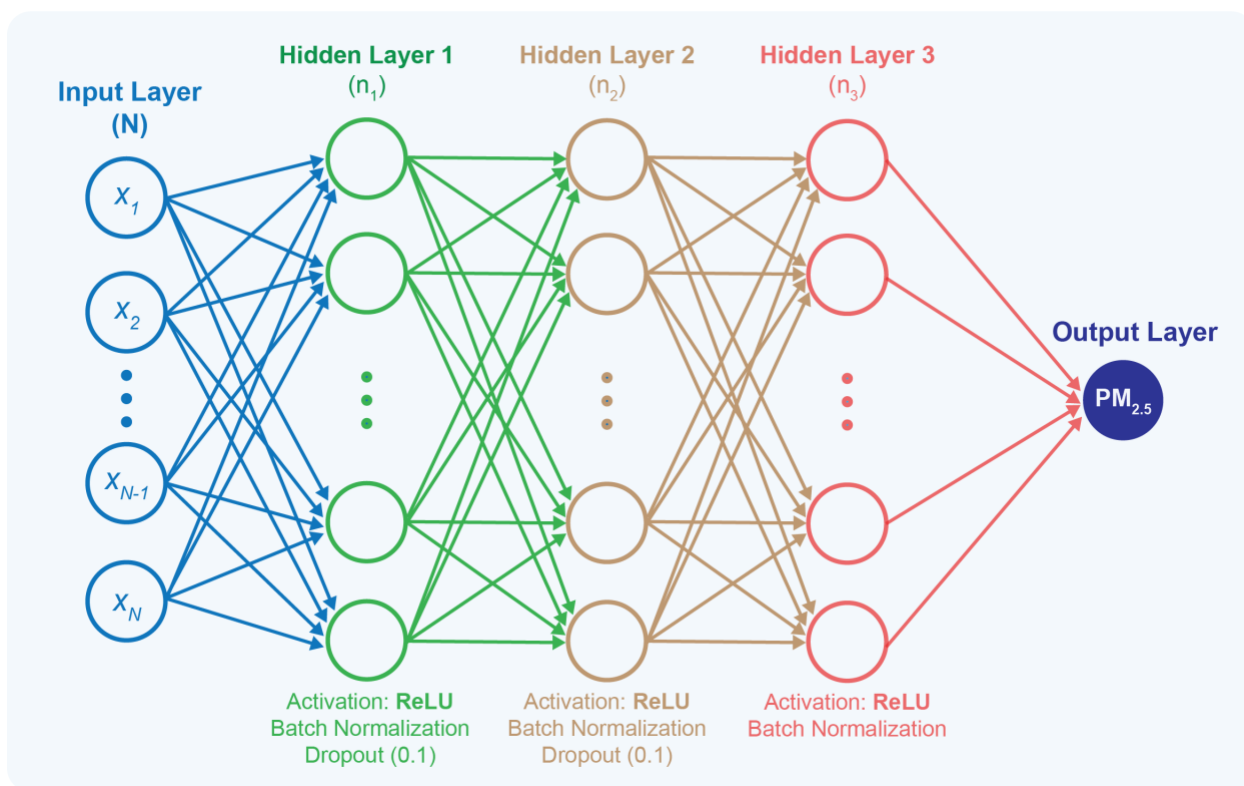
**Fig. S5.** Correlation between training features used in MERRA2-based model. Pearson correlation matrix of PM<sub>2.5</sub>, meteorological (ERA5), and MERRA-2 reanalysis variables used in the modeling framework. Color intensity represents the strength and direction of linear correlations, ranging from -1 (strong negative correlation) to +1 (strong positive correlation).



**Fig. S6.** Surface PM<sub>2.5</sub> measurements from ground monitors. Probability distribution of surface-observed PM<sub>2.5</sub> concentrations ( $\mu\text{g m}^{-3}$ ) stratified into Harmattan (orange) and non-Harmattan (blue) periods. The histogram represents the relative frequency of observations, where the red dashed vertical line indicates the 95<sup>th</sup> percentile ( $80.0 \mu\text{g m}^{-3}$ ) and black dash-dotted line denotes the mean concentration ( $31.4 \mu\text{g m}^{-3}$ ). The distribution is positively skewed, with most observations concentrated at lower concentrations and a long tail toward higher pollution events.



**Fig. S7.** Violin plots of mean daily PM<sub>2.5</sub> concentrations measured by ground-based sensors across all study locations in Ghana. Each violin represents the kernel density of daily PM<sub>2.5</sub> for a location. Embedded boxplots display the median (white horizontal line), mean (white solid circle), and interquartile range (IQR). Whiskers extend to  $1.5 \times \text{IQR}$ , and values beyond the whiskers are considered outliers.



**Fig. S8.** Architecture of the multilayer perceptron (MLP) model used for  $PM_{2.5}$  prediction. The MLP consists of an input layer with  $N$  predictor variables ( $X_1, X_2, \dots, X_N$ ). The input layer is followed by three fully connected hidden layers containing  $n_1, n_2$ , and  $n_3$  nodes, respectively. Each hidden layer applies a ReLU activation function and batch normalization, while dropout (0.1) is applied to the first two hidden layers to reduce overfitting. The output layer contains a single neuron that produces continuous  $PM_{2.5}$  concentrations using a linear activation function. The number of nodes in each hidden layer varies across tested model configurations and is summarized in Tables S5 to S7.

**Table S5.** Optimized hyperparameters for TROPOMI model. Final tuned hyperparameter values for each machine learning model trained for TROPOMI model of the multi-model framework.

<b>Model</b>	<b>Optimized hyperparameters</b>
LASSO	<i>{'regressor__alpha': 0.01}</i>
SVM	<i>{'regressor__C': 100, 'regressor__epsilon': 0.2, 'regressor__gamma': 'scale'}</i>
XGBoost	<i>{'regressor__learning_rate': 0.1, 'regressor__max_depth': 7, 'regressor__n_estimators': 50, 'regressor__subsample': 1.0}</i>
RF	<i>{'regressor__max_depth': 20, 'regressor__min_samples_split': 5, 'regressor__n_estimators': 500}</i>
MLP	<i>{'layers': [input (N=14) → 256 (n<sub>1</sub>) → 128 (n<sub>2</sub>) → 64 (n<sub>3</sub>) → 1], 'activation': ReLU, 'batch_norm': True, 'dropout': 0.1, 'optimizer': AdamW, 'learning_rate': 0.0005, 'weight_decay': 1e-4, 'loss': Huber(delta=10), 'batch_size': 64, 'epochs': 150}</i>

**Table S6.** Optimized hyperparameters for OMI Model. Final tuned hyperparameter values for each machine learning model trained for OMI model of the multi-model framework.

<b>Model</b>	<b>Optimized hyperparameters</b>
LASSO	<i>{'regressor__alpha': 0.01}</i>
SVM	<i>{'regressor__C': 100, 'regressor__epsilon': 0.2, 'regressor__gamma': 0.1}</i>
XGBoost	<i>{'regressor__learning_rate': 0.1, 'regressor__max_depth': 7, 'regressor__n_estimators': 50, 'regressor__subsample': 1.0}</i>
RF	<i>{'regressor__max_depth': 20, 'regressor__min_samples_split': 5, 'regressor__n_estimators': 500}</i>
MLP	<i>{'layers': [input (N=14) → 256 (n<sub>1</sub>) → 128 (n<sub>2</sub>) → 64 (n<sub>3</sub>) → 1], 'activation': ReLU, 'batch_norm': True, 'dropout': 0.1, 'optimizer': AdamW, 'learning_rate': 0.0005, 'weight_decay': 1e-4, 'loss': Huber(delta=10), 'batch_size': 64, 'epochs': 150}</i>

**Table S7.** Optimized hyperparameters for MERRA model. Final tuned hyperparameter values for each machine learning model trained for MERRA model of the multi-model framework.

<b>Model</b>	<b>Optimized hyperparameters</b>
LASSO	<i>{'regressor__alpha': 0.001}</i>
SVM	<i>{'regressor__C': 100, 'regressor__epsilon': 0.2, 'regressor__gamma': 0.1}</i>
XGBoost	<i>{'regressor__learning_rate': 0.1, 'regressor__max_depth': 7, 'regressor__n_estimators': 50, 'regressor__subsample': 1.0}</i>
RF	<i>{'regressor__max_depth': 20, 'regressor__min_samples_split': 5, 'regressor__n_estimators': 500}</i>
MLP	<i>{'layers': [input (N=14) → 256 (n<sub>1</sub>) → 128 (n<sub>2</sub>) → 64 (n<sub>3</sub>) → 1], 'activation': ReLU, 'batch_norm': True, 'dropout': 0.1, 'optimizer': AdamW, 'learning_rate': 0.0005, 'weight_decay': 1e-4, 'loss': Huber(delta=10), 'batch_size': 64, 'epochs': 150}</i>

**Table S8.** Performance metrics for TROPOMI Model. Performance comparison of machine learning models for PM<sub>2.5</sub> prediction under random, temporal, and spatial cross-validation schemes. Model performance is evaluated using coefficient of determination ( $R^2$ ), Pearson correlation ( $r$ ), bias, root mean squared error (RMSE), and mean absolute error (MAE). Normalized RMSE (nRMSE) and normalized MAE (nMAE) are reported in parentheses. Temporal cross-validation evaluates model generalization across years, while spatial cross-validation assesses transferability across locations. RMSE, MAE and bias are expressed in  $\mu\text{g m}^{-3}$ .

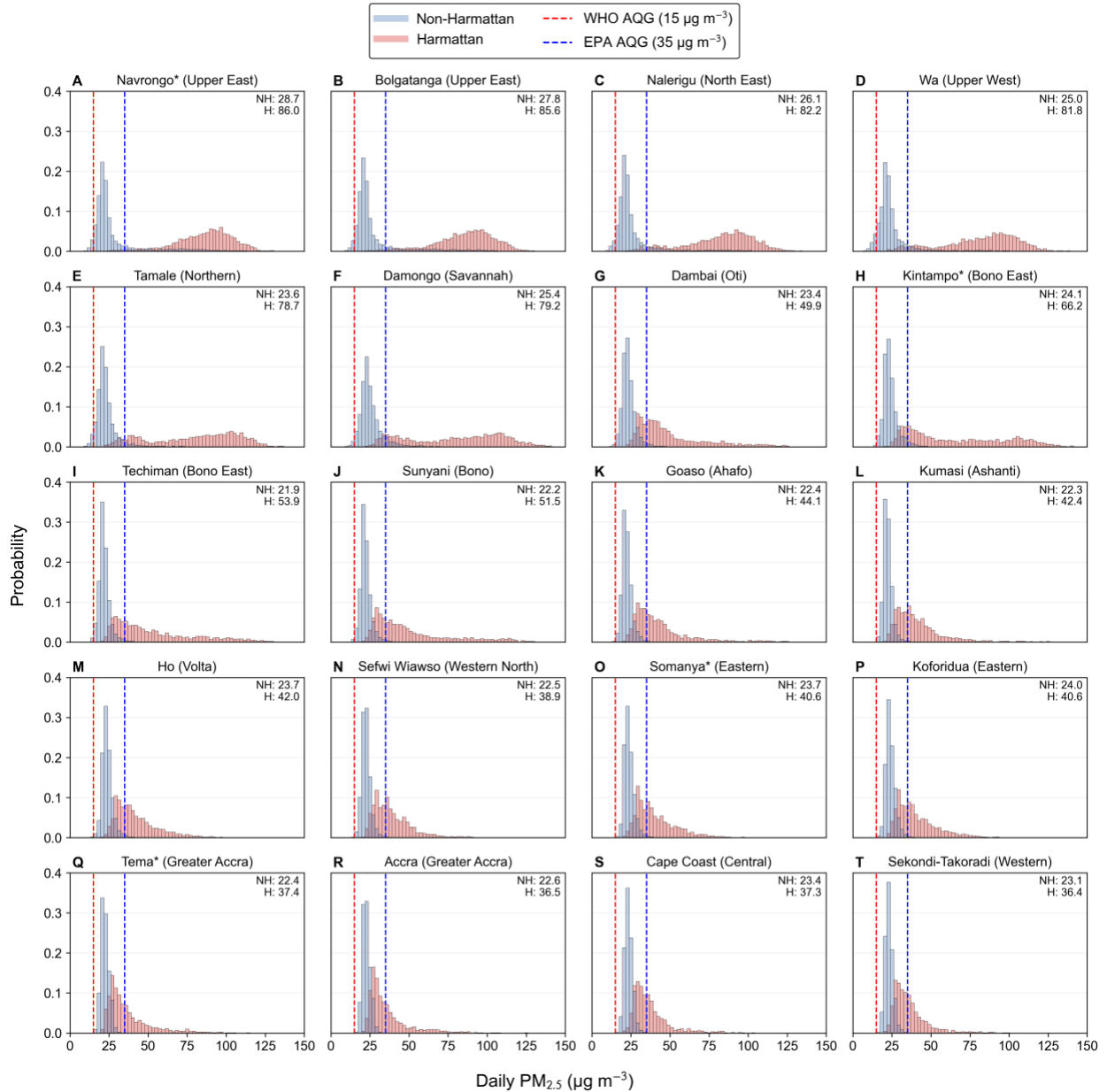
Model	Random CV				Temporal CV				Spatial CV			
	$R^2$ ( $r$ )	Bias	RMSE (nRMSE)	MAE (nMAE)	$R^2$ ( $r$ )	Bias	RMSE (nRMSE)	MAE (nMAE)	$R^2$ ( $r$ )	Bias	RMSE (nRMSE)	MAE (nMAE)
LASSO	0.60 (0.78)	-0.17	15.0 (0.48)	9.7 (0.31)	0.56 (0.75)	0.33	15.7 (0.50)	10.3 (0.33)	0.45 (0.68)	2.96	19.8 (0.57)	13.8 (0.39)
SVM	0.69 (0.83)	-1.55	13.2 (0.42)	7.8 (0.25)	0.55 (0.77)	-0.36	15.8 (0.50)	9.8 (0.31)	0.52 (0.75)	-5.91	19.4 (0.55)	13.3 (0.38)
RF	0.71 (0.85)	0.09	12.7 (0.40)	7.6 (0.24)	0.55 (0.77)	0.77	15.9 (0.50)	10.1 (0.32)	0.56 (0.76)	-1.52	18.4 (0.53)	13.1 (0.37)
<b>XGB*</b>	0.72 (0.86)	-0.01	12.5 (0.40)	7.3 (0.23)	0.57 (0.78)	0.66	15.6 (0.49)	9.6 (0.30)	0.57 (0.76)	-0.93	17.3 (0.49)	10.4 (0.33)
MLP	0.71 (0.84)	-0.12	12.8 (0.40)	7.8 (0.25)	0.54 (0.76)	-1.56	16.0 (0.51)	10.4 (0.33)	0.42 (0.66)	-2.75	18.7 (0.54)	13.5 (0.38)

**Table S9.** Performance metrics for OMI Model. Performance comparison of machine learning models for PM<sub>2.5</sub> prediction under random, temporal, and spatial cross-validation schemes. Model performance is evaluated using coefficient of determination (R<sup>2</sup>), Pearson correlation (r), bias, root mean squared error (RMSE), and mean absolute error (MAE). Normalized RMSE (nRMSE) and normalized MAE (nMAE) are reported in parentheses. Temporal cross-validation evaluates model generalization across years, while spatial cross-validation assesses transferability across locations. RMSE, MAE and bias are expressed in  $\mu\text{g m}^{-3}$ .

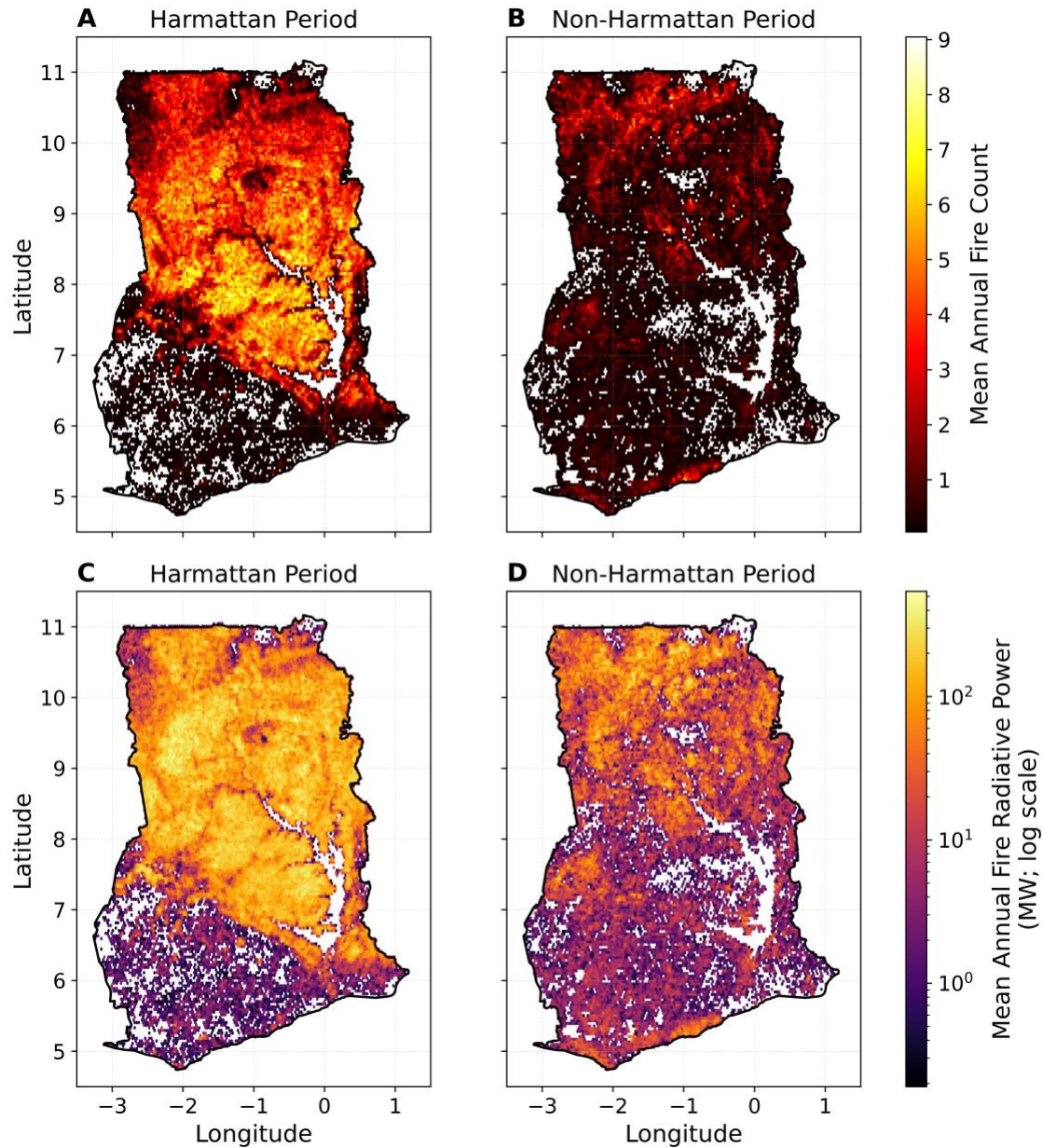
Model	Random CV				Temporal CV				Spatial CV			
	R <sup>2</sup> (r)	Bias	RMSE (nRMSE)	MAE (nMAE)	R <sup>2</sup> (r)	Bias	RMSE (nRMSE)	MAE (nMAE)	R <sup>2</sup> (r)	Bias	RMSE (nRMSE)	MAE (nMAE)
LASSO	0.58 (0.76)	-0.19	15.4 (0.49)	10.2 (0.32)	0.53 (0.73)	0.35	16.3 (0.52)	10.9 (0.35)	0.38 (0.64)	-4.19	21.7 (0.59)	14.1 (0.41)
SVM	0.68 (0.83)	-1.79	13.4 (0.42)	7.8 (0.25)	0.52 (0.75)	-1.31	16.4 (0.52)	10.0 (0.32)	0.48 (0.73)	-5.55	20.2 (0.57)	13.9 (0.40)
RF	0.71 (0.84)	-0.16	13.0 (0.41)	7.9 (0.25)	0.53 (0.75)	1.46	16.3 (0.52)	10.3 (0.33)	0.48 (0.75)	-1.53	19.3 (0.55)	13.6 (0.39)
<b>XGB*</b>	0.72 (0.85)	.04	12.8 (0.41)	7.7 (0.24)	0.56 (0.76)	0.75	15.6 (0.50)	9.6 (0.31)	0.54 (0.75)	-1.01	18.9 (0.54)	13.5 (0.38)
MLP	0.70 (0.83)	-1.19	13.3 (0.42)	8.1 (0.26)	0.50 (0.71)	1.78	16.6 (0.53)	10.6 (0.34)	0.46 (0.70)	-1.83	19.8 (0.57)	15.5 (0.48)

**Table S10.** Performance metrics for MERRA-2 model. Performance comparison of machine learning models for PM<sub>2.5</sub> prediction under random, temporal, and spatial cross-validation schemes. Model performance is evaluated using coefficient of determination (R<sup>2</sup>), Pearson correlation (r), bias, root mean squared error (RMSE), and mean absolute error (MAE). Normalized RMSE (nRMSE) and normalized MAE (nMAE) are reported in parentheses. Temporal cross-validation evaluates model generalization across years, while spatial cross-validation assesses transferability across locations. RMSE, MAE and bias are expressed in  $\mu\text{g m}^{-3}$ .

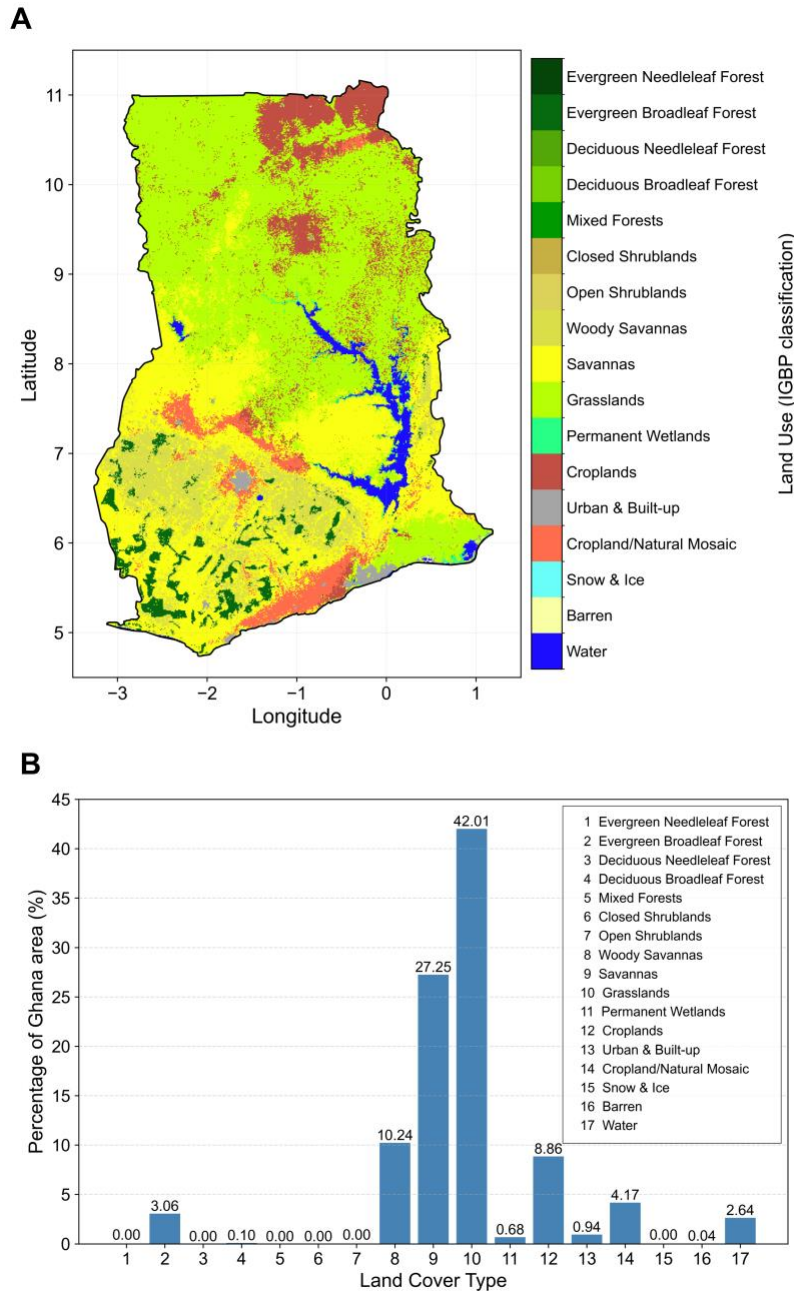
Model	Random CV				Temporal CV				Spatial CV			
	R <sup>2</sup> (r)	Bias	RMSE (nRMSE)	MAE (nMAE)	R <sup>2</sup> (r)	Bias	RMSE (nRMSE)	MAE (nMAE)	R <sup>2</sup> (r)	Bias	RMSE (nRMSE)	MAE (nMAE)
LASSO	0.63 (0.79)	-0.19	14.5 (0.46)	9.1 (0.29)	0.58 (0.75)	-0.06	15.2 (0.48)	9.6 (0.30)	0.41 (0.67)	-4.63	18.2 (0.57)	12.0 (0.38)
SVM	0.70 (0.83)	-1.68	12.7 (0.41)	7.6 (0.25)	0.55 (0.76)	-1.17	15.9 (0.50)	9.6 (0.30)	0.37 (0.66)	5.60	19.0 (0.60)	11.9 (0.37)
RF	0.71 (0.84)	0.38	12.6 (0.40)	7.5 (0.24)	0.51 (0.75)	1.37	16.6 (0.53)	10.4 (0.33)	0.47 (0.70)	2.16	17.2 (0.54)	10.8 (0.34)
<b>XGB*</b>	0.72 (0.85)	-0.18	12.6 (0.40)	7.2 (0.23)	0.59 (0.78)	0.03	15.2 (0.48)	9.2 (0.29)	0.49 (0.73)	0.39	16.8 (0.50)	10.1 (0.32)
MLP	0.70 (0.83)	1.03	12.9 (0.42)	7.8 (0.26)	0.55 (0.74)	1.92	16.0 (0.51)	9.8 (0.31)	0.44 (0.69)	-1.54	18.0 (0.55)	11.2 (0.35)



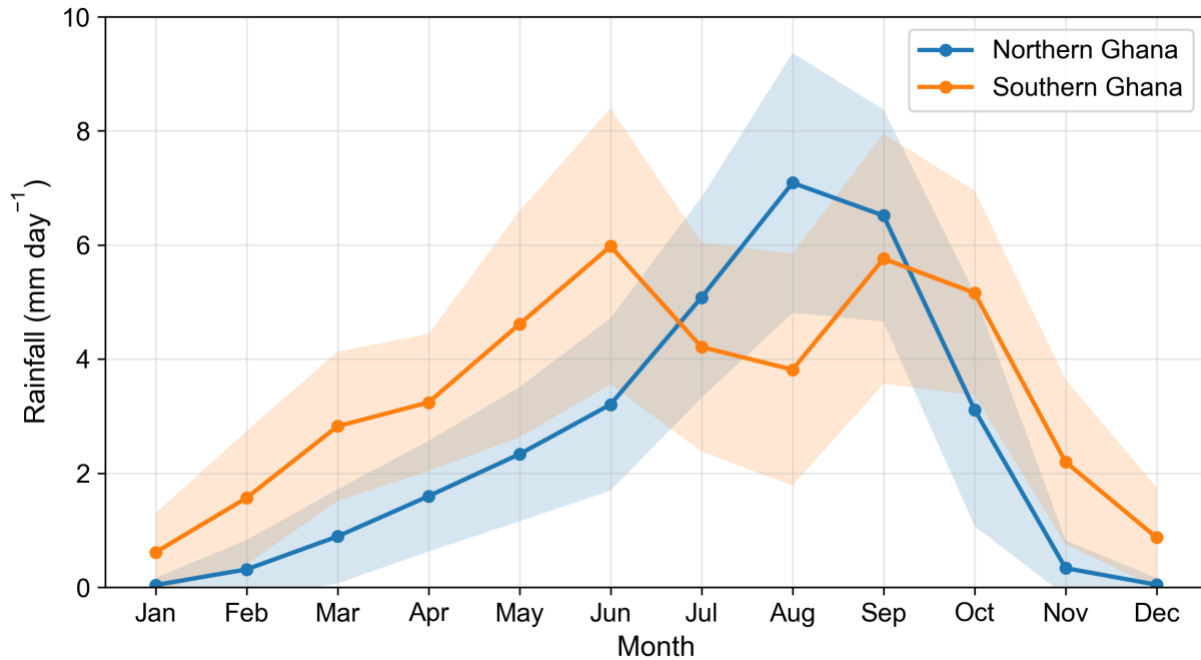
**Fig. S9.** Distribution of  $PM_{2.5}$  concentrations between 2005–2025. (A–T) City-level probability distributions of daily  $PM_{2.5}$  concentrations across Ghana during Harmattan and non-Harmattan periods (2005–2025). Panels show histograms of daily mean  $PM_{2.5}$  concentrations for each regional capital, arranged geographically from north (top-left) to south (bottom-right). Blue bars represent non-Harmattan conditions, while red bars represent Harmattan conditions (mid-November to mid-March). Average  $PM_{2.5}$  (in  $\mu g m^{-3}$ ) for both periods are denoted as NH and H for non-Harmattan and Harmattan periods. The red and blue dashed vertical lines indicate the 24-hour air quality guidelines from WHO ( $15 \mu g m^{-3}$ ) and Ghana EPA ( $35 \mu g m^{-3}$ ), highlighting extremely high number of exceedances, particularly during Harmattan months. Overall, the figure highlights a pronounced north–south gradient in particulate pollution and demonstrates the dominant role of Harmattan dust in shaping seasonal air quality patterns across Ghana.



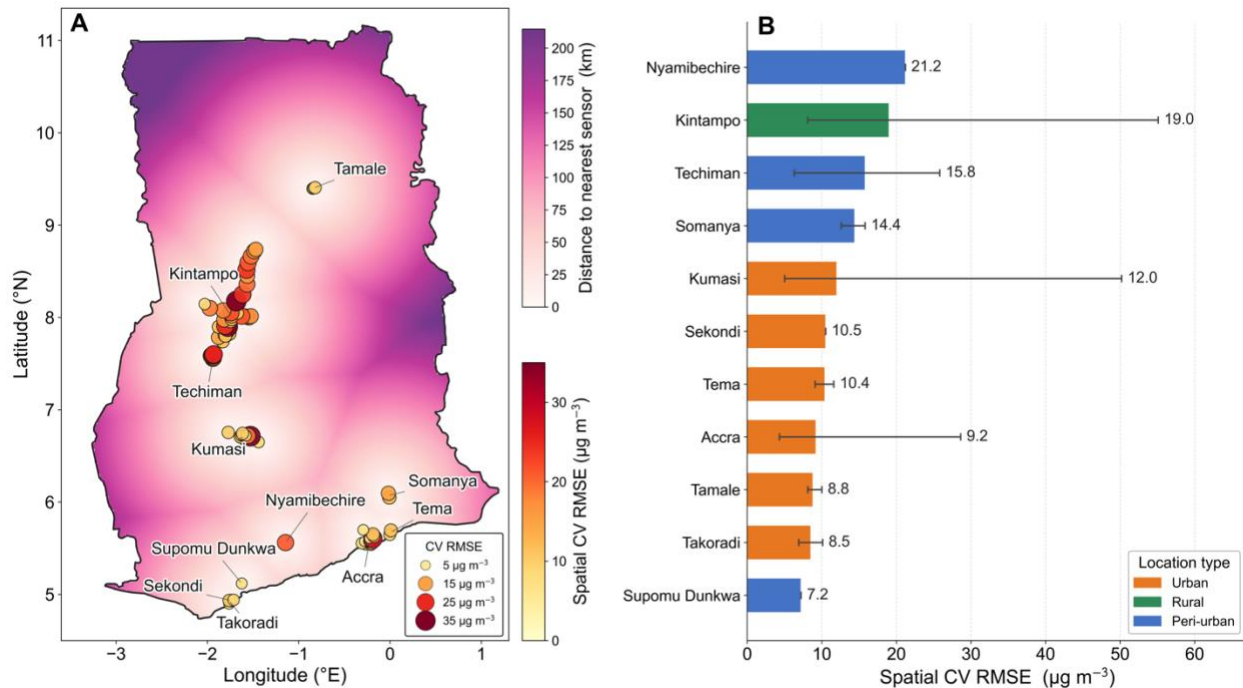
**Fig. S10.** Spatial distribution of fire activity across Ghana during Harmattan and non-Harmattan seasons (2005–2025). (A–B) Fire detection counts derived from MODIS MCD14ML active fire daily observations aggregated over a 21-year period. (C–D) Corresponding cumulative fire radiative power (FRP), indicating the spatial distribution of fire intensity in megawatts. Harmattan-season fires (December–February; A, C) are concentrated across the northern and middle savanna regions of Ghana, reflecting widespread biomass burning associated with seasonal land management practices and dry atmospheric conditions. In contrast, non-Harmattan fires (March–November; B, D) occur less frequently and are more spatially heterogeneous, with localized hotspots observed across both northern and southern regions. Log-transformed cumulative FRP highlights persistently higher fire intensity across northern Ghana relative to southern forested and coastal zones. All maps include only fire detections with confidence  $\geq 30\%$ .



**Fig. S11.** Land cover distribution across Ghana in 2024. (A) Annual spatial distribution of land cover classes derived from the MCD12Q1 Version 6.1 MODIS Land Cover Type product (International Geosphere–Biosphere Programme, IGBP classification). The original 500 m sinusoidal grid product was regridded to 1 km resolution for Ghana. Savannas and grasslands dominate northern and central Ghana, while croplands and cropland–natural vegetation mosaics are prevalent in southern and transitional zones. Water bodies correspond primarily to Lake Volta and major river systems. (B) Fractional area coverage of each IGBP land cover class within Ghana. Grasslands (42.01%), Savannas (27.25%), woody savannas (10.24%), and croplands (8.86%) represent the dominant land cover types, together accounting for more than 85% of national land area. Only 0.94% of Ghana’s land is classified as urban.



**Fig. S12.** Monthly rainfall climatology for Northern and Southern Ghana (2005–2025). Lines show the long-term mean daily rainfall for each calendar month derived from ERA5 data, averaged over 2005–2025. Shaded regions represent  $\pm 1$  standard deviation, indicating interannual variability. Southern Ghana exhibits a bimodal rainfall regime with peaks in May–June (major wet season) and September–October (minor wet season). In contrast, Northern Ghana shows a unimodal rainfall pattern peaking during August–September, a typical characteristic of the West African monsoon.



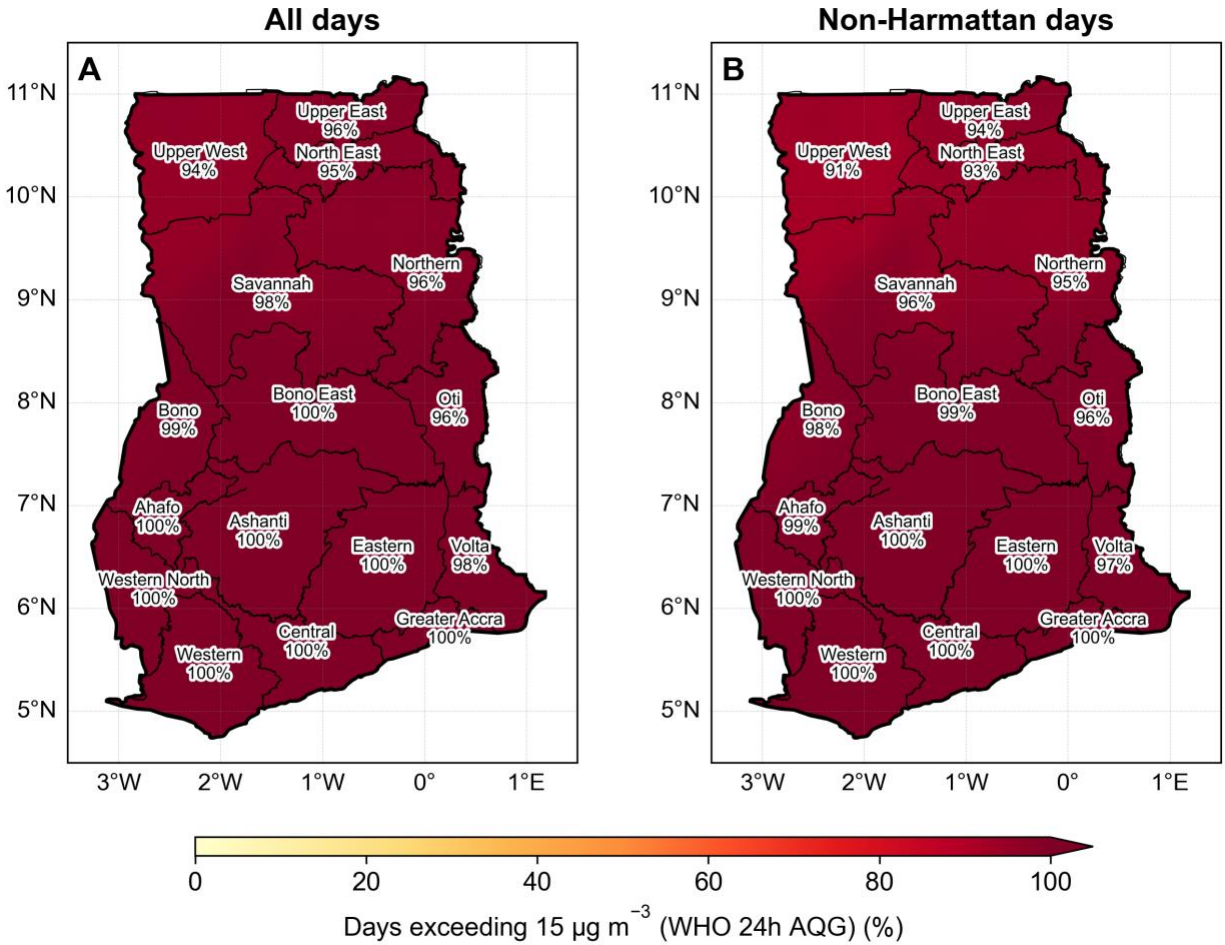
**Fig. S13.** Spatial uncertainty assessment of the  $\text{PM}_{2.5}$  model across Ghana. (A) Background color shows the distance to the nearest ground monitoring site, where darker regions indicate areas farther from sensor locations. Circles represent leave-one-site-out cross-validation (CV) RMSE at individual sensor locations, with circle size and color intensity proportional to RMSE magnitude. (B) Mean spatial CV RMSE by city, with horizontal whiskers indicating the full range across monitoring sites within each location. Bar colors denote site classification (urban, peri-urban, and rural). Overall, most monitoring locations exhibit CV RMSE values below  $20 \mu\text{g m}^{-3}$ .

## **Text S2. Estimation of long-term PM<sub>2.5</sub> Trends**

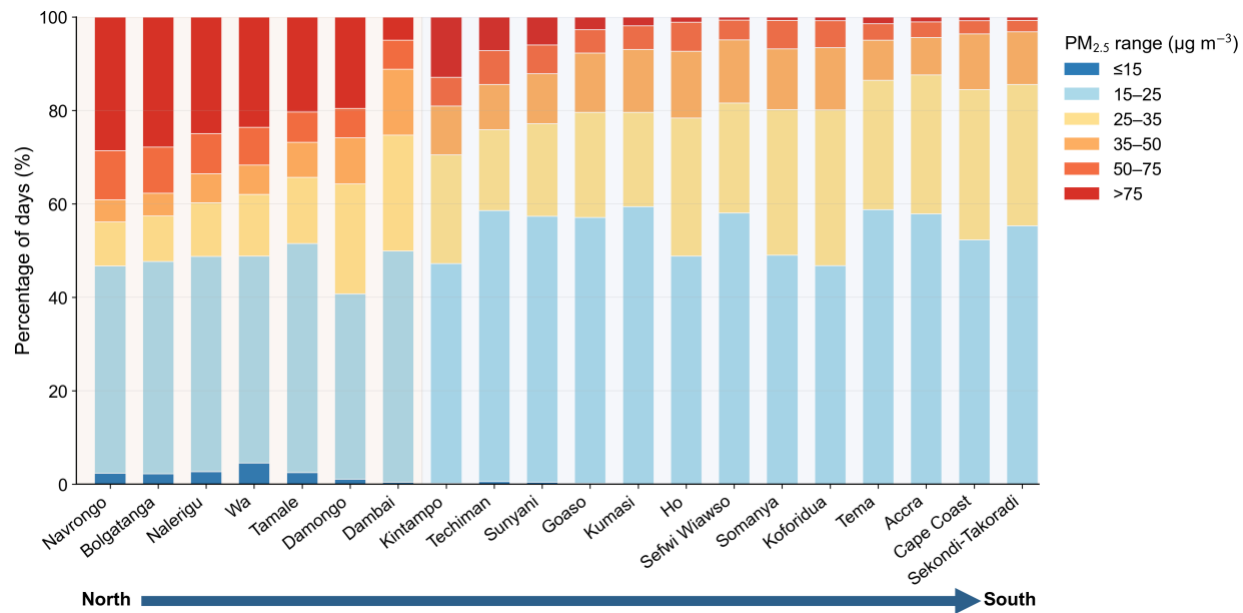
Long-term trends in PM<sub>2.5</sub> concentrations were estimated at each grid cell using a non-parametric trend analysis approach applied to the annual mean PM<sub>2.5</sub> time series for the period 2005–2025. First, annual mean PM<sub>2.5</sub> concentrations were calculated from the gridded daily PM<sub>2.5</sub> dataset for each year. The magnitude of the temporal trend was then quantified using Sen’s slope estimator, which computes the median of all pairwise slopes between observations in the time series and is robust to outliers and non-normal data distributions. Statistical significance of the monotonic trend was evaluated using the Mann–Kendall test, a rank-based method commonly used for environmental time series that does not assume linearity or normality. Grid cells with a Mann–Kendall *p*-value less than 0.05 were considered to exhibit statistically significant trends and are highlighted in the figure using stippling. This approach provides a robust assessment of spatial patterns in long-term changes in PM<sub>2.5</sub> concentrations across Ghana over the study period.

**Table S11.** Annual and seasonal long-term trends in PM<sub>2.5</sub> concentrations across Ghanaian locations. *Slope* is the Sen’s slope estimates ( $\mu\text{g m}^{-3} \text{ yr}^{-1}$ ), representing the magnitude and direction of monotonic trends derived using monthly PM<sub>2.5</sub> data (2005–2025). *p-value* is derived from the non-parametric Mann–Kendall significance test. Trends with p-values < 0.05 are considered statistically significant and are indicated with an asterisk (\*). Non-regional capitals are denoted with a hash (#).

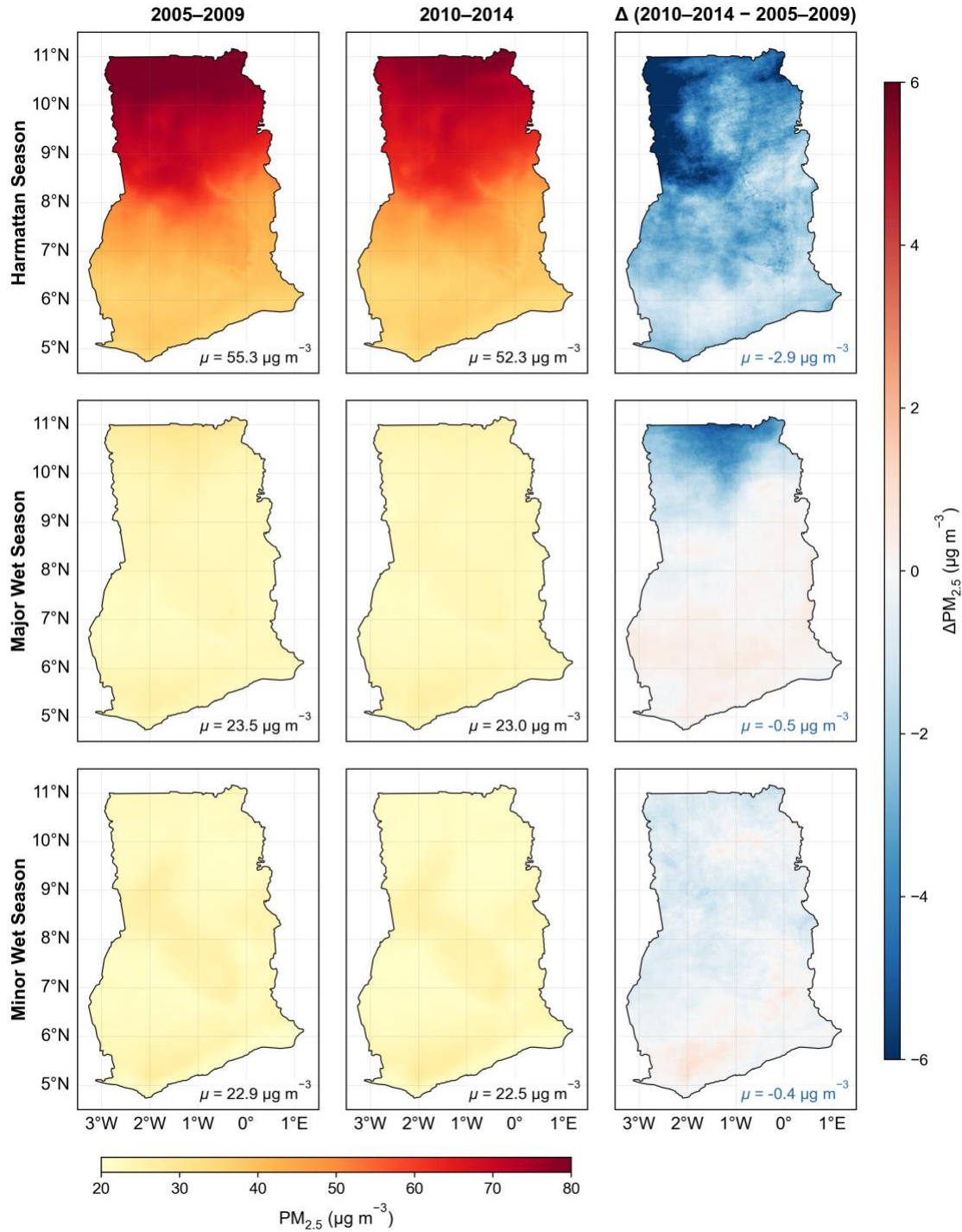
Location	Annual		Harmattan		Non-Harmattan	
	Slope	p-value	Slope	p-value	Slope	p-value
Navrongo <sup>#</sup>	0.003	0.206	-0.057	0.007*	0.011	0*
Bolgatanga	0.001	0.308	-0.051	0.026*	0.007	0.001*
Nalerigu	0.004	0.003*	-0.031	0.474	0.011	0*
Wa	0.008	0*	-0.028	0.903	0.018	0*
Tamale	0.006	0.001*	0.008	0.395	0.01	0*
Damongo	0.004	0.002*	0.037	0.061	0.006	0.013*
Dambai	0.001	0.469	0.016	0.262	0.001	1
Kintampo <sup>#</sup>	0.002	0.127	0.038	0.173	0.003	0.405
Techiman	0.003	0.001*	0.05	0.039*	0.004	0.008*
Sunyani	0.002	0.002*	0.033	0.05*	0.003	0.021*
Goaso	0	0.166	0.017	0.19	0.001	0.486
Kumasi	-0.002	0.253	-0.008	0.925	-0.003	0.16
Ho	-0.007	0*	-0.01	0.251	-0.011	0*
Sefwi Wiawso	-0.001	0.841	0.007	0.338	-0.002	0.278
Somanya <sup>#</sup>	-0.009	0*	-0.018	0.047*	-0.014	0*
Koforidua	-0.007	0*	-0.003	0.636	-0.013	0*
Tema <sup>#</sup>	-0.003	0.003*	-0.004	0.24	-0.004	0.004*
Accra	-0.002	0.16	0.004	0.925	-0.003	0.054
Cape Coast	-0.003	0.001*	-0.011	0.077	-0.005	0.004*
Sekondi-Takoradi	-0.003	0.003*	-0.011	0.148	-0.005	0.008*



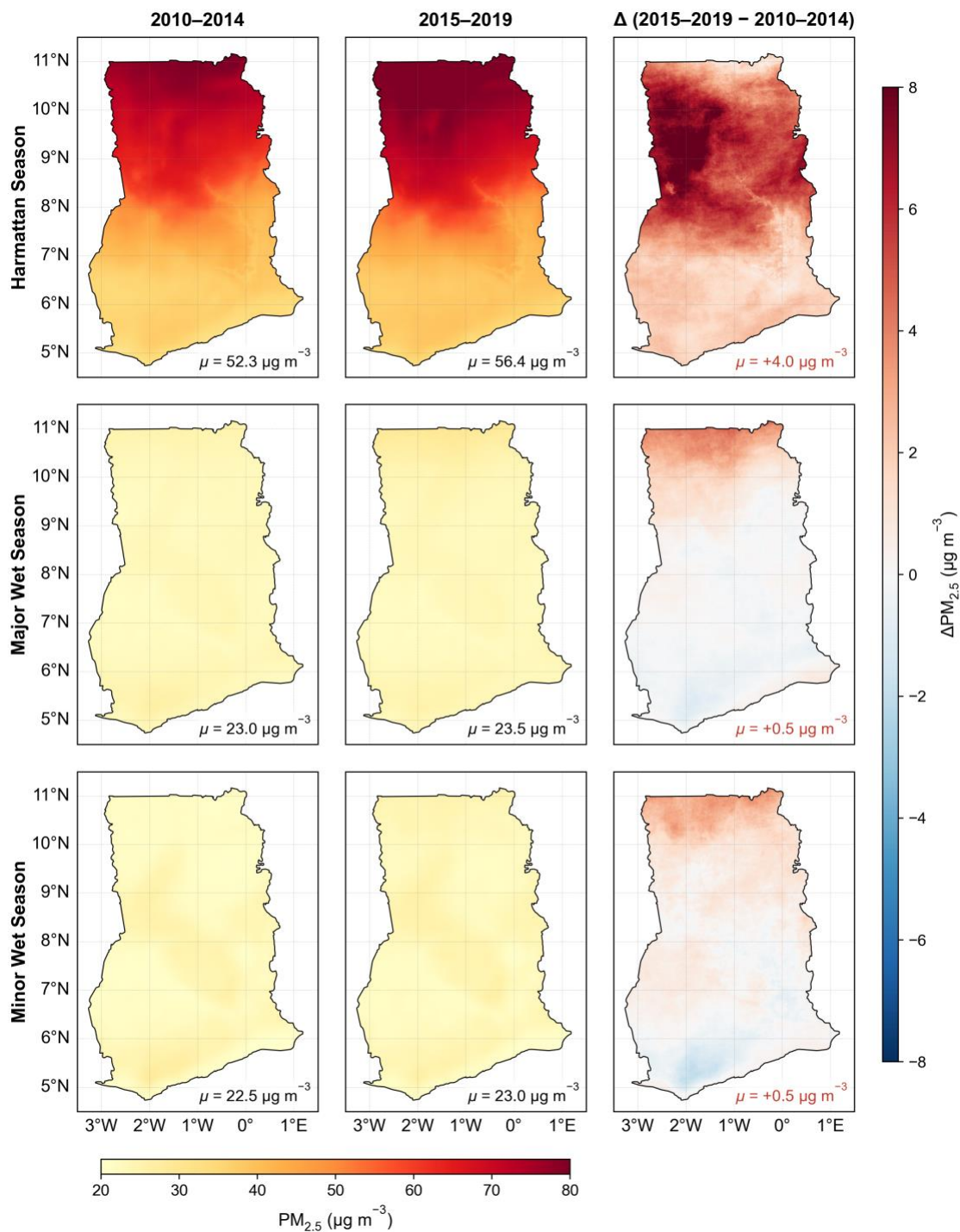
**Fig. S14.** Spatial distribution of the percentage of days exceeding the WHO 24-hour  $\text{PM}_{2.5}$  guideline. Maps show the percentage of days during 2005–2025 in which daily  $\text{PM}_{2.5}$  concentrations exceeded the WHO 24-hour air quality guideline ( $15 \mu\text{g m}^{-3}$ ) across Ghana, calculated using (a) all available days and (b) non-Harmattan days. Values represent the proportion of days (%) at each grid cell exceeding the guideline threshold.



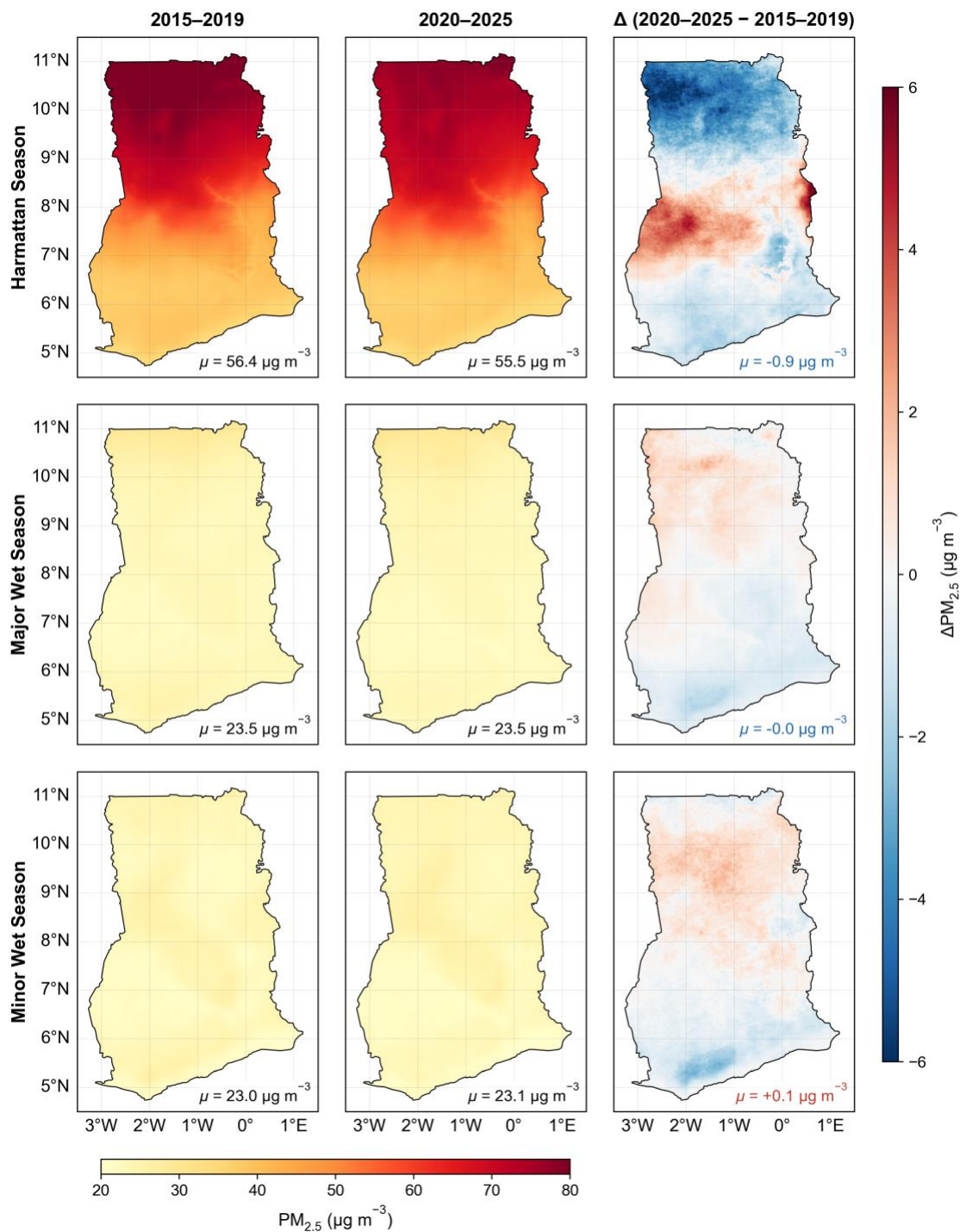
**Fig. S15.** Distribution of daily PM<sub>2.5</sub> concentrations across WHO air quality guideline (AQG) categories. Stacked bars show the percentage of days within each concentration range ( $\leq 15$ , 15–25, 25–35, 35–50, 50–75, and  $>75 \mu\text{g m}^{-3}$ ) for locations across Ghana (2005–2025). Cities are ordered geographically from north (left) to south (right) to highlight spatial gradients in air quality.



**Fig. S16.** Multi-year changes in seasonal mean PM<sub>2.5</sub> concentrations across Ghana (2005–2014). Maps show spatial patterns of mean PM<sub>2.5</sub> concentrations for the Harmattan, major wet, and minor wet seasons for two multi-year periods (2005–2009 and 2010–2014), along with the corresponding differences ( $\Delta = 2010\text{--}2014$  minus 2005–2009).  $\mu$  denotes the spatial mean concentration for each panel. The color scale for mean concentration levels is shown at the bottom, while the diverging color scale on the right indicates the difference.



**Fig. S17.** Multi-year changes in seasonal mean  $PM_{2.5}$  concentrations across Ghana (2010–2019). Maps show spatial patterns of mean  $PM_{2.5}$  concentrations for the Harmattan, major wet, and minor wet seasons for two multi-year periods (2010–2014 and 2015–2019), along with the corresponding differences ( $\Delta = 2015\text{--}2019$  minus 2010–2014).  $\mu$  denotes the spatial mean concentration for each panel. The color scale for mean concentration levels is shown at the bottom, while the diverging color scale on the right indicates the difference.



**Fig. S18.** Multi-year changes in seasonal mean  $PM_{2.5}$  concentrations across Ghana (2015–2025). Maps show spatial patterns of mean  $PM_{2.5}$  concentrations for the Harmattan, major wet, and minor wet seasons for two multi-year periods (2015–2019 and 2020–2025), along with the corresponding differences ( $\Delta = 2020\text{--}2025$  minus 2015–2019).  $\mu$  denotes the spatial mean concentration for each panel. The color scale for mean concentration levels is shown at the bottom, while the diverging color scale on the right indicates the difference.

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