

# Quantifying Spatio-Temporal Dynamics of African Dust Detection Threshold for *PM*<sub>10</sub> Concentrations in the Caribbean Area Using Multiscale Decomposition

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Plocoste T, Euphrasie-Clotilde L, Calif R and Brute F-N (2022) Quantifying Spatio-Temporal Dynamics of African Dust Detection Threshold for PM<sub>10</sub> Concentrations in the Caribbean Area Using Multiscale Decomposition. Front. Environ. Sci. 10:907440. doi: 10.3389/fenvs.2022.907440 Due to African dust, the Caribbean area is known to have one of the highest incidences of asthma on the planet. Consequently, it is crucial to dissociate the impact of local sources from large scale sources in this region. The aim of this study was to estimate the  $PM_{10}$ detection threshold for dusty events using a statistical approach and a dynamic approach. To carry out this analysis, PM10 time series from Martinique (MAR), Guadeloupe (GPE) and Puerto-Rico (PR) were used between 2006 and 2016. The statistical analysis highlighted that the distance from the African coast is a key feature for  $PM_{10}$  concentrations distribution with the highest at MAR (26.52 µg/m<sup>3</sup>) and the lowest at PR (24.42 µg/m<sup>3</sup>). The probability density function analysis showed that MAR-GPE-PR distributions converge towards a same point between the first and the second maximum probability value at 28 µg/m<sup>3</sup>. The dynamical analysis with the Complete Ensemble Empirical Mode Decomposition with Adaptive Noise (CEEMDAN) and the Improved CEEMDAN (ICEEMDAN) validated the 28 µg/m<sup>3</sup> found with the statistical analysis. The analysis of HYSPLIT back trajectories confirmed this threshold. Thus, our results indicated that 28  $\mu$ g/m<sup>3</sup> is the *PM*<sub>10</sub> detection threshold for African dust in the Caribbean basin. It will therefore be a good indicator allowing the competent authorities to take the appropriate decisions to protect vulnerable populations during dusty events.

Keywords: PM10, african dust threshold, statistical analysis, multiscale analysis, caribbean area

# **1 INTRODUCTION**

Mineral dust is the most abundant type of aerosol on Earth (Jaenicke, 1988; Ramanathan et al., 2001). The Saharan and the Sahelian desert are the main mineral dust sources (d'Almeida, 1986; Tegen and Schepanski, 2009; Knippertz and Todd, 2012). Since 1967, many studies have demonstrated that large amount of African dust were routinely being transported across the Atlantic to the Caribbean (Delany et al., 1967; Parkin et al., 1967; Huang et al., 2010; Adams et al., 2012; Chin et al., 2014; Kim et al., 2017; Tegen and Schepanski, 2018; Euphrasie-Clotilde et al., 2020; Prospero et al., 2021), to mention a few.

In ecology, mineral dust plays a key role in nutrient cycling, allowing the fertilization of ecosystems in the American continent. The transport of these nutrients helps nourish the Amazon, compensating for the scarcity of nutrients in the soil of the region (Griffin, 2007). For example, soils in the Amazon Basin are

deficient in phosphorus, essential to soil fertility (Prospero et al., 2020). The iron found in mineral dust is also essential in the ocean for the growth of phytoplankton, which is the basis of oceanic food chains (Villar-Argaiz et al., 2018). These nutrients reach the surface layer and the marine boundary layer through dry deposition or wet deposition (Rizzolo et al., 2017; López-García et al., 2021). In addition, the dry and hot conditions of dusty air masses alter atmospheric stability, inhibiting the formation and intensification of tropical cyclones (Dunion and Velden, 2004; Braun et al., 2012; Huang et al., 2020). As some of the solar energy does not reach the ocean surface when there is airborne dust, this lowers the temperature and prevents evaporation of water which is essential to the formation of hurricanes and tropical storms (Nowottnick et al., 2018).

Despite all these positive aspects, mineral dust also has harmful effects on health. In the literature, many studies associated it with cardiovascular and respiratory diseases, prematurity, general mortality, and a series of infectious diseases (Comrie, 2005; Griffin, 2007; Chen et al., 2010; Baughman et al., 2011; Tobías et al., 2011; Schweitzer et al., 2018; Dominguez-Rodriguez et al., 2020; Urrutia-Pereira et al., 2021), to cite a few. The populations most susceptible to shortterm effects of airborne dust are: 1) the elderly, due to their lower immune capacity (Jiménez et al., 2010); 2) individuals affected by chronic cardiopulmonary diseases (Dominguez-Rodriguez et al., 2020); and (iii)children, whose lungs and airways are not fully developed (Yu et al., 2012). Furthermore, African dust can also carry bacteria, fungi and viruses (Sakhamuri and Cummings, 2019).

Many studies have highlighted the impact of desert dust on Caribbean population. In Trinidad, Gyan et al. (2005) exhibited a relationship between African dust and respiratory stress, increased asthma exacerbations and emergency admissions, and increased daily rates of pediatric hospitalization. In Barbados and Trinidad, Monteil (2008) also observed a significant increase in pediatric admissions 7 days after major dust outbreaks. Cadelis et al. (2014) found the same behavior in Guadeloupe with a relationship between particulate matter from African dust and an increased risk for visits to emergency services in children with asthma. In Grenada, Akpinar-Elci et al. (2015) has shown that mineral dust coupled with seasonal humidity allows the formation of inhalable particles that aggravate asthma among residents. Thus, the Caribbean are known to have some of the highest incidences of asthma on the planet due to dust outbreaks (Urrutia-Pereira et al., 2021). A recent study made by Viel et al. (2020) in Guadeloupe also indicated that Saharan dust seems to influence weight but not length or head circumference at birth. For all the reasons aforementioned, it is therefore crucial to determine the detection threshold for African dust in the Caribbean basin. Mineral dust is a mixture of fine and coarse particles (Does et al., 2016). In this study, the authors focused on particles lower or equal to 10 µm in diameter called PM<sub>10</sub> which are present in significant quantities in dust plumes (Petit et al., 2005) and can reach the trachea and the bronchi after inhalation (Urrutia-Pereira et al., 2021).

Previous studies have shown that the daily concentration of  $PM_{10}$  in the Caribbean area frequently exceeds the 50 µg/m<sup>3</sup>

recommended by the World Health Organization for an average of 24 h during the boreal summer (Martet et al., 2009; Prospero et al., 2014). Recently, Euphrasie-Clotilde et al. (2020) highlighted a dusty threshold of 35  $\mu$ g/m<sup>3</sup> using daily *PM*<sub>10</sub> concentrations and optical measurements, i.e. Aerosols Optical Depth (AOD). However, for daily *PM*<sub>10</sub> concentrations below 35  $\mu$ g/m<sup>3</sup>, this study showed that the associated daily AOD data are also characterized by marine aerosols. The aim of this study is therefore to refine this threshold using statistical and dynamic methods with daily *PM*<sub>10</sub> time series from three Caribbean islands: Martinique, Guadeloupe and Puerto-Rico.

In nature, it is well known that physical processes are often non-linear and non-stationary, showing the coexistence of different spatial-temporal scales (Huang et al., 2003; Bai et al., 2016; Plocoste et al., 2019). Many studies have demonstrated the non-linear properties of  $PM_{10}$  time series in the Caribbean area (Plocoste et al., 2017, 2020b, 2021b; Plocoste and Calif, 2021; Plocoste, 2022a; Plocoste, 2022b). This is the reason why multiscale decomposition methods were used to estimate  $PM_{10}$ detection threshold between local sources and large scale sources in a dynamical way. Empirical Mode Decomposition (EMD) is an adaptive time-frequency data analysis proposed by Huang et al. (1998) whose objective is to decompose any time series into a sum of different Intrinsic Mode Functions (IMFs) with a sifting procedure (Huang et al., 1999; Flandrin and Goncalves, 2004; Huang and Schmitt, 2014). However, this technique introduces a serious drawback which is the mode mixing problem (Yeh et al., 2010), i.e. the presence of very similar oscillations in different modes (Cao et al., 2019). To overcome the scale separation problem, the Ensemble EMD (EEMD) is introduced by Wu and Huang (2009). In this approach, randomly generated white noise series are added to the original signal to help the sifting process to avoid mode mixing. Nevertheless, the EEMD method cannot completely eliminate white noise after signal reconstruction (Luukko et al., 2016). To solve this issue, the Complete Ensemble Empirical Mode Decomposition with Adaptive Noise (CEEMDAN) was proposed by Torres et al. (2011). As CEEMDAN modes have some residual noise and the signal information presents some spurious modes in the early stages, Colominas et al. (2014) introduced the Improved CEEMDAN (ICEEMDAN), obtaining components with less noise and more physical meaning. Recently, CEEMDAN and ICEEMDAN approaches have been applied in several fields such as finance (Cao et al., 2019; Wu et al., 2020), entropy (Kuai et al., 2018; Kou et al., 2020), air pollution (Du et al., 2020; Xiao et al., 2021; Plocoste, 2022b) and renewable energy (Zhang et al., 2017; Rezaie-Balf et al., 2019; Gao et al., 2020; Sibtain et al., 2021), to mention a few. Usually, these methods are used for forecasting purposes. To our knowledge, no study has yet used CEEMDAN and ICEEMDAN frameworks to investigate the background atmosphere of an air pollutant.

# **2 EXPERIMENTAL DATA**

 $PM_{10}$  data of three Caribbean islands were included in this analysis with respectively Martinique (MAR), Guadeloupe





(GPE) and Puerto-Rico (PR) (see Figure 1). MAR and GPE measurements are made by Les Associations Agréées de Surveillance de Qualité de l'air, a national organization that overseas air quality in each of the French administrative regions while PR measurements is carried out by the

United States air quality network. For MAR-GPE-PR,  $PM_{10}$  data are respectively released by MadininAir, Gwad'Air and Air Now agencies. It is important to emphasize that each island uses the same sensor and the same measurement protocol. Using the Thermo Scientific Tapered Element

Oscillating Microbalance (TEOM) models 1400 ab and 1400-FDMS,  $PM_{10}$  data is continuously sampled and stored as 15 min averages which were used to calculate daily averages concentrations analyzed in this study. For 24 h, the measurement accuracy is  $\pm 0.5 \,\mu\text{g/m}^3$  (Euphrasie-Clotilde et al., 2020).

For MAR-GPE-PR,  $PM_{10}$  measurements are respectively performed at Schoelcher (14.648°N 61.099°W, 2006–2016, urban area); Pointe-à-Pitre (16.242°N 61.541°W, 2006–2014, urban area) and Baie-Mahault (16.256°N 61.590°W, 2015–2016, suburban area); and Cataño (18.431°N 66.142°W, 2006–2016, suburban area). Consequently, 11 years of daily database is available for MAR-GPE-PR with 3,903–3,503–3,718 data points. A sequence of  $PM_{10}$  time series is displayed in **Figure 2**. One can observe that  $PM_{10}$  time series seem to follow the same temporal pattern for the three islands.

# **3 THEORETICAL FRAMEWORK**

# **3.1 Hybrid Single Particle Lagrangian** Integrated Trajectory

In literature, many studies have shown that Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) is a robust tool to investigate the origin of air masses (McGowan and Clark, 2008; Ashrafi et al., 2014; Stein et al., 2015). In air pollution, HYSPLIT back trajectories are commonly performed to assess the origin of dusty air masses in the Caribbean basin (Prospero et al., 2014; Gläser et al., 2015; Euphrasie-Clotilde et al., 2020; Plocoste et al., 2020a). In HYSPLIT, the meteorological database used is the National Center for Atmospheric Research/National Centers for Environmental Prediction (NCAR/NCEP) re-analysis data (Kalnay et al., 1996). The following parameters are used to generate all daily back trajectories for MAR, GPE, and PR islands between 2006 and 2016: 1) Altitude: 1,500 m according to the properties of the Saharan Air Layer (SAL); 2) Starting locations: MAR (14.64°N; 61.09°W), GPE (16.24°N, 61.53°W), PR (18.43°N; 66.14°W); 3) Start time: 12 UTC (8 a.m. local time); 4) Duration: 10 days (240 h). Once back trajectories data as latitude/ longitude and positions are created by HYSPLIT, this latter are transferred to QGIS geographic information system for visualisation (version QGIS-2.18.14, http://www.qgis.org/fr/site/ ). The protocol used in this study was validated and widely explained by Euphrasie-Clotilde et al. (2020).

# 3.2 CEEMDAN

As CEEMDAN is based on EEMD frame, the decomposition theory of EEMD is firstly presented. Let *s* the target signal. The EEMD algorithm can be describe as follow (Wu and Huang, 2009; Colominas et al., 2014):

- Step 1: Create  $s^{(i)} = s + \beta w^{(i)}$ , where  $w^{(i)}$  (i = 1, ..., I) is a different white Gaussian noise and  $\beta > 0$ .
- Step 2: Fully decompose each  $s^{(i)}$  (i = 1, ..., I) by EMD, obtaining the modes  $IMF_k^{(i)}$ , where k = 1, ..., K represents the mode.

• Step 3: Put  $\overline{IMF}_k$  as the *k*th mode of *s*, obtained by averaging the corresponding modes:  $\overline{IMF}_k = \frac{1}{I}\sum_{i=1}^{I}IMF_k^{(i)}$ .

In order to extract each  $IMF_k^{(i)}$ , a different number of sifting iterations is required. It can be underlined that in EEMD, each  $s^{(i)}$  is decomposed independently from the other realizations and for every one of them a residue  $r_k^{(i)} = r_{k-1}^{(i)} - IMF_k^{(i)}$  is obtained at each stage, with no connection between the different realizations. Nevertheless, this latter introduces some problems in EEMD. Firstly the decomposition is not complete. Then, different realizations of signal plus noise might produce different number of modes. To overcome these drawbacks, Torres et al. (2011) and Colominas et al. (2012) introduced a new ensemble method termed the CEEMDAN.

The main concept behind the CEEMDAN frame is the following:  $s^{(i)}$  are generated from *s* and the first mode  $\widetilde{IMF_1} = \overline{IMF_1}$  is calculated as in EEMD. Independently of the realization of the noise, a first unique residue is then obtained by (Colominas et al., 2012):

$$r_1 = s - I\widetilde{MF}_1. \tag{1}$$

Thereafter, EMD first mode is computed from an ensemble of  $r_1$  plus different realizations of a particular noise. The second mode  $\widehat{IMF}_2$  is the average of these modes. The next residue is defined as  $r_2 = r_1 - \widehat{IMF}_2$ . Other modes continue this process until a stopping criterion is met.

The following algorithm presents the CEEMDAN method in details. Specify  $E_j(\cdot)$  as the operator that creates the *k*th mode obtained by EMD frame and let  $w^{(i)}$  be a realization of zero mean unit variance white noise. If *s* is the target signal (Torres et al., 2011; Colominas et al., 2012):

Step 1: For every *i* = 1, ... *I*, decompose each s<sup>(i)</sup> = s + β<sub>0</sub>w<sup>(i)</sup> by EMD, until its first mode, and set the first CEEMDAN mode as:

$$\widetilde{IMF}_{1} = \frac{1}{I} \sum_{i=1}^{I} IMF_{1}^{(i)} = \overline{IMF_{1}}.$$
(2)

- Step 2: For k = 1, compute the first residue as in Eq. (1):  $r_1 = s - I\widetilde{MF}_1$ .
- Step 3: For every i = 1, ..., I, decompose each  $r_1 = s + \beta_1 E_1(w^{(i)})$  by EMD and define the second CEEMDAN mode as:

$$\widetilde{IMF}_{2} = \frac{1}{I} \sum_{i=1}^{I} E_{1} (r_{1} + \beta_{1} E_{1} (w^{(i)})).$$
(3)

• Step 4: For k = 2, ..., K, compute the k-th residue:

$$r_k = r_{(k-1)} - I\widetilde{MF}_k.$$
 (4)

• Step 5: For every i = 1, ..., I, decompose each  $r_k + \beta_k E_k(w^{(i)})$  by EMD, until define the (k + 1)th CEEMDAN mode as:

$$I\widetilde{MF}_{(k+1)} = \frac{1}{I} \sum_{i=1}^{I} E_1 \left( r_k + \beta_k E_k \left( w^{(i)} \right) \right).$$
(5)

• Step 6: Go to the step 4 for next k.

Iterate the steps 4–6 until the obtained residue can no longer be further decomposed by the EMD, because either it satisfies IMF criteria or it has been less than the three local extrema.

After construction of CEEMDAN, the final residue can be expressed as:

$$r_k = s - \sum_{k=1}^{K} I \widetilde{MF}_k, \tag{6}$$

with k the total number of modes. Thus, the given signal s can be expressed as:

$$s = \sum_{k=1}^{K} I \widetilde{MF}_k + r_k.$$
<sup>(7)</sup>

**Equation** 7 makes the proposed decomposition complete and gives an exact reconstruction of the original data. It is important to emphasize that the final number of modes is determined only by the data and the stop criterion. The selection of the Signal to Noise Ratio (SNR) are determined by the coefficients  $\beta_k = \varepsilon_k std(r_k)$ .

#### 3.3 ICEEMDAN

Even if CEEMDAN solved the problem of the number of modes for different realizations of signal plus noise, it still has some problems which may be improved (Colominas et al., 2014; Thuraisingham, 2021). Indeed, its modes contain some residual noise and the signal information presents some spurious modes in the early stages of the decomposition (Colominas et al., 2014). To improve on this, Colominas et al. (2014) proposed the Improved CEEMDAN (ICEEMDAN), obtaining components with less noise and more physical meaning. The ICEEMDAN algorithm is described based on CEEMDAN as follows (Colominas et al., 2014):

• Step 1: Compute by EMD the local means of *I* realizations  $s^{(i)} = s + \beta_0 E_1(w^{(i)})$  to get the first residue:

$$r_1 = \langle M(s^{(i)}) \rangle. \tag{8}$$

• Step 2: For k = 1, compute the first mode:

$$\widetilde{IMF}_1 = s - r_1. \tag{9}$$

Step 3: Estimate the second residue as the average of local means of the realizations r<sub>1</sub> + β<sub>1</sub>E<sub>2</sub>(w<sup>(i)</sup>); then the second mode is defined as:

$$\widetilde{IMF}_2 = r_1 - r_2 = r_1 - \langle M(r_1 + \beta_1 E_2(w^{(i)})) \rangle.$$
(10)

• Step 4: For k = 3, ..., K, compute the *k*th residue:



**FIGURE 3** Box-plot of daily  $PM_{10}$  concentrations at Martinique (MAR), Guadeloupe (GPE) and Puerto-Rico (PR) from 2006 to 2016. The horizontal lines within the box represent the median while the bottom and top of each box are the 25th and 75th percentiles. The whiskers are the 5th and 95th percentiles.

$$r_{k} = \langle M(r_{(k-1)} + \beta_{(k-1)}E_{k}(w^{(i)})) \rangle.$$
(11)

• Step 5: Calculate the *k*th mode:

$$\widetilde{IMF}_k = r_{(k-1)} - r_k. \tag{12}$$

• Step 6: Repeat step 4 for the next k.

## **4 RESULTS AND DISCUSSION**

# 4.1 Descriptive Statistics

### 4.1.1 Overall Analysis

To understand the behavior of  $PM_{10}$  concentrations in the Caribbean area, the descriptive statistics are firstly computed. **Figure 3** shows the box-plot of daily  $PM_{10}$  concentrations at MAR, GPE and PR from 2006 to 2016. At first glance, the box-plots seem to present the same pattern. The 25th percentiles and the medians are almost similar at MAR-GPE-PR with respectively 15.0-16.7-16.0  $\mu g/m^3$  and 21.0-20.7-20.0  $\mu g/m^3$ . As regards the 75th and the 95th percentiles, a decrease is observed from MAR to PR with respectively 32.0-30.7-27.0  $\mu g/m^3$  and 57.0-52.0-43.0  $\mu g/m^3$ . For each island, one can observe the presence of outliers. They are mainly due to African dust outbreaks. Indeed, a study made by Euphrasie-Clotilde et al. (2020) with NOAA-HYSPLIT day-to-day back trajectories highlighted that 98% of concentrations higher or equal to 35  $\mu g/m^3$  in the Caribbean area are related to air masses coming from the African coast.

**Table 1** presents the statistical parameters for each studied island. The mean  $(\overline{M})$ , standard deviation  $(\sigma)$ , skewness (S) and kurtosis (K) were chosen to respectively analyze the trend, fluctuation, asymmetry and intermittency of  $PM_{10}$  concentrations. In pollution studies, highly intermittent time series will have a higher kurtosis (Windsor and Toumi, 2001). Overall, from MAR to PR, the values of  $\overline{M}$  and  $\sigma$  decrease while S

**TABLE 1** Statistical parameters (Mean ( $\overline{M}$ ), Standard deviation ( $\sigma$ ), Minimum (*Min*), Maximum (*Max*), Skewness (S) and Kurtosis (K) of *PM*<sub>10</sub> data at Martinique (MAR), Guadeloupe (GPE) and Puerto-Rico (PR) over 11 years, for low (October to April) and high (May–September) dust seasons.  $\overline{M}$ ,  $\sigma$ , *Min* and *Max* are in  $\mu g/m^3$  and N represents the data point number.

Period	Location	Ŵ	σ	Min	Max	S	к
Overall	MAR (N = 3,903)	26.52	17.65	2.0	200.2	2.12	9.98
	GPE (N = 3,503)	26.34	15.61	3.3	164.4	2.27	10.98
	PR (N = 3,718)	24.42	15.11	1.0	197.0	3.19	19.27
Low season	MAR (N = 2,271)	21.19	13.79	2.0	200.2	3.55	25.97
	GPE (N = $2,040$ )	21.80	11.49	3.3	164.4	3.86	29.60
	PR (N = 2,217)	19.90	9.44	1.0	143.0	4.98	49.59
High season	MAR (N = 1,632)	33.93	19.65	3.0	149.4	1.37	5.38
	GPE $(N = 1,463)$	32.67	18.19	4.0	157.2	1.44	6.10
	PR (N = 1,501)	31.09	18.96	7.0	197.0	2.27	11.55



and *K* values increase. However, we notice that MAR and GPE which are geographically very close have roughly the same values for  $\overline{M}$ , *S* and *K*. For PR, these values are different.

It is important to emphasize that  $PM_{10}$  concentrations in the Caribbean  $(24.42 \le \overline{M} \le 26.52 \ \mu g/m^3)$  are lower than those measured in Jawaharlal Nehru Port India 66.1 µg/m<sup>3</sup> (Gupta et al., 2004), Belgrade Serbia 68.3 μg/m<sup>3</sup> (Mijić et al., 2009), Nilai Malaysia 59.1 µg/m<sup>3</sup> (Sansuddin et al., 2011), Beijing China 145.1 µg/m<sup>3</sup> (Xi et al., 2013), Anatolia Turkey 78.0 µg/ m<sup>3</sup> (Ozel and Cakmakyapan, 2015) or Abadan Iran 186.1 µg/m<sup>3</sup> (Momtazan et al., 2018) to name a few. In addition, the standard deviation value computed (15.11  $\leq \sigma \leq$  17.65 µg/m<sup>3</sup>) is lower than those found in megalopolis as Anatolia Turkey (Ozel and Cakmakyapan, 2015), Abadan Iran (Momtazan et al., 2018), Nilai Malaysia (Sansuddin et al., 2011) and Beijing China (Xi et al., 2013) with respectively 26.0, 26.4, 28.6 and 91.4  $\mu\text{g/m}^3.$  This is due to the wide heterogeneity of  $PM_{10}$  sources in these large cities where anthropogenic pollution is high. Thus, if the standard deviation is high, so is the variability, indicating large concentrations (Plocoste et al., 2020a).

Moments of higher order, such as skewness (third order) and kurtosis (fourth order) are often estimated for a sharp characterization and therefore the highlighting of the Gaussianity of the considered process. Note that the third moment is zero for symmetric distributions. The kurtosis of a normal distribution is equal to 3. A positive kurtosis is therefore an indicator of the degree of intermittency. With  $2.12 \le S \le 3.19$  and  $9.98 \le K \le 19.27$ , the skewness and kurtosis are positive for all  $PM_{10}$  time series. For *S*, it means that the frequency distribution moves away from a normal distribution on the right with a larger right tail. As K > 3, this indicates that the values we get are greater than the peak of a Gaussian distribution (Dong et al., 2017). In other words, there is in  $PM_{10}$  data some values related to extreme events and there is an irregularity in the process of  $PM_{10}$  emissions.

### 4.1.2 Seasonal Analysis

To refine the analysis of  $PM_{10}$  concentrations behavior, a statistical analysis is performed according to African dust seasonality. In the literature, many studies have shown that the low dust season runs from October to April while the high dust season runs from May to September in the Caribbean area (Prospero et al., 2014; Velasco-Merino et al., 2018; Plocoste and Pavón-Domínguez, 2020b). As expected, Table 1 shows that  $PM_{10}$  average concentrations are higher in the high dust season. As observed in section 4.1.1, there is a decrease in the average from MAR to PR during that period which is not the case in the low season. During summer months, a large amount of sand travels from the African coast towards the Caribbean in a Saharan Air Layer at an altitude between 1 and 5 km height (Prospero and Carlson, 1972; Tsamalis et al., 2013) and at an average speed of 10 ms<sup>-1</sup> (Petit et al., 2005; Jury and Jiménez, 2021). Euphrasie-Clotilde et al. (2020) showed that ~ 84% of dusty air masses in the Caribbean area come directly from the East African coasts. Many studies have already shown that Barbados (see Figure 1) is the first Caribbean island impacted by dust outbreaks (Prospero et al., 1970; Chiapello et al., 2005; Zuidema et al., 2019). Dry deposition (gravitational settling) and wet deposition (precipitation and cloud sweeping) are the two main processes that will remove these mineral dust from the atmosphere (Schepanski, 2018; Plocoste et al., 2021a). In other words, the further one moves away from the African coast, the lower the concentrations of  $PM_{10}$  linked to mineral dust.



Due to insular context,  $PM_{10}$  are mainly composed by marine aerosols and anthropogenic pollution in the low dust season (Clergue et al., 2015; Rastelli et al., 2017). In other words, this season is more representative of local pollution and sea spray. Contrary to African dust (large scale source), marine aerosols and anthropogenic pollution (mesoscale and local scale) fluctuate less over time. Indeed, anthropogenic pollution is related to daily human activities (Plocoste et al., 2018) while marine aerosols are advected by the trade winds which blow continuously during the year (Plocoste and Pavón-Domínguez, 2020a). This is the reason why  $\sigma_{Low} < \sigma_{High}$ .

In Table 1, one can observe that the minimum values are higher for the high dust season. This is due to the fact that a residual amount of dust remains in the atmosphere due to the continuous alternation between African easterly waves and dust outbreaks during that period (Plocoste et al., 2021c). As regards the maximum values, the behavior is different between both seasons. In the high dust season, these values correspond to the same haze of sand that impacted the Caribbean basin from May 14 to 20, 2007 with daily peaks on the 14 in MAR (149.4 µg/ m<sup>3</sup>), 15 in GPE (157.2  $\mu$ g/m<sup>3</sup>) and 16 in PR (197.0  $\mu$ g/m<sup>3</sup>). This event seems special because although there is a daily lag between these peaks, the phenomenon increases in intensity instead of decreasing. One hypothesis would be the meeting between a mass of air coming from Africa and another one coming from cold higher latitudes, e.g., Alaska or Iceland, where main quantities of dust are transported southward and deposited in the North Atlantic (Prospero et al., 2012). This episode will be analyzed

more precisely in a future study. Contrary to PR, one can notice that the higher maximum values for MAR and GPE are in the low dust season. During that period, dust outbreaks from Africa are more sporadic but can occur. With 200.2  $\mu$ g/m<sup>3</sup> on 16 February 2007 and 143.0  $\mu$ g/m<sup>3</sup> on 28 April 2010, MAR and PR are the perfect example. For GPE, the 164.4  $\mu$ g/m<sup>3</sup> is due to the eruption of Soufrière volcano at Montserrat in 13 February 2010 (mesoscale source) (Plocoste and Calif, 2019). All these results show that dust outbreaks are not the only natural source that can generate high *PM*<sub>10</sub> concentrations in the Caribbean.

As regards the skewness and kurtosis, the same behavior is observed for both parameters, i.e.  $S_{High} < S_{Low}$  and  $K_{High} < K_{Low}$ . This may be due to the fact that African dust are recurrent in the high dust season (Plocoste et al., 2020a). During this period, days with  $PM_{10}$  concentrations exceeding the 50 µg/m<sup>3</sup> recommended by the World Health Organization (2006) are more frequent. According to Huang et al. (2010), there is an average of ~6 dust outbreak days per month during the summer months. Whatever the season, it may be noted that *S* and *K* values are higher for PR. The authors believe this is mainly due to the location of  $PM_{10}$  sensor. MAR and GPE seem to be undergoing the same plume of  $PM_{10}$  concentrations. To confirm this assumption, an analysis will have to be carried out with  $PM_{10}$  sampled at high frequency.

### 4.2 Distribution Analysis

To sharply describe the statistical information contained in a dataset, the probability density function (PDF) is a robust tool. It



(2020) classification: NWAP, SWAP, NEAP, SA, North.

will allow to exhibit the entire range, mean and probability of occurrences of  $PM_{10}$  data. Figure 4 presents the PDFs of  $PM_{10}$  time series at MAR, GPE and PR over 11 years. Overall, one

notice that all the PDFs show the same pattern. In "Area 1," the first maximum probability value (peak of the PDF) is close between each island with 17.0  $\mu g/m^3$  at MAR, 17.2  $\mu g/m^3$  at



classification: NWAP, SWAP, NEAP, SA, North.

PR and 19.2  $\mu$ g/m<sup>3</sup> at GPE. The higher value in GPE is due to the fact that the measurements are taken very close to the largest industrial area of the island (Plocoste et al., 2018). Then, all the

distributions converge towards a same point at  $28\,\mu g/m^3.$  In "Area 2," it is important to underline that MAR and GPE curves are almost similar while that of PR diverges. The



second maximum probability value is  $46.2 \ \mu g/m^3$  at PR and  $53.2 \ \mu g/m^3$  for MAR and GPE. Unlike the first peaks where the values seem homogeneous, the second peaks present a greater shift between PR and MAR-GPE. Intuitively, the first peak seems more representative of  $PM_{10}$  from local and mesoscale sources while the second seems strongly related to  $PM_{10}$  from large scale source. These results confirm that the further away from the African coast, the more the intensity of dusts outbreaks seems to decrease. Consequently, the authors believe that  $28 \ \mu g/m^3$  is the detection threshold between local sources and large scale sources, i.e., African dust detection threshold.

**Figure 5** shows the distribution plot of  $PM_{10}$  concentrations by year. For some years, the same result as Figure 4 seems to be observed while for the others, the convergence point of the distributions, when it exists, is before or after the estimated threshold over 11 years. From one year to another, one can notice a strong heterogeneity in PM10 distribution for each island. This behavior is mainly due to the activation of dust sources in Africa. In summer, many African dust source in the Sahara and Sahel region become more active (Schepanski et al., 2007; Bou Karam et al., 2008; Kim et al., 2017). These activations are related to the development and movement of African easterly coupled with extra-tropical disturbances. waves The modifications of terrain properties in these source regions (vegetation cover or land use) combined with climate processes that affect them will act to modulate transport to the Caribbean area (Ginoux et al., 2012; Prospero et al., 2014). Indeed,  $PM_{10}$  inter-annual variability is also connected to dust removal and deposition mechanisms (e.g., changes in precipitation) (Gavrouzou et al., 2021). Drought years in the global dust belt zone are associated with a high presence of dust in the atmosphere due to reduced wet deposition (Dey and Di Girolamo, 2010; Prospero et al., 2021).

### 4.3 Air Mass Back Trajectory

In order to validate the threshold previously determined in section 4.2, the daily HYSPLIT back trajectories are generated for dusty days  $(PM_{10} \ge 28 \,\mu\text{g/m}^3)$  and non-dusty days  $(PM_{10} <$ 28 µg/m<sup>3</sup>) from 2006 to 2016 according to Euphrasie-Clotilde et al. (2020) classification: North-West Atlantic Path (NWAP), South-West Atlantic Path (SWAP), North-Est Atlantic Path (NEAP), South America (SA) and North. NWAP and SWAP are air masses coming directly from the dust belt while NEAP is a circulation originating from the North of the United States which rotates nearby the African coasts. The seasonal behavior of these paths has been widely described in Euphrasie-Clotilde et al. (2020) works. On Figures 6, 7, one can observe that these paths (NWAP + SWAP + NEAP) correspond to the main air masses routes moving towards the Caribbean with on average 89 and 80% respectively for dusty days and non-dusty days. For both periods, the number of back trajectories passing near the eastern part of SA is of the same order of magnitude. Concerning the air masses coming directly from the North of the United States, these cases are at least 3 times greater during non-dusty days. In Figure 6, it is important to emphasize that  $PM_{10}$  average concentrations related to the back trajectories are well above the 28  $\mu g/m^3$  threshold estimated. Furthermore,  $PM_{10}$  standard deviations of Figure 6 are higher than those of Figure 7. All these results highlight that dust sources activation in Africa is a key process allowing the increase of  $PM_{10}$  concentrations in the Caribbean area. Knowing that  $PM_{10}$  emissions from local sources are frequently below this threshold (see low season average in Table 1), these results confirm that  $28 \,\mu g/m^3$  is a transition value between local sources and large scale sources as dust outbreaks. This threshold is lower than the  $50 \,\mu\text{g/m}^3$ recommended by the World Health Organization for an average of 24 h (World Health Organization, 2006). It refines the threshold of  $35 \,\mu\text{g/m}^3$  previously found in the literature to detect dust events (Euphrasie-Clotilde et al., 2020).

**TABLE 2** Statistical parameters (Mean ( $\overline{M}$ ), Standard deviation ( $\sigma$ ), Minimum (Min), Maximum (Max)) of  $PM_{10}$  residual at Martinique (MAR), Guadeloupe (GPE) and Puerto-Rico (PR) over 6 years with CEEMDAN and ICEEMDAN methods. For each island the data point number N is 2,190.

Method	Location	Μ̄ (μg/m³)	σ (μg/m³)	<i>Min</i> (μg/m³)	<i>Max</i> (μg/m <sup>3</sup> )
CEEMDAN	MAR	27.32	1.62	25.31	30.14
	GPE	28.10	1.26	26.43	30.20
	PR	26.83	2.18	24.47	30.52
ICEEMDAN	MAR	27.40	1.41	25.41	29.61
	GPE	27.62	0.55	26.35	28.29
	PR	25.86	2.55	22.87	31.39



Guadeloupe (GPE) and **(C)** Puerto-Rico (PR) by year. The horizontal pi dash-dot line represents the  $PM_{10}$  statistical threshold estimated over 11 years at 28 µg/m<sup>3</sup>.



**(B)** Guadeloupe (GPE) and **(C)** Puerto-Rico (PR) by year. The horizontal purple dash-dot line represents the  $PM_{10}$  statistical threshold estimated over 11 years at 28 µg/m<sup>3</sup>.

# 4.4 Multiscale Analysis

After studying the detection threshold between the local sources and large scale sources in a statistical way, we decide to analyze this threshold dynamically. Unlike statistical analysis, temporal analysis requires complete data to not compromise the dynamics of the signal, i.e. not modify the physical meaning of the studied parameter. Here, the difficulty was to have the maximum of complete and continuous data simultaneously for the three islands. This is the reason why 6 years were retained from 2006 to 2011 for this investigation.

In CEEMDAN and ICEEMDAN framework, the first mode characterizes the fast fluctuations while the last mode characterizes the slowest fluctuations (Torres et al., 2011; Colominas et al., 2014). The trend of the data is given by the residual part of the decomposition process (Hu et al., 2013). To perform the CEEMDAN and ICEEMDAN methods, the number of realizations was fixed at 300 and the Gaussian noise amplitude was set to 0.15 (Plocoste, 2022b). For each island,  $PM_{10}$  time series were decomposed into 10 IMFs and one residue. In order to assess the dynamic behavior of the  $PM_{10}$  detection threshold between local sources and large scale sources, the authors focused on the residual part of both approaches by comparing them to the statistical threshold estimated in **section 4.2**.

Figure 8 illustrates the residual from CEEMDAN and ICEEMDAN between 2006 and 2011. One can notice a heterogeneity in the residual behavior which seems to stabilize over the period of 6 years. Table 2 presents the statistical parameters for each method. Whatever approach is used, both methods converge towards the same result. Indeed, the average values of the residuals for each island are close to the statistical threshold of 28  $\mu$ g/m<sup>3</sup>. The difference between the minimum and maximum values is smaller for MAR and GPE. As regards the standard deviation, the higher values is for PR. The authors assume that its greater distance from the African coasts and the location of the sensor may play a major role in this behavior for this location. In other words, the dynamical results are in agreement with the statistical results. Indeed, even with 6 years of data, the dynamic approach confirms the statistical threshold estimated with 11 years of data.

Figures 9, 10 show the yearly residual part of  $PM_{10}$  times for each island respectively with CEEMDAN and ICEEMDAN methods. In both Figures one can observe that for some years the residual follows a seasonal cycle while for the others the residual can be almost constant. Overall, the residuals in Figure 10 seem closer to the statistical threshold of 28 µg/ m<sup>3</sup>. It is important to recall that ICEEMDAN is the improved version of CEEMDAN (Colominas et al., 2014). For those following the seasonal cycle, the maximum values are generally between June and August, i.e., the period when the African dust sources are more active (Zuidema et al., 2019; Euphrasie-Clotilde et al., 2021; Gavrouzou et al., 2021). We must emphasize that the cases where the residue does not present seasonality are not anomalies. As its definition indicates, the residual gives the overall trend of  $PM_{10}$  time series, i.e., the background atmosphere. As expected, between MAR-GPE-PR, PR is the location where the seasonality is least marked in **Figure 10** due to the arrival of air mass in Cataño from San Juan, i.e., the capital and the most populated area.

# **5 CONCLUSION**

It is well known that air quality of the Caribbean basin is frequently deteriorated by African dust. Thus, vulnerable populations (the elderly, asthmatics or children) are often exposed to particulate pollution. That is why the aim of this study was to assess the  $PM_{10}$  detection threshold between local sources and large scale sources. To carry out this investigation,  $PM_{10}$  time series from Martinique (MAR), Guadeloupe (GPE) and Puerto-Rico (PR) islands were used.

The statistical analysis showed that the distance from the African coast is a key element for the distribution of  $PM_{10}$  concentrations during the high dust season in summer. Indeed, MAR which is the closest island has the highest  $PM_{10}$  average and the lowest kurtosis. Conversely, PR which is the furthest away has the lowest  $PM_{10}$  average and the highest kurtosis. During this period, high  $PM_{10}$  concentrations are mainly related to large scale sources. In the low dust season,  $PM_{10}$  concentrations are mainly related to local sources. Using the PDF analysis, a statistical threshold was estimated at 28 µg/m<sup>3</sup>. HYSPLIT back trajectories analysis confirmed this threshold. The authors assume that this value corresponds to the  $PM_{10}$  detection threshold between local sources and large scale sources.

In order to investigate this threshold in a dynamical way, the residual part of two multiscale decomposition methods was analyzed. CEEMDAN and ICEEMDAN results showed that the behavior of the residuals of each island over time remains close to the  $PM_{10}$  statistical threshold. Consequently, the dynamical results validate the  $PM_{10}$  detection threshold found with the statistical analysis.

To conclude, the results of this study are relevant because they define a new detection threshold for dust outbreaks in the Caribbean from ground measurements, i.e., after dry and wet deposition of mineral dust. These phenomena being events that last several days, this new threshold will make it possible to better determine the beginning and the end of a dusty episode in the atmospheric boundary layer. The competent authorities will therefore be able to take adequate measures to protect vulnerable populations. According to the European legislation,  $28 \,\mu g/m^3$  corresponds to "medium" air quality (from 21 to  $40 \,\mu g/m^3$ ). In view of the high rate of asthma in this area, the authors believe that this new threshold is more related to "poor" air quality, which actually corresponds to concentrations of  $41-50 \,\mu g/m^3$  in European legislation. In this study, the impact of weather conditions on  $PM_{10}$  concentrations were not taken into account. This will be the aim of a future study.

# DATA AVAILABILITY STATEMENT

The data analyzed in this study is subject to the following licenses/ restrictions: The data presented are available on request from the corresponding author. The data are not publicly available due to privacy or ethical reasons. Requests to access these datasets should be directed to thomas.plocoste@karusphere.com.

# **AUTHOR CONTRIBUTIONS**

TP: Writing-original draft, Conceptualization, Methodology, Software, Resources, Formal analysis, Data Curation, Supervision, Visualization, Validation, Investigation, Project administration, Writing-review and editing. LE-C: Investigation, Writing-original draft, Data Curation. RC: Supervision, Validation, Writing-review and editing. F-NB: Data Curation, Formal analysis.

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