RESEARCH PAPER

SPATIO-TEMPORAL CHANGES OF PARTICULATE MATTER ($PM_{2.5}$) OVER BRAZIL AND ITS CORRELATION WITH METEOROLOGICAL VARIABLES

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Received: September 10th 2024 / Accepted: January 23rd 2025 / Published: June 30th 2025
https://doi.org/10.24057/2071-9388-2025-3600

ABSTRACT. Fine particulate matter ($PM_{2.5}$), classified as airborne, adversely affects human health and the environment. This study examined the concentration and variability of $PM_{2.5}$ and its correlation with meteorological variables in Brazil. The annual average highest concentration of $PM_{2.5}$ (kg-m^{-3}) 5.65×10^{-9} was found in the western part of the country. A low concentration of $PM_{2.5}$ (kg-m^{-3}), 0.21×10^{-9} was reported in North, East, and South Brazil. Mann-Kendall and Sen's slope statistics were applied to find the trend and magnitude in the time series. Mann-Kendall (MAK)-Tau shows a positive significant trend (1 to 0.41) detected in the south, midwest, and southeastern Brazil. The Mann-Kendall (MAK)-Tau trend test was applied. The Sen's Slope rate ranged from 6.98 to 4.54 in the midwest, south, and southeast regions of Brazil, respectively. In 24 years, an overall negative $PM_{2.5}$ trend of -3.17 and -5.18 is shown in the north and northeast, respectively. This study evaluated $PM_{2.5}$ correlation with prevailing meteorological variables using various statistical techniques computed in R-Studio. Cross-wavelet Transform (CWT) analysis was used to examine the time and magnitude of $PM_{2.5}$ with prevailing meteorological variables. The CWT analysis is statistically significant. The application of CWT analysis has revealed high leading and lagging in-phase and anti-phase correlations with prevailing meteorological variables, e.g., relative humidity, precipitation, temperature, and wind speed variables that have influenced the temporal concentration of $PM_{2.5}$.

KEYWORDS: Mann-Kendall test, wavelet transformation, $PM_{2.5}$, meteorological variables, Brazil

CITATION: Shakrullah K., Tariq S., Shirazi S. A., Nasar-u-Minallah M., Shahzad H., Mariam A. (2025). Spatio-Temporal Changes Of Particulate Matter ($Pm_{2.5}$) Over Brazil And Its Correlation With Meteorological Variables. Geography, Environment, Sustainability, 2 (18), 82-90
https://doi.org/10.24057/2071-9388-2025-3600

Conflict of interests: The authors reported no potential conflict of interests.

INTRODUCTION

$PM_{2.5}$ is classified as airborne fine particulate matter. Its diameter is 2.5 microns or less (≤ 2.5 µm). $PM_{2.5}$ is generated from combustion sources and is documented as an important contributor to air quality (Chen et al. 2020; Wu et al. 2023). $PM_{2.5}$ mass concentration standard value is diverse for each country. In 2006, the safety limits for particulate matter (PM) in the atmosphere were established by the World Health Organization (WHO). The value range for $PM_{2.5}$ was determined to be 10 µg/m^3(per annum) and 25 µg/m^3 (24-hour average). In 2021, WHO issued the updated values of $PM_{2.5}$ and PM_{10} concentration. The maximum annual value should be correspondingly 5 µg/m^3 and 15 µg/m^3, for $PM_{2.5}$ and PM_{10} (WHO 2021). $PM_{2.5}$ poses a significant adverse effect not only on climate, ecosystems, and visibility but also on human health (Faridi et al. 2019; Fatima et al. 2023; Wang et al. 2023).

Biomass burning and energy use are the main contributors to the emission and concentration of particulate matter in the atmosphere. Other anthropogenic activities, e.g., brick kilns, agricultural activities, industrial and vehicle emissions, and waste incineration, are key factors of atmospheric particulate matter (Guttikunda et al. 2019; Amnuaylojaroen et al. 2020; Amit et al. 2021; Nasar-u-Minallah et al., 2024a; Nasar-u-Minallah et al., 2024b; Nasar-u-Minallah et al., 2025). These variables largely affect changing air circulation conditions, particle proliferation, and distribution. Furthermore, the metrological parameters can be used as a gauge to improve the projected values of $PM_{2.5}$ concentration at ground level (Liu et al. 2009). Climatic variability also affects the concentration of $PM_{2.5}$. The meteorological parameters (e.g., humidity, precipitation, temperature, and wind speed) can affect the mass concentration of $PM_{2.5}$ its dispersion, dilution, and accumulation in the air on a large scale (Tai et al. 2010; Westervelt et al. 2016). $PM_{2.5}$ is correlated with precipitation and wind speed. The high wind velocity will lead to turbulence and advection that increase the dispersion of pollutants. On the other hand, precipitation reduces the concentration of $PM_{2.5}$ through wet deposition and wet removal (Westervelt et al. 2016; Nguyen et al. 2017; Zhang et al. 2018). Several studies reported that meteorological factors are critical in the circulation and removal of particulate matter from the lower atmosphere (Sharma et al. 2017; Das et al. 2021; Singh et al. 2021; Nasar-u-Minallah 2024c). Saraswati et al. (2019) reported that the air pollutant's dispersion was primarily affected by the diurnal variation in boundary layer conditions and other meteorological factors. Precipitation, relative humidity, ambient temperature, wind velocity, and direction affect the concentration of particulate matter in ambient air and scatter them from areas of high to lower concentration (Begum et al. 2008; Saha et al. 2019; P. Sharma et al. 2022) A negative correlation is found between meteorological factors (such as dew point, wind gust, and ambient temperature) and $PM_{2.5}$ concentration at any given location (altitude and latitude) whereas, positively correlated with relative humidity (Das et al. 2021; Singh et al. 2021)

The carbon content in $PM_{2.5}$ scatters and absorbs the light and impacts atmospheric visibility (Shen et al. 2019). Approximately 50 percent variation in the concentration of $PM_{2.5}$ is reported due to diurnal variation in meteorological parameters (Tai et al. 2012). Wang & Ogawa (2015), they studied $PM_{2.5}$ and meteorological conditions in Nagasaki (Japan). The result indicated that $PM_{2.5}$ has a strong correlation with precipitation and a weak correlation with temperature. Several studies have carried out the correlation between $PM_{2.5}$ and meteorological parameters. In China (Wuhan) a study was led using Generalized Additive Models (GAM) on the correlation between $PM_{2.5}$ and meteorological parameters. The result indicated a 37% decrease in $PM_{2.5}$ concentration observed during precipitation (Zhang et al. 2018). Some studies have also reported seasonal variations of $PM_{2.5}$ with meteorological variables. For instance, Yang et al. (2017) stated that seasonal variation of $PM_{2.5}$ with temperature in different cities in China. In the winter, $PM_{2.5}$ has a strong correlation with temperature and a weak correlation in the autumn.

The disparity between $PM_{2.5}$ and temperature in different seasons was reported. Temperature showed a weak correlation with $PM_{2.5}$ in summer and autumn; however, there was a strong correlation in the spring and winter seasons (Chen et al. 2017). Conversely, an increase in temperature can cause variation in the formation of $PM_{2.5}$. The higher temperature increases the photochemical reaction involving $PM_{2.5}$. Additionally, a study conducted in Hong Kong from January – December 2013 reported a negative relationship between $PM_{2.5}$ and temperature (Zhao et al. 2019). Brazil is experiencing several socio-environmental challenges linked to air quality and climate variability. Brazil, being a continental country, is home to a diverse type of biomes. Apart from anthropogenic activities, diverse biomes are also a source of natural air pollution and spatio-temporal weather changes. Numerous studies have been carried out in Brazil focused on the concentration of primary pollutants, biomass burning (Squizzato et al. 2021; Castelhano et al. 2022) effect of particulate matter on health (Leão et al. 2023) $PM_{2.5}$ and PM_{10} concentrations (Braga et al. 2005) in major urban centers of Brazil. However, there is a dire need to conduct studies to evaluate the $PM_{2.5}$ concentration in Brazil (Pacheco et al., 2017). Finally, the literature on the correlation of particulate matter $PM_{2.5}$ with meteorological variables in Brazil is scarce, and to the best of our knowledge, no study has been carried out using cross-wavelet over the whole of Brazil.

This study aims to identify the gaps in previous studies and evaluate the variability of $PM_{2.5}$ and its correlation with meteorological parameters in Brazil over 24 years (2000-2024). For that purpose, the average maps of $PM_{2.5}$ and prevailing meteorological variables were prepared; in addition to that, we also used wavelet coherence to identify the relationship of $PM_{2.5}$ with other meteorological parameters. The Mann-Kendall test and Sen's slope methods were also used. The $PM_{2.5}$ concentration study is critical to getting a clear picture of the impact of anthropogenic activities on the environment. It is essential to develop effective planning and strategies to reduce air pollution. The effect of meteorological factors on $PM_{2.5}$ concentration is well-recognized and understood (Chen et al. 2020). There is a dire need for correct and precise daily $PM_{2.5}$ concentration assessment and projection to discourse environmental issues (Wang et al. 2022). Precise and accurate estimation of $PM_{2.5}$ could benefit the policymakers and enable them to initiate the measures that can help the public manage the means of transportation and travelling, thus decreasing the effect of $PM_{2.5}$ on their daily lives (Huang et al. 2021; Dong et al. 2022).

METHODS AND MATERIALS

Study Area

Brazil (geographic coordinates 10.00 S, 55.00 W) is the 5th largest country in the world and the largest in South America by geographical area (8,514,877 sq. km.) while 7th in terms of population size (217,663,781 souls). The climatic setup of the country is dominated by the equatorial and subtropical type of climate with high temperatures and erratic rainfall throughout the country, apart from the northeast of Brazil, which receives less rainfall and is virtually a semi-arid region (less than 700 mm of rain per annum).

Data sets

$PM_{2.5}$ (kg-m^{-3}) with spatial resolution 0.5°×0.625° was retrieved from the MERRA-2 reanalysis model. The surface radiative temperature (K) monthly product was taken at a spatial resolution of 1°×1° from the FLDAS model. Relative humidity (RH) (%) 700 hpa monthly product with a spatial resolution of 1°×1° was collected from Aqua Satellite through the AIRS instrument from 1-9-2002 to 01-01-2024. The wind speed (m s^{-1}) product was acquired at 0.5°×0.625° spatial resolution monthly through the MERRA-2 reanalysis model. Precipitation (kg m^{-2}s^{-1}) product with 0.5°× 0.625° spatial resolution and temporal resolution (monthly) was obtained from MERRA-2 reanalysis. The data sets were retrieved from NASA's Giovanni online web source[1]. Table 1 displays the data set used for this study.

ArcGIS 10.5 and R Studio were used to prepare the averaged maps of $PM_{2.5}$ and meteorological variables, such as humidity, precipitation, temperature, and wind speed, and calculate the Sen's slope and Mak Tau.

Mann-Kendal & Sen's Slope

The non-parametric Mann- Kendall (MK) test is adopted to evaluate the trend of $PM_{2.5}$. It is a robust method for analyzing the monotonic trend in time series, helping to

[1] https://giovanni.gsfc.nasa.gov/

Table 1. Satellite and model data used for the analysis

Data sets	Source	Spatial	Temporal	Duration
$PM_{2.5}$ (kg-m^{-3})	MERRA-2	0.05°×0.625°	Monthly	2000-2024
Relative Humidity (%)	AIRS	1°×1°.	Monthly	2002-2024
Wind Speed (m s^{-1})	MERRA-2	0.05°×0.625°	Monthly	2000-2024
Temperature (K)	FLDAS	1°×1°.	Monthly	2000-2024
Precipitation (kg m^{-2} s^{1})	MERRA-2	0.05°×0.625°	Monthly	2000-2024

identify increasing and decreasing trends in time series. It is calculated by following the equation (Eq. 1).

$$S = \sum_{l=1}^{n-1} \sum_{m=l+1}^{n} Sign\left(X_m - X_l\right) \quad (1)$$

From Eq. 1, S represents the MK-trend statistics, also known as Kendall's Tau (MAK-Tau). Whereas X_m and X_l are time series observations (Eq. 2).

$$Sign\left(X_m - X_l\right) = Sign\left(R_j - R_i\right) = \begin{cases} -1 for\left(X_m - X_l\right) < 0 \\ 0 for\left(X_m - X_l\right) = 0 \\ +1 for\left(X_m - X_l\right) > 0 \end{cases} \quad (2)$$

In the above equation (Eq. 2), R_j and R_i symbolize the rank of X_m and X_l time series values. The number of values tends to be normally distributed, and significance at 95% was determined using a p-value < 0.05. MAK-Tau is primarily used to test the correlation and strength between two variables. The values range from −1 to +1. Sen's slope is also a non-parametric test primarily used to identify the slope in time series data (Agarwal et al. 2021; Ray et al. 2021). Sen's slope is computed in Eq. 3.

$$Q_m = \frac{X_k + X_m}{K - m} for\ m = 1 \ldots N \quad (3)$$

From Eq. 3, Q_m represents the estimated slope for each pair of observations. The subscripts k and m are the time steps, where $K > m$. The Mann-Kendall's and Sen's slopes were calculated in R Studio.

Wavelet analysis

Several researchers have extensively used wavelet transformations for time series data. Firstly, wavelet analysis was used for seismic signal analysis (Chen et al. 2020). Nowadays, wavelet analysis has been extensively used in all fields (e.g., mathematics, science, engineering, and geophysics) (Zhang et al. 2017). Discrete wavelet (DWT) and continuous wavelet (CWT) are the two main types of wavelet transform analysis (Cholianawati et al. 2024a). Several studies used a cross-wavelet to analyze and find out the correlation between $PM_{2.5}$ and meteorological variables (Barik et al. 2020; Meng & Sun 2021; Fattah et al. 2023; Jang & Jung 2023; Cholianawati et al. 2024b). The Morlet wavelet function (ψ) and cross-wavelet power are used to understand the correlation between $PM_{2.5}$ and other meteorological variables (Eq. 4).

$$Wave(\tau, s) = \sum_t xt \frac{1}{\sqrt{s}} \psi^* \left(\frac{t - \tau}{s}\right) \quad (4)$$

In Eq. 4, xt denotes the time domain. Whereas $Wave(\tau,s)$ indicates time series in continuous wavelets, (xt, T =1,2,3…N) concerning wavelet ψ is defined. where s represents the wavelet scale, τ stands as the position of the wavelet window in time or the translated time index, and Ψ is the mother wavelet function with * representing/indicating its complex conjugate solution. The wavelet analysis of the time series of $PM_{2.5}$ and meteorological variables was computed in R-Studio using the wavelet comp package.

RESULTS AND DISCUSSION

Spatio-temporal distribution of $PM_{2.5}$ and meteorological variables

Fig. 1 shows the spatiotemporal correlation of $PM_{2.5}$ with meteorological variables from 2000-2024 in Brazil. Fig. 1a illustrates Brazil's spatiotemporal $PM_{2.5}$ (kg-m^{-3}) patterns from 2000-2024. The map indicates that the maximum $PM_{2.5}$ (kg-m^{-3}) concentration of 5.65×10^{-9} was found in the western part of the country. A low concentration of $PM_{2.5}$ (kg-m^{-3}), 0.21×10^{-9} was reported in eastern Brazil. The descriptive statistics of all study variables are provided in Table 2. The mega-cities of Brazil are home to millions of people residing there and vehicles as well, and they face numerous problems related to air pollution and particulate matter, which is one of them. Vehicles are considered the primary reason for pollutants' emission into the atmosphere in major urban centers of Brazil (de Fatima Andrade et al. 2012; Requia & Azevedo de Melo 2024). Numerous studies documented the other sources of particulate matter $PM_{2.5}$, e.g., biomass burning, aerosols from sea salt, and industrial waste and traffic congestion (Gioia et al. 2010; de Fatima Andrade et al. 2012; Souza et al. 2014). The central areas of Brazil receive a large number of gases and particulate matter emitted into the atmosphere due to the burning of biomass from July to October (dry season) (Butt et al. 2020).

Table 2. Descriptive statistics

	$PM_{2.5}$ (kg-m^{-3})	Humidity (%)	Wind Speed (m s^{-1})	Temperature (K)	Precipitation (kg m^{-2} s^{-1})
Mean	1.81×10^{-9}	46.71	3.55	298.41	4.96 ×10^{-5}
Median	5.82×10^{-11}	46.92	3.45	298.25	1.41×10^{-6}
Standard Deviation	1.61×10^{-9}	1.97	0.38	1.40	2.40×10^{-5}
Minimum	1.97×10^{-9}	41.34	2.83	295.54	1.24×10^{-5}
Maximum	5.78×10^{-9}	51.00	4.53	302.92	1.09×10^{-4}

$PM_{2.5}$ concentration induced by wildfire has been reported as high in the central-western part of Brazil, affecting the health of human beings (Butt et al. 2020; Ye et al. 2022; Jang & Jung 2023). A study found, that approximately 80% of $PM_{2.5}$ concentration in Brazil (Amazon region) was associated with deforestation (Urrutia-Pereira et al. 2021). It has been reported that wildfires from the north and west sides and transportation of wildfires from suburbs also influence the concentration of $PM_{2.5}$ in the country (Jang & Jung 2023). Fig.1b highlights the relative humidity in (%). The maximum relative humidity was observed in the north and midwest. The minimum RH was observed in the northeast, southeast, and south. Fig. 1c illustrates the spatio-temporal variability of precipitation over 24 years in the study area. The map depicts the highest precipitation, 9.88 (kg $m^{-2}s^{-1}\times10^{-5}$), in the east and north of Brazil. The precipitation of 1.37 (kg $m^{-2}s^{-1}\times10^{-5}$) is observed in the western and central parts of the country. Fig. 1d shows the temperature (K) in Brazil. The maximum temperature is 303K, found in the northeast and midwest of Brazil. In the north, the temperature range is approximately 296K-299K. The lowest temperature is found from southeast to south. The temperature range lies between 285K and 288K from southeast to south. Our findings are aligned with previous literature. Several studies have revealed that an increase in temperature and a substantial decrease in precipitation are observed in northeast Brazil (Marengo et al. 2017; Da Silva et al. 2019; Costa et al. 2020). Intensified extreme dry spells or events due to the El Niño Southern Oscillation were reported in northeast Brazil (Marengo et al. 2017)

Fig. 1e illustrates the wind speed (m s^{-1}) in the study area. The highest wind velocity, 5.62 (m s^{-1}) was found in the north and west of Brazil. The wind velocity between 4.56 (m s^{-1}) to 3.51 (m s^{-1}) is observed in the west of Brazil. The wind speed, ranging from 1.41 (m s^{-1}) to 0.35 (m s^{-1}) is seen from the east and central parts of the study area. Chen et al. (2017), reported that stronger wind speeds lead to faster parallel diffusion of pollutants, which eventually drops the $PM_{2.5}$ concentration. Wind speed conditions are considered the critical factor in the evaporation process of $PM_{2.5}$. The high wind velocity increased the dispersion of pollutants horizontally, eventually decreasing the $PM_{2.5}$ concentration (Chen et al. 2018). The acceleration of wind speed intensifies the $PM_{2.5}$ evaporation rate, leading to a significant decrease in the concentration of meteorological parameters such as temperature, wind velocity, temperature inversion, relative humidity, and atmospheric pressure, which significantly influence the dispersal and accretion of $PM_{2.5}$ (Ocak & Sezer Turalioglu 2008; Wang et al. 2023). Relevant studies have shown that the effect of $PM_{2.5}$ varies with different weather conditions. A study in China from 2015 to 2017 found that there was a weak link between particulate matter and both rain and humidity (Han et al. 2018).

Fig. 2a exhibits the variability in the spatial trends and magnitude of $PM_{2.5}$ using the Mann-Kendall trend test (Sen's slope) over Brazil during the study period 2000-2024. The trend rate ranges from 6.98 to 4.54 in the midwest, south, and southeast regions of Brazil, respectively. A negative trend of $PM_{2.5}$ ranging from -3.17 to -5.18 is observed in the north and northeast respectively. Fig. 2b spatial trend's magnitude (Mak-Tau) shows a positive trend (1 to 0.41) detected in the south, midwest, and southeastern Brazil, but this trend seems highly significant.

Cross Wavelet Analysis $PM_{2.5}$ and meteorological parameters

The Cross Wavelet Transform (CWT) is a tool used to study the relationship of time and magnitude in two-time series. Coherency explains a constant pattern and identifies the correlation between two variables. The wavelet uses the arrows to describe the pattern; the right arrows show an in-phase relationship. On the other hand, the left arrows depict the anti-phase, or inverse, correlation. In addition, the leading relationship is indicated by upward arrows. The downward arrow shows the lagging correlation between the two variables (Aguiar-Conraria et al. 2008).

In Fig. 3, the red color indicates the highest value, while the blue color indicates the lowest value. The coherency between $PM_{2.5}$ and relative humidity (Fig. 3a) mostly shows the leading and lagging situations. It illustrates a periodic cycle of 8 to 16 days, with cross wavelet power of ~1.2 to 1.5. The in-phase (direct) relationship is found between two variables. The value range indicates a strong positive relationship. The cross-wavelet transfer between $PM_{2.5}$ and precipitation is shown in Fig. 3b a positive covariance/coherence in the dataset. A leading and lagging sequence phase is detected between two variables. The lagging variables, from 2000 to 2024, are dominated by an anti-phase relationship. Cross wavelet power of ~1.3 to 1.6 is found. The wavelet displays an 8- to 16-day periodic cycle.

There is a strong connection between $PM_{2.5}$ and temperature (Fig. 3c), which displays an anti-phase (inverse) relationship, along with leading and lagging states. A strong Cross wavelet power of ~1.3- 1.6 is seen in datasets. Substantial periodic cycles of 8–16 days are observed. The relationship between $PM_{2.5}$ and wind speed (Fig. 3d) exhibits in-phase (direct) situations in datasets, whereas leading and lagging phases. It displays a significant periodic cycle of 8–16 days, having cross wavelet power of ~1.5 to 1.8, and with several days are 150-200. Wavelet transformation helps in comprehending the aerosol nature in Brazil's regions. For instance, the long-term significant periodicities in the cross wavelet between precipitation and $PM_{2.5}$ indicate the presence of fine-mode aerosols in the atmosphere, which maintain the air pressure in the upper atmosphere. Similarly, the results of $PM_{2.5}$ with relative humidity indicate aerosol plumes exposed to sufficient atmospheric moisture, resulting in scattering and diffusion of PM particles. Moreover, $PM_{2.5}$ has an inverse relationship with temperature (Vaishali et al. 2023) as can be seen through the out-of-phase relationships seen from the wavelet figure. Overall, the links between $PM_{2.5}$ and all the meteorological variables help to understand these variables in Brazil. The dataset used in this study is a monthly dataset rather than a daily one, which is one reason why the periodic cycles mostly appear between 8-16. Moreover, the frequency shown in the period has prolific short-term periodicities between 2 and 4 periodic cycles, but high wavelet powers seem to be between 8-16 throughout the days. The periodic cycle shows the relationship of PM particles with meteorological variables in the upper atmosphere, which is highly significant in the medium-run range (8-16).

CONCLUSION

The harmful impact of $PM_{2.5}$ on visibility, climate change, and human health has recently attracted the attention of scientists worldwide. The findings of this study uncover the spatio-temporal variations of $PM_{2.5}$ concentration across Brazil. The highest concentration was observed in northern and western Brazil. The current study evaluates the correlation of $PM_{2.5}$ on meteorological variables using cross-wavelet analysis over Brazil. The averaged maps of $PM_{2.5}$ and meteorological parameters have been prepared. The cross-wavelet transformation was calculated in RStudio to determine the correlation of $PM_{2.5}$ with all meteorological

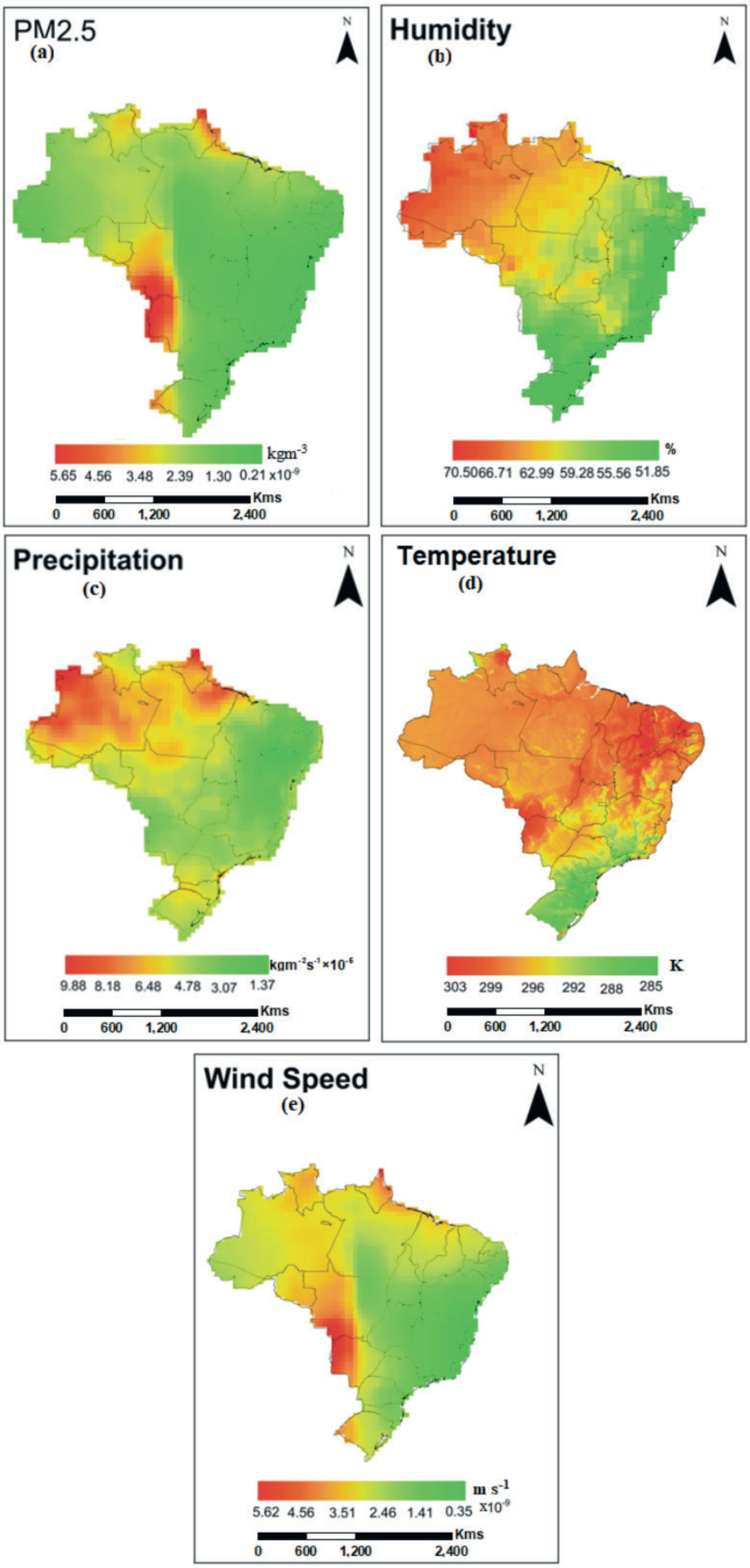

Fig. 1. Averaged maps of (a) $PM_{2.5}$, (b) humidity, (c) precipitation, (d) temperature, and (e) wind speed over the 2000-2024 period

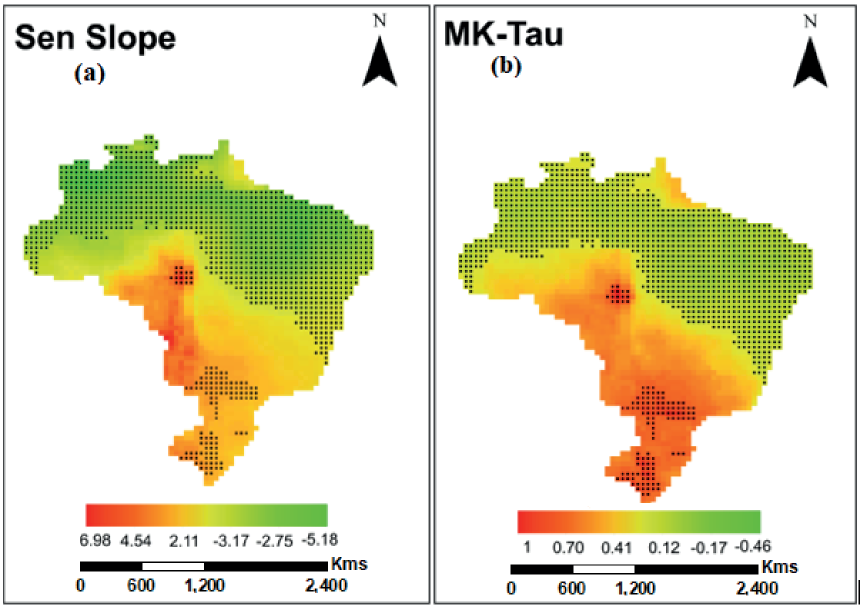

Fig. 2. (a) Sen's Slope and (b) Mann-Kendall (MAK)-Tau over the 2000-2024 period

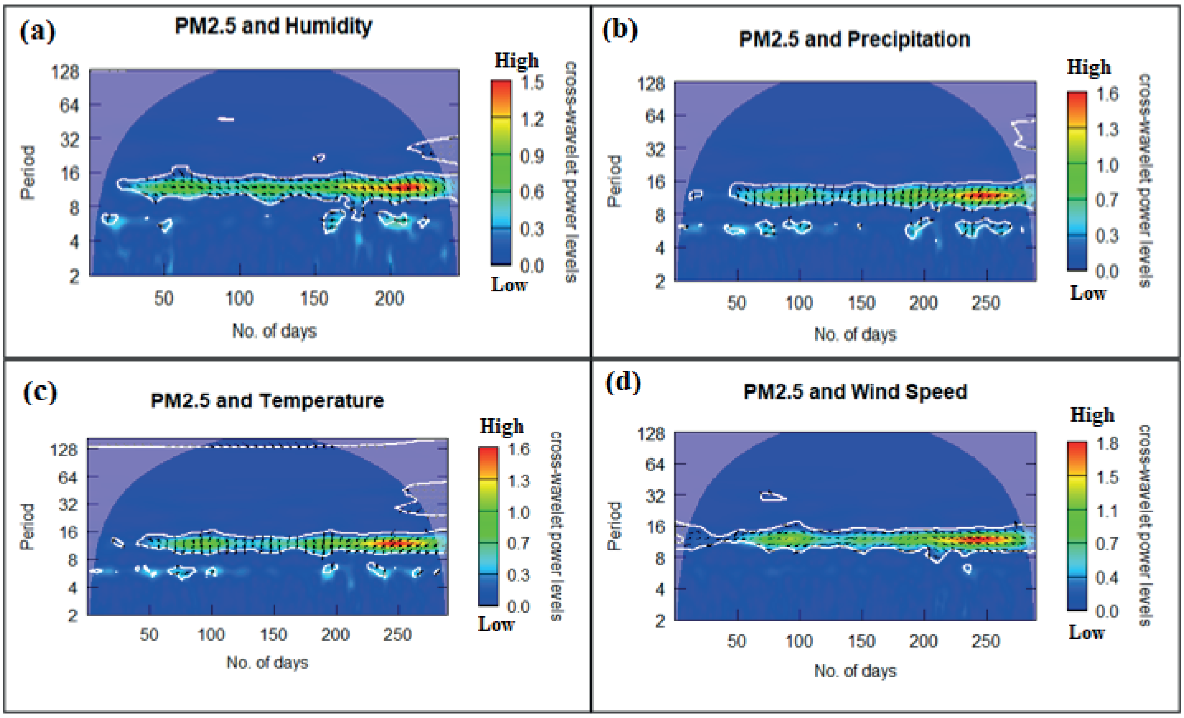

Fig. 3. $PM_{2.5}$ and meteorological variable cross wavelet spectra: (a) $PM_{2.5}$ and humidity, (b) $PM_{2.5}$ and precipitation, (c) $PM_{2.5}$ and temperature, and (d) $PM_{2.5}$ and wind speed

parameters, e.g. humidity, precipitation, temperature, and wind speed. All meteorological parameters exhibit/show remarkable periodicities of 8-16 days. $PM_{2.5}$ correlation was detected as an anti-phase (inverse) relationship with temperature and precipitation. In contrast, humidity and wind speed depicted the in-phase (synchronous/positive) relationship with $PM_{2.5}$. A leading and lagging phase is observed in all covariates. $PM_{2.5}$ results are slightly higher than the WHO standards in northern and western Brazil. There is a dire need to take measures to reduce the $PM_{2.5}$ concentration in the study area. The outcomes of this study would provide valuable insights for future research in Brazil. This study will help stakeholders create policies and strategies to reduce $PM_{2.5}$ in the atmosphere, which is more harmful to human health than any other particulate matter. This study has certain limitations due to the non-availability of the data set, and we used model data. The results and observations can be compared with ground data to validate the results to get a clear picture and help make robust decisions. Therefore, higher levels of $PM_{2.5}$ disagree with Sustainable Development Goal (SDG) 11.6.2. The main objective of SDG is to lessen the adverse effects of air pollutants on human health, including fine particulate pollution. ■

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