Meteorol Atmos Phys (2014) 124:83–96
DOI 10.1007/s00703-013-0304-2

ORIGINAL PAPER

A new method of quantifying aerosol concentrations in atmosphere

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Received: 7 April 2012 / Accepted: 25 November 2013 / Published online: 14 December 2013

Abstract Africa is one of the sources of biomass burning emissions. It is estimated that about 6 million tons of fuel per day is consumed in the southern hemisphere. Biomass burning has an important contribution on aerosol particle concentrations in the atmosphere. Efforts have been made to conduct research in Gaborone to monitor the concentration of atmospheric aerosol particles. These studies were mainly confined to measurement of concentration of aerosol particles and establishing a relation with determinants such as carbon dioxide concentration, biomass burning, and precipitation among others. However, very little seems to have been done in relating the empirical data to levels of aerosol concentrations through a mathematical model. In this paper an objective criterion of classifying levels of aerosol concentrations in terms of their severity is provided. A mathematical model for severity levels is built. Furthermore, two indices, namely, an index of dispersion when applied to the observed annual data indicated that intensity of atmospheric aerosol are on increase in the city of Gaborone, Botswana, and an index of drift which establishes that aerosol severity states showed larger drift during the year 2006–2007 than in the year 2007–2008.

Responsible editor: S. Trini Castelli.

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1 Introduction

Precipitation is the process of transporting water from the atmosphere back to earth's surface. It links climate, weather and the global hydrological cycle. Atmospheric aerosol particles can contribute to climate change by absorbing and scattering solar and infrared radiation. It can indirectly influence climate by altering the properties of clouds (Lohmann and Feichter 2005). It can also collect other cloud droplets and lead to in cloud scavenging (Andronache et al. 2005). Removal of atmospheric aerosol particles during precipitation is the major way of sink of aerosol particles. The atmospheric particle concentration measurements were made over different parts of the world by various researchers (Wallace and Hobbs 1977; Verma and Jayaratne 2001; Hitchins et al. 2000). Many studies have been conducted to monitor the aerosol particles due to biomass burning. For example, Xuejiao et al. 2008 showed that the biomass burning in south East Asia has an important contribution on aerosol particle concentrations. A study in Southeast Asia (Ma et al. 2003) showed that biomass burning plume contributes approximately 35–40 % of the fine organic aerosol particle mass in the Pacific Ocean. The concentration of aerosol particles was observed to decrease during rain (Sisterson et al. 1985). The studies conducted in Gaborone, Botswana (Jayaratne and Verma 2001; Verma and Thomas 2007), also showed that there is an increase in atmospheric particle concentration during winter season due to biomass burning.

The various studies on aerosol particles were mainly confined to measurements of concentration and establishing a

possible relation with determinants such as carbon dioxide concentration, biomass burning and precipitation among others. All these studies in fact established that the amount of aerosols in the atmosphere increases when (1) biomass is burnt and/or (2) there is not precipitation. However, while these are partly foregone conclusions and do not strictly need to be demonstrated, but better quantified in any subsequent research studies. Therefore, the main purpose of this article is focused around a novel way of building a mathematical model given the levels of aerosol concentration measurements. A review of the literature suggests that very little seems to have been done in relating the empirical data to levels of aerosol concentrations through a mathematical model. In this paper, an objective criterion for defining levels of aerosol concentration measurements in terms of their severity is provided. A mathematical model and two indices that help to understand, for example, variations in weekly aerosol particle concentrations are constructed.

The study reported here was an ongoing research in the Department of Physics, University of Botswana. Botswana, as shown in the map in Fig. 1, is a land-locked country in southern Africa surrounded by Namibia to the west, South Africa to the east and south and Zambia and Zimbabwe to the north. The country lies between longitudes 20° and 30° east of Greenwich and between the latitudes 18° and 27° approximately south of the equator. It is approximately 500 km from the nearest coastline, to the southwest (Geographical info 1996).

Botswana is hot and dry for much of the year. The rainy season is in summer, which brings high temperature and is between November and March, with the peak in January and February. Rain is unpredictable and regional, sometimes followed by sunshine. The mean annual rainfall is about 650 mm in the north and 250 mm in extreme south (Botswana geographical info 1996). The winter is between May and August and days are sunny and warm; but night temperature can drop below freezing point in some places. According to Botswana Meteorological Services, annual rainfall in Gaborone during the years 2006, 2007 and 2008 were, respectively, 506, 348 and 475 mm.

The primary objectives of this research article are as follows:

1. To propose a new paradigm, namely, severity states for atmospheric aerosol concentrations based on weekly mean measurements.
2. To build a probability model that best explains the variations in the severity states for the weekly data.
3. To propose two new absolute indices namely; (a) a drift index, that helps to measure overall variations in terms of frequency of severity states and (b) a dispersion index that indicates how far the weekly atmospheric aerosol concentrations are deviating from the normal threshold level.
4. To (a) classify weekly mean atmospheric aerosol concentrations based on the new paradigm (b) compute the likelihood of different states of severity (c) compute drift and dispersion indices using the daily aerosol concentration measurements observed during the experimental period from September 2006 to August 2008.

Fig. 1 Map of Botswana showing the experimental site

5. To interpret the significance of levels of aerosol concentration measurements observed during the experimental period from September 2006 to August 2008 based on the new indices proposed.

2 Data and methodology

The atmospheric particle concentrations were monitored in the Physics Department at the University of Botswana in Gaborone. The measurements were made using automatic laser scattering particle counters from Rion model KC-01 which detects particles larger than or equal to 0.3 μm diameter and separates them into 6 categories (≥0.3, ≥0.5, ≥0.7, ≥1.0, ≥2.0 and ≥5.0 μm). For the sake of analysis, we classify these six categories into PM 2.5(particles of size in between 0.3 and 2 μm) and PM 10 (particles of size ≥5 μm). The air sample was drawn through plastic tubes of diameter 5 mm and of length 1.8 m from outside and about 10 m above the ground. After running the instrument for certain period of time the error in the measurement were calculated and error factor was applied to minimize the error due to the piping and other effects. The humidity and temperature at all times of observations were also recorded. The readings were taken at 12 noon every day. This time is chosen assuming that, at that time of the day atmosphere was relatively calm. The instrument counts the particle concentrations, average and displays the data. The rain fall data was collected from Botswana Meteorological Services in Gaborone. The study period consisted of 91 weeks from September 2006 to August 2008. In each week measurements were taken on successive days. The frequency of sampling in each week varied from two to 7 days depending on the availability of experimental, human and capital resources. From Table 1, it is evident that, for the year September 2006–August 2007, the weekly mean aerosol particle concentration measurements vary between 13.86 particles cm^{-3} and 164.15 particles cm^{-3} with a weekly mean of 68.78 particles cm^{-3} while for the year 2007–2008, the corresponding measurements are respectively, 33.46, 138.28 and 75.65 particles cm^{-3}. It may be pointed out that smaller mean measurements could be due to presence of scavenging factors such as precipitation, while larger mean measurements could be due to factors such as biomass burning, secondary aerosol formation from combustion due to NO_x and SO_2. Further, large variations in weekly means are indicative of high drift in levels of aerosol concentrations.

In general, significance of the aerosol concentration measurements may be captured by traditional statistical parameters like the standard deviation and skewness of the distribution. However, at times these may fail to explain the status of atmospheric pollution or other aerosol concentration conditions. In Table 1, we display summary statistics for PM 2.5 and PM 10(those inside the brackets) for the years 2006–2007. For example, it is seen that for PM 2.5, the variance and skewness of the distribution of weekly measurements during the years 2006–2007 and 2007–2008 are respectively 1,340.81, 0.93 and 888.65 and 0.67. These conventional summary indices indicate that year 2006–2007 showed more weekly variations, and also weekly means are more distorted to the right of the annual mean measurement than year 2007–2008. A similar pattern is seen with respect to PM 10 concentration. One of the demerits of these conventional measures is that they fail to track part of the data which contribute to the pattern portrayed by these indices. Such information may be needed to account for the presence of high or low aerosol concentration measurements during the study period. Furthermore, it may be desirable to have estimates of proportion of weeks during the study period that would result in different status of atmospheric conditions. Therefore these considerations would require construction of new indices that would better explain the status of atmospheric conditions than conventional indices.

As the atmospheric aerosol particles are known to influence climate adversely, it may be of interest to study the weekly variations in an objective way. A natural question in atmospheric data analysis, apart from establishing a relationship between aerosol concentration measurements and factors responsible for it, is how best one can possibly quantify levels of aerosol concentration measurements to render comparisons within and across time period of study. The quantitative analysis should be grounded on ideas beyond the narrative interpretations of tables and graphs. A naive approach would be to categorize weekly variations into certain classes, based on percentage increase or decrease in the corresponding weekly means, with respect to a reference time. However, this categorization may appear subjective as percentage cut-off points may have been arbitrarily defined, and more over, such

Table 1 Summary statistics: weekly PM 2.5 (PM 10) aerosol concentrations (cm^{-3})

Year	Weeks	Minimum	Maximum	Mean	Variance	Skewness
2006–2007	42	13.74 (0.013)	164.00 (0.249)	67.77 (0.071)	1,340.81 (0.003)	0.93 (1.488)
2007–2008	49	32.49 (0.013)	138.13 (0.195)	72.22 (0.069)	888.65 (0.002)	0.67 (0.929)

categorization is not data driven. An objective way of categorizing weekly variations is to adopt a rule that is defined by the underlying parameters of the sampling distribution of the weekly averages; for example, the mean and standard deviation. The parameters, mean and standard deviation, are chosen for simplicity, as it is well known that mean measures the central tendency, while spread of the underlying distribution of the data is measured by the standard deviation. Moreover, the interval (mean ± 3 standard error) covers almost all the data. For example, when the underlying sampling distribution is Gaussian, as may be the case in several atmospheric data, the intervals (mean ± 3 standard error), (mean ± 2 standard error), (mean ± standard error), are respectively known to cover approximately 97, 95 and 68 % of the observations in a given data. The notion of levels of aerosol concentrations is linked to these severity intervals. These intervals inter alia may be used to define the severity or criticality of aerosol concentrations characterizing how far individual observations are spread from the center of the data. This principle is essentially used in this study to define classes that categorize weekly atmospheric aerosol concentration measurements.

3 Severity states for aerosol particle concentrations

Suppose that there is a time series data of daily aerosol particle concentration measurements, $P_t, t = 1, \ldots, N$, where 'N' is the length of the study period in days. Here 't' refers to the day. The total number of weeks during the study period shall be denoted by 'n' and the number of days in a week 'w' by 'n_w' (Usually n_w varies between 2 and 7, the number of days in a typical week during which particle concentrations were monitored). Then the weekly mean and variance of aerosol particle concentration are respectively given by

$$\overline{P}_w = \frac{1}{n_w}\sum_{t=1}^{n_w} P_t, w = 1, \ldots, n, \tag{1}$$

and

$$s_w^2 = \frac{1}{(n_w - 1)}\sum_{t=1}^{n_w} (P_t - \overline{P}_w)^2. \tag{2}$$

Assuming that the conditions related to weekly mean aerosol particle concentrations can be classified into seven states depending on their severity, the following seven states are accordingly defined: S_1: Absolutely safe state, S_2: Safe state, S_3: sub-normal state, S_4: normal state, S_5: sub-critical state, S_6: critical state, S_7: extremely critical state. Further, if it is assumed that $\overline{P}_w$'s comes from a population with finite mean μ and finite variance σ^2, then $E(\overline{P}_w) = \mu$ and $\mathrm{Var}(\overline{P}_w) = \frac{\sigma^2}{n_w}$. Then one may use the standardized statistic $Z = \sqrt{n_w}\left(\frac{\overline{P}_w - \mu}{\sigma}\right)$ to construct appropriate classification rules for the severity of states. In Z, the unknown population mean μ is replaced by its unbiased estimator $\overline{P}_w$ and the unknown population variance σ^2 by its unbiased estimator s_w^2, respectively given by (1) and (2) above. Thus, the severity of atmospheric aerosol particle concentrations in weekly data may be classified into seven states $S_j, j = 1, \ldots, 7$ as follows. For a current week $(w+1), w = 1, \ldots, n-1$ the aerosol particle concentration measurement is said to belong to

$$\left.\begin{array}{l}
\text{(i)}\quad \text{State}\quad S_1, \text{ if } \overline{P}_{w+1} < \overline{P}_w - 3\dfrac{S_w}{\sqrt{n_w}} \\
\text{(ii)}\quad \text{State}\quad S_2, \text{ if } \overline{P}_w - 3\dfrac{S_w}{\sqrt{n_w}} \le \overline{P}_{w+1} < \overline{P}_w - 2\dfrac{S_w}{\sqrt{n_w}} \\
\text{(iii)}\quad \text{State}\quad S_3, \text{ if } \overline{P}_w - 2\dfrac{S_w}{\sqrt{n_w}} \le \overline{P}_{w+1} < \overline{P}_w - \dfrac{S_w}{\sqrt{n_w}} \\
\text{(iv)}\quad \text{State}\quad S_4, \text{ if } \overline{P}_w - \dfrac{S_w}{\sqrt{n_w}} \le \overline{P}_{w+1} < \overline{P}_w + \dfrac{S_w}{\sqrt{n_w}} \\
\text{(v)}\quad \text{State}\quad S_5, \text{ if } \overline{P}_w + \dfrac{S_w}{\sqrt{n_w}} \le \overline{P}_{w+1} < \overline{P}_w + 2\dfrac{S_w}{\sqrt{n_w}} \\
\text{(vi)}\quad \text{State}\quad S_6, \text{ if } \overline{P}_w + 2\dfrac{S_w}{\sqrt{n_w}} \le \overline{P}_{w+1} < \overline{P}_w + 3\dfrac{S_w}{\sqrt{n_w}} \\
\text{(vii)}\quad \text{State}\quad S_7, \text{ if } \overline{P}_{w+1} \ge \overline{P}_w + 3\dfrac{S_w}{\sqrt{n_w}}
\end{array}\right\} \tag{3}$$

In the above stated classification rule, the current week's mean particle concentration is compared with previous week's mean particle concentration plus or minus a multiplier of the standard error of that week's mean to decide to which severity state the current week's mean belongs. The multipliers ±3 and ±2 of $\frac{s_w}{\sqrt{n_w}}$ suggested here is quite appropriate in the sense that the intervals formed with these multipliers can be shown to capture almost all variations that exist in the weekly means. For example, when the underlying distribution of weekly mean is Gaussian, the interval $\left(\overline{P}_w - \frac{3}{\sqrt{n_w}}s_w, \overline{P}_w + \frac{3}{\sqrt{n_w}}s_w\right)$ is known to cover approximately 99.73 % of the variations in the weekly means (See for example, Stuart and Ord 1994). In (3) above, the states S_1, S_2 and S_3 prior to the normal threshold state S_4 refer to decreased levels of aerosol concentrations possibly due to scavenging activities such as precipitation, emission control mechanisms etc.; while the states S_5, S_6 and S_7 beyond the normal threshold state S_4 refer to increased levels of aerosol concentrations possibly due to activities such as biomass burning, industrial emissions, etc.

4 A multinomial probability model for severity states

An empirical model building exercise for severity states that characterize weekly variations in mean aerosol particle concentration measurements can be approached from the classical probability point of view in that one may model different states of severity based on a certain probability distribution. Here the severity states refer to the quantum jumps in weekly means during the study period. In general, suppose that there are k mutually exclusive and exhaustive states, say S_1, …, S_k to which the weekly mean $\overline{P}_w$ can be assigned based on a certain classification rule, say for example, rule (3) and let $P_r(S_j) = \theta_j$ denote the probability that a typical weekly mean $\overline{P}_w$ belongs to the state j for $j = 1, \ldots, k$. We let $n_j, \quad j = 1, \ldots, k$ to denote the number of occurrences of the state S_j in an independent sequence of the phenomena observed say, for $n = \sum_{j=1}^{k} n_j$ weeks. Then the vector $\mathbf{n} = (n_1, \ldots, n_k)$ follows a multinomial distribution (See for example, Stuart and Ord (1994) with the joint probability mass function (p.m.f.) given by

$$p(n_1, n_2, \ldots, n_k) = \prod_{j=1}^{k} \frac{n!}{n_j!} \theta_j^{n_j}, \tag{4}$$

where, $0 < \theta_j < 1, \sum_{j=1}^{k} \theta_j = 1, n = \sum_{j=1}^{k} n_j$.

The validity of the assumed multinomial model (4) rests on the assumption that the components in the vector of weekly means $\overline{P}_w$, $w = 1, \ldots, n$ are independent. This can be demonstrated by carrying out a test of randomness based on the total number of runs R in an ordered sequence of weekly means of aerosol concentration measurements. The weekly means data may be classified into a dichotomous sequence according as each observation is above or below some fixed number, often the calculated sample median or mean. Let n_1, n_2 respectively denote the number of observations below and above the sample median, where $n_1 + n_2 = n$. Then for $n_1 > 12$ and $n_2 > 12$, the two-sided run test consists in rejecting the null hypothesis of independence versus hypothesis of non-randomness, if

$$|Z| = \left| \frac{R + 0.5 - (1 + 2n_1n_2/n)}{\sqrt{2n_1n_2(2n_1n_2 - n)/n^2(n-1)}} \right| \geq z_{\alpha/2}, \tag{5}$$

where, $z_{\alpha/2}$ is the upper $\alpha/2$ percentile of a standard normal distribution (See for example, Gibbons and Chakraborti (2003).

In general, the parameters θ_j's in the model given by (4) are unknown and can be estimated by their empirical estimates

$$\widehat{\theta}_j = \frac{n_j}{n}, \quad j = 1, \ldots, k. \tag{6}$$

It may be pointed out that θ_j's are in fact the unrestricted maximum likelihood estimators of $\theta_j, j = 1, \ldots, k$. For the aerosol particle concentration data, the multinomial model that best describes likelihood of different severity states is given by

$$p(n_1, \ldots, n_7) = \frac{n!}{n_1!n_2!n_3!n_4!n_5!n_6!n_7!} \theta_1^{n_1} \theta_2^{n_2} \theta_3^{n_3} \theta_4^{n_4} \theta_5^{n_5} \theta_6^{n_6} \theta_7^{n_7} \tag{7}$$

for $0 < \theta_j < 1, j = 1, \ldots, 7, \theta_7 = 1 - (\theta_1 + \cdots + \theta_6), n = \sum_{j=1}^{7} n_j$.

Given the data on mean weekly aerosol particle concentrations, using the classification rule (3) we can obtain n_j's $j = 1, \ldots, 7$ and then estimate the probabilities using (6). These probabilities may be used to interpret the likelihood of different states of severity in the long run.

5 Indices of drift and dispersion for severity states

One may use the multinomial model given by (7) to propose an index of drift for aerosol particle concentrations. It is but natural to assume that if the aerosol particle concentrations are subject to significant cause variations; like biomass burning, industrial emissions, vehicular emissions, precipitation and so on, then the severity states may tend to be volatile from 1 week to another. On the other hand, if aerosol particle concentrations are subject to only random chance fluctuations, then one would expect a symmetric multinomial model with $\theta_1 = \theta_7, \theta_2 = \theta_6$ and $\theta_3 = \theta_5$ in (7) above, given by

$$p(n_1, \ldots, n_7) = \frac{n!}{n_1!n_2!n_3!n_4!n_5!n_6!n_7!} \theta_1^{n_1+n_7} \theta_2^{n_2+n_6} \theta_3^{n_3+n_5} \theta_4^{n_4} \tag{8}$$

for $0 < \theta_j < 1, \quad j = 1, \ldots, 4, \quad \theta_4 = 1 - 2(\theta_1 + \theta_2 + \theta_3)$, $n + n_4 = \sum_{j=1}^{4} (n_j + n_{8-j})$.

Ideally, in a symmetric multinomial model, most of the weekly variations in aerosol particle concentrations will be spread around the state S_4 in equal measure. As a measure of such a drift, a suitable index is proposed below. As before, let $n_j, \quad j = 1, \ldots, 7$ denote the number of occurrences of the state S_j in an independent sequence of the phenomena observed say, for $n = \sum_{j=1}^{k} n_j$ weeks. Suppose $E(n_j)$ denotes the expected number of occurrences of the state S_j under the hypothesis of symmetric multinomial model (8). Then it can be shown that the maximum likelihood estimators of θ_j's in the model (8) are given by

$$\widehat{\theta}_j = \frac{n_j + n_{8-j}}{2n}, \quad j = 1, \ldots, 4. \tag{9}$$

Thus an estimate of $E(n_j) = E(n_{8-j})$ can be obtained as

$$\widehat{n}_j = n\widehat{\theta}_j = \frac{(n_j + n_{8-j})}{2}, \quad j = 1, \ldots, 4. \tag{10}$$

It is reasonable to expect that under the hypothesis of symmetric multinomial model, the observed and the expected number of occurrences of the state S_j must be more or less the same, and therefore a suitable function of their difference would reflect the magnitude of drift. Accordingly, it is appropriate to consider an index of drift given by

$$I_{\mathrm{D}} = \sum_{j=1}^{7} (n_j - \widehat{n}_j)^2 = \frac{1}{2}\sum_{j=1}^{7} (n_j - \widehat{n}_{8-j})^2. \tag{11}$$

It is easily seen that when the severity states are symmetric, $I_{\mathrm{D}} = 0$; otherwise non-zero. Large values of $\mathrm{I_D}$ suggest that drifts in severity states are high. It is pointed out that the index of drift can be used in a micro analysis, for example, to ascertain how the weekly means of aerosol concentration measurements vary in a given unit of time; say a year or two.

Finally, given the severity state of each week, one can propose a measure or an index of dispersion which may be used to compare severity of aerosol particle concentrations across different years. The index of dispersion measures the spread of the severity states around the normal state. As before, let n denote the number of weeks in a year and $t_w, w = 1, \ldots, n$ be the value of the severity state corresponding to the week w. Note that t_w will take one of the values from 1 to 7. For example, if $t_1 = 2$; for the first week, then the severity state is '2', if $t_2 = 6$; for the second week, then the severity state is '6' and so on. Then an index of dispersion is given by

$$I_{\mathrm{d}} = \frac{1}{n}\sum_{w=1}^{n} (t_w - 4)^2 \tag{12}$$

It is seen that when weekly aerosol particle concentration measurements are more or less deterministic, then for each week, $\mathrm{S_4}$ will be the severity state, in which case $t_w = 4$ for all w and therefore, $I_{\mathrm{d}} = 0$. On the other hand if weekly means show swings on either side of the normal threshold state $\mathrm{S_4}$, I_{d} will be significantly different from 0. In the extreme case, it can be easily shown that the dispersion index I_{d} will be equal to 9. Thus, the greater the values of dispersion index, more dispersed are the weekly states from the normal state $\mathrm{S_4}$. These considerations precisely constitute the rationale behind the measure I_{d} proposed here and in particular given two or more series of aerosol particle concentration measurements one can compare them in terms of the index I_{d}. It is to be noted that drift and dispersion indices are defined in order to point up the symmetry of the severity states, since it is expected that in case of strong pollution events the status of the distribution moves towards the high-level states, and their dispersion. Further, it is to be noted that these measures have the statistical properties such as location and scale invariance.

6 Results and discussion

Figures 2 and 3 show the weekly means and standard deviations of PM 2.5 and PM 10 particle concentrations during the experimental period September 2006–August 2008. It is evident that both weekly means and standard deviations display significant variations from 1 week to another throughout the study period. But, these descriptive statistics alone do not seem to throw any light on the impact of aerosol concentrations over the time span of the study. Nonetheless, the magnitude of variations is a critical factor to be accounted for in any study related to assessing the impact of aerosol concentrations.

On a different note, for the purpose of the comparisons, the recorded measurements were grouped into three distinct non overlapping seasons namely dry season (April, September and October), rainy season (November, December, January, February and March) and winter season (May, June, July and August). This classification of seasons is consistent with the practice followed by Botswana Meteorological Services. However, based on the actual rain fall data, those weeks which recorded rain fall in non-rainy seasons were also included in the rainy season in this study. Thus, for the dry season, observations were obtained for 19 weeks, for winter season for 32 weeks and for rainy season for 40 weeks. The corresponding weeks of three seasons were identified among the total study period of

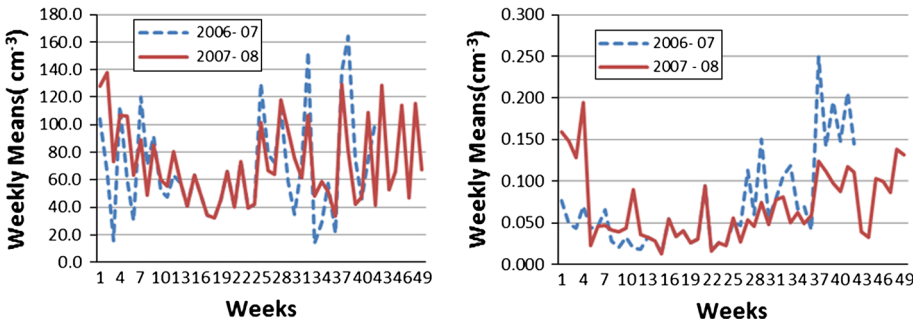

Fig. 2 Weekly means of PM 2.5 and PM 10 concentrations

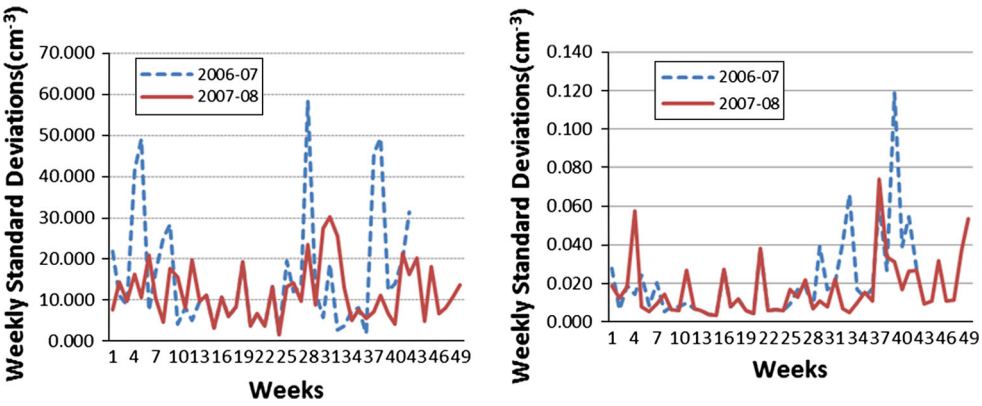

Fig. 3 Weekly standard deviations of PM 2.5 and PM 10 concentrations

91 weeks and the season-wise weekly means and standard deviations were plotted. It was observed from the plotted figure that the highest concentration of particles of size $\geq$0.3 μm was in the winter season and the lowest concentration of particles was mainly in the rainy season. During the winter and dry seasons, when the rain fall is low, the particle counts of size $\geq$0.3 μm are comparatively higher than those during the rainy season for most of the weeks. The increase in particles count during the absence of rainfall could have been due to the increase in biomass burning for heating purposes during that time as June–July months are the coldest months of the year in Botswana. Biomass usage for heating and cooking is very common in Botswana. The biomass contain large concentrations of small particles that can be activated as cloud condensation nuclei, which increase cloud droplet concentrations, decrease cloud droplet sizes and hence tend to inhibit precipitation due to the lack of sufficient numbers of large drops.

As outlined in Sect. 2 above, the aerosol particles were measured according to their size and separated into 6 groups, namely, 0.3, 0.5, 0.7, 1.0, 2.0 and 5.0 μm. Figure 4 shows the particle size distribution (particle concentration per cc) for 2006–2007 and 2007–2008. It is seen that the particle concentration is greatest around 0.3 μm and lowest around 5 μm. Figure 5 shows typical number size distribution spectrum of aerosol particles larger than 0.3 μm. According to Junge's principle (see Wallace and Hobbs 1977) the slope of the number size distribution gives the value of 'v' at that place. Its value is normally below 4. For continental aerosols larger than 0.2 μm this value is closer to 3. The value of 'v' obtained is around 0.6 during the study period. This shows that the air quality in Gaborone is of good standard. Further, it is worthwhile to provide at least some statistics about size distribution of aerosol particles and these could be useful for climate studies and modeling. Figure 6 shows particle size distributions of weekly means for the year 2006–2007 and 2007–2008. It is seen that distribution corresponding to particle size 0.5, 0.7, 1.0 and 2.0 μm show peak fluctuations during the weeks 28–31 for the year 2006–2007. This tendency is consistent with the extremely cold climatic conditions during the winter months of May–June 2007 as reported by the Botswana Meteorological Services. Table 2 summarizes the findings of PM 2.5 and PM 10 size distribution analysis in nutshell. The summary statistics refer to the mean and variance of particle size distribution during the study period 2006–2007 and 2007–2008. To establish whether PM 2.5 and PM 10 particle size distributions are homogeneous over the two study periods with respect to variance, we carry out an F test of homogeneity for variances. The findings reported in Table 2 suggest that the magnitude of variations in PM 10 weekly means is significant while it is not so for PM 2.5 weekly means. Next, to see if the weekly means of particle size distributions are the same for the two periods, we carry out appropriate two-sample t test and the findings reported in Table 2 suggest that the differences in weekly means are insignificant. Thus, the approach to compare weekly particle size distributions has been primarily based on the well-known techniques such as summary statistics and test of hypotheses (see Stuart and Ord 1994). However, the focus of this article is to suggest an alternative technique; which is supposedly a new line of approach to analyze particle concentrations. To demonstrate the new method we prefer to focus on PM 2.5 and PM 10 particle concentrations of aerosol rather than on different particle sizes captured in the experiment. The findings of the new line of analysis are dealt in the next few paragraphs.

To validate the multinomial model suggested here, we first carry out a test of randomness for the observed sequence of PM 2.5 and PM 10 mean concentration measurements as reported in the Table 3 for the years 2006–2007 and 2007–2008. Following Gibbons and Chakraborti (2003), for PM 2.5 size, the application of the formula (5) and straight forward calculations lead to $Z = -1.5$ and $Z = 0.58$ for the 2 years respectively. The corresponding values for PM 10 are respectively, $Z = -2.03$ and $Z = -1.44$. The hypothesis of independence of weekly means is accepted at 5 % level of significance; except that for PM 10 size, there, appears to be a

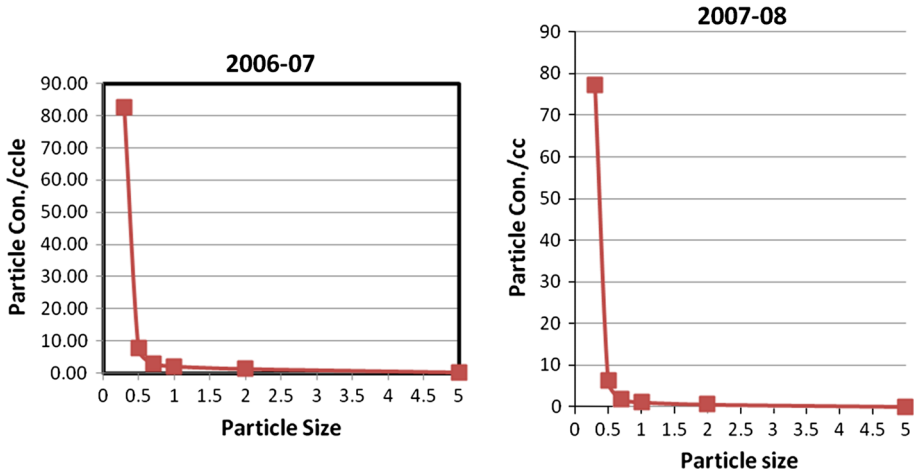

Fig. 4 Particle size distribution for 2006–2007 and 2007–2008

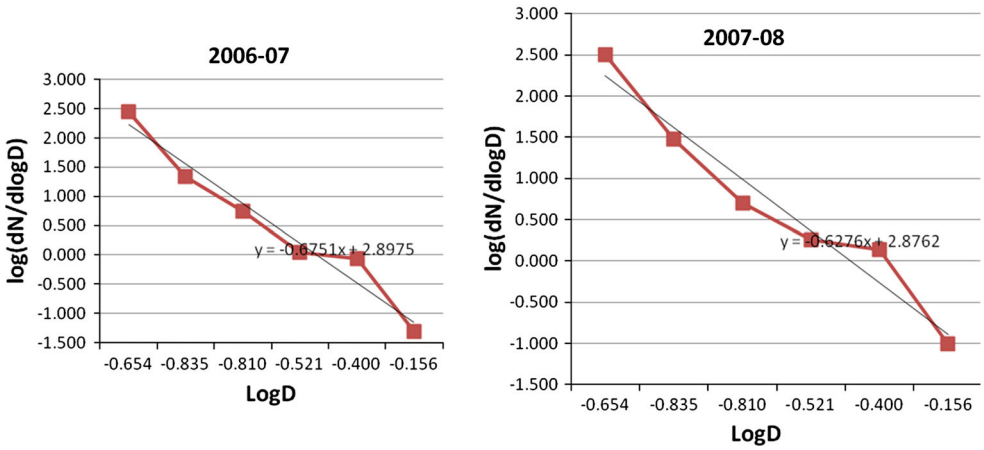

Fig. 5 Number size distribution spectrum of aerosol particles larger than 0.3 μm (log (d*N*/dlog *D*) vs. log D)

marginal evidence of dependence for the year 2006–2007. However, broadly speaking these findings suggest that the classification rule based on the weekly means result in independent states.

Next, we proceed to classify the weekly means of aerosol particle concentrations into seven severity states S_j, $j = 1,\ldots,7$ following the classification rule outlined in (3). The different severity states resulting from the elaborate but straight forward calculations are exhibited in Table 4. The Fig. 7 depicts the severity states of PM 2.5 and PM 10 concentrations during the weeks of September 2006–August 2008. By placing the weekly severity states in correspondence with the weeks of the seasons as discussed earlier, it turned out that, the aerosol particle concentrations attain weekly peaks more often during the winter seasons followed by dry and rainy seasons. Similarly, the weekly troughs occurred more often during the rainy seasons than the winter and dry seasons. The frequencies of severity states of PM 2.5 and PM 10 concentrations during the 2 years of study are reported in Table 5. The multinomial probability models for severity states can be constructed using the frequency of severity states displayed in Table 5. Table 6 provides empirical estimates of probabilities of severity states for the 2 years. From Table 5 and Table 6, it is seen that

1. The extremely critical state (S_7) is one of the most frequent states where the aerosol concentrations are appreciably higher. During the years 2006–2008, in between 7 and 11 weeks recorded excessively high PM 2.5 and PM 10 aerosol concentrations in atmosphere.
2. The chance that the mean weekly aerosol concentration measurements exceed the threshold or normal state (S_4) varies between 28 and 38 %. In other words, in about 15–20 weeks in a year, the PM 2.5 and PM 10 aerosol concentrations are rising in the city of Gaborone. This finding suggests that aerosol particle concentrations are likely to be one of the agents to influence climate change in Botswana.
3. The city witnessed more number of absolutely safe states (S_1) i.e. in about one-fifth of the year 2007–2008, than during the year 2006–2007; considering PM 2.5 concentrations. The frequency of absolutely safe states are more less the same in respect of PM 10, but appreciably smaller in number.

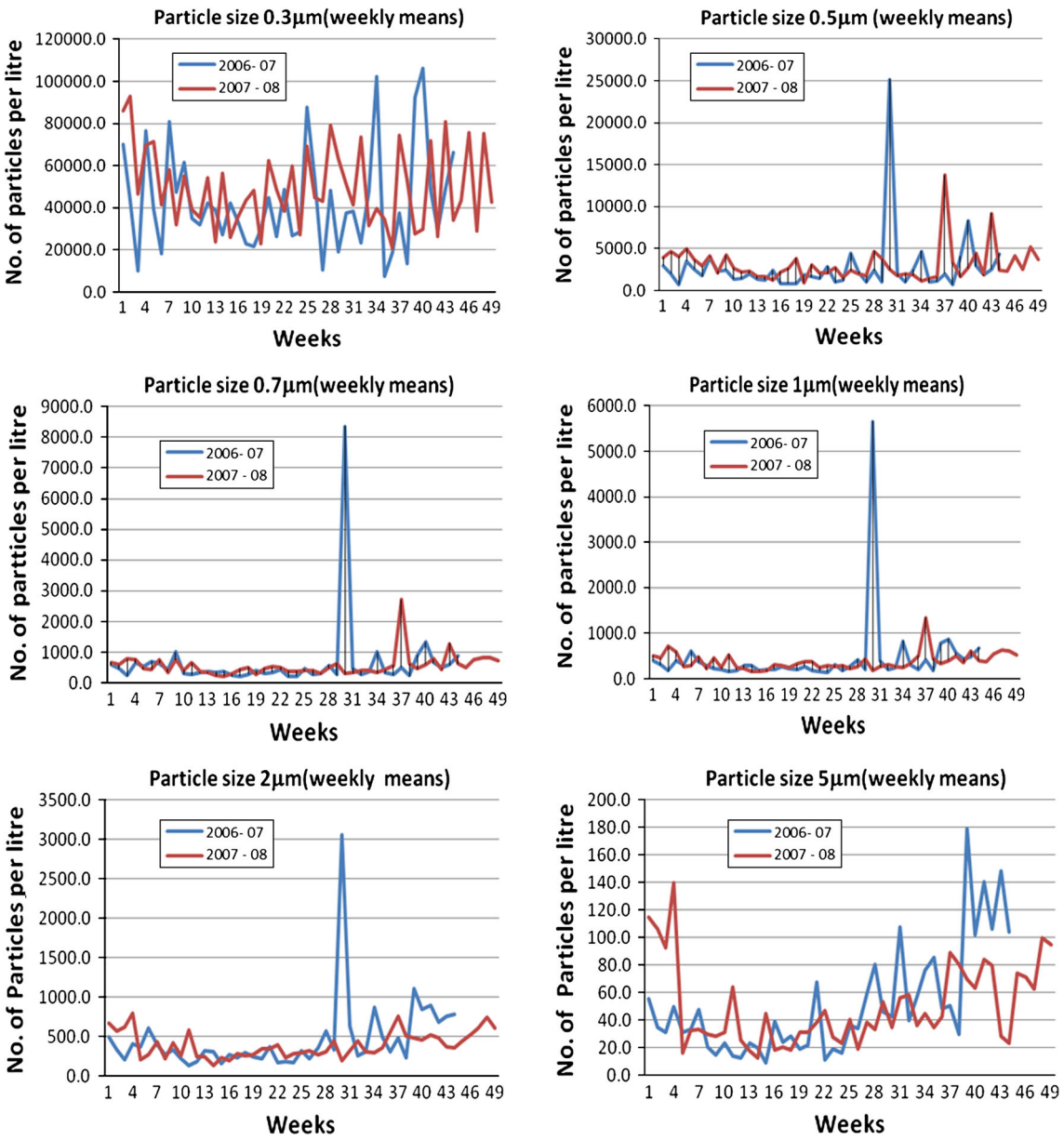

Fig. 6 Particle size distribution of weekly means for the years 2006–2007 and 2007–2008

Next we compute the volatility index I_D for the 2 years using the formula (11) and the frequency of severity states reported in Table 4. It can be verified that for PM 2.5,

$$I_D(2006-2007) = 13.0 \text{ and } I_D(2007-2008) = 4.5 \tag{13}$$

and for PM 10,

$$I_D(2006-2007) = 10.5 \text{ and } I_D(2007-2008) = 24.5 \tag{14}$$

Thus the drift indices reveal that PM 2.5 severity states in the year 2007–2008 followed a symmetric model more often than in the year 2006–2008; while PM 10 severity states were asymmetric in both the years. This analytical finding is consistent with the Botswana Meteorological Services' records which showed more rain fall during the weeks of the year 2007–2008 than in the weeks of the year 2006–2007; coupled with a shorter winter season factoring for increased precipitation and lesser biomass burning. However, PM 10 severity states were asymmetric in both years, indicative of the fact that reasons alluded to PM 2.5 concentrations seemed to have little impact.

Next we compute, dispersion index I_d using the formula (12) and values in Table 4. It turns out that for PM 2.5,

Table 2 Tests of homogeneity of variances and equality of means of PM 2.5 and PM 10 concentrations

Description	Years	Summary	Particle size	
			PM 2.5	PM 10
Test for homogeneity of variances of weekly means	2006–2007	F statistic	1.51	1.74
	2007–2008	Degrees of freedom	41.48	41.48
		p value	0.09	<0.03
		Conclusion	Insignificant	Significant
Test of equality of means (variances unequal) of weekly means	2006–2007	t statistic	−0.64	0.18
	2007–2008	Degrees of freedom	89	76
		p value	0.53	0.86
		Conclusion	Insignificant	Insignificant

$$I_{\mathrm{d}}(2006-2007)=4.02 \text{ and } I_{\mathrm{d}}(2007-2008)=4.76 \quad (15)$$

and for PM 10,

$$I_{\mathrm{d}}(2006-2007)=2.71 \text{ and } I_{\mathrm{d}}(2007-2008)=3.0 \quad (16)$$

It can therefore be concluded that both PM 2.5 and PM 10 severity states comparatively exhibited more dispersion during the year 2007–2008 than 2006–2007. In other words, the year 2007–2008 exhibited more activity in the atmospheric aerosol concentrations than the year 2006–2007 in Gaborone, Botswana.

7 Conclusions

The focus of this paper is to propose alternative ways of quantifying levels of variations in weekly mean aerosol concentration measurements, in particular PM 2.5 and PM 10 and apply the proposed techniques to atmospheric data collected in Gaborone, Botswana. The analysis is based on a new approach wherein natural variations in weekly means are perceived to belong to different severity categories depending on the magnitude of variations. These categories are defined in terms of magnitude of the spread in weekly means, a concept used in statistical theory. Two new measures called, index of drift and index of dispersion for aerosol particle concentrations have been proposed. These measures help to quantify both within and across aerosol particle concentrations given the daily data during one or more periods of study. From the analysis of the data it is revealed that

1. Severity of aerosol particle concentrations differs appreciably over the two years under study.
2. In the city of Gaborone, Botswana, extremely critical state of aerosol concentrations tends to be present more often than the other states. This may have far reaching implications on pollution and health related issues and therefore needs the attention of the city council in general and policy makers in particular.
3. In general, the city witnessed more atmospheric aerosol concentration activities in the year 2007–2008 than the year 2006–2007.

A similar analysis can be done by considering the monthly mean concentrations rather than the weekly mean concentrations. The findings of the analysis are on similar lines with those based on the weekly means data. The details of the analysis can be obtained from the authors and are not reported here. Finally, the technique developed here can be used to analyze aerosol concentrations data based on seasons, such as dry, rainy and winter to examine the relationship between aerosol concentrations and influential factors such as precipitation, biomass burning etc. The findings of such an analysis can be obtained from the authors and they are not reported here except that the analysis analytically confirms the well-established narrative relationship between aerosol concentrations, precipitation and biomass burning, eluded by earlier researchers.

In this work, the 'severity states' are defined from the variation of aerosol concentration with respect to the previous week statistics. While this is an important indicator of the atmospheric trend, but one may also take into account the absolute concentration. Most of the air pollution regulations are based on the PM 10 and PM 2.5 daily load. For example, in Europe the E.C. guidelines allow a maximum PM 10 annual concentration of 40 $\mu g/m^3$, with a maximum of 35 days of load exceeding 50 $\mu g/m^3$. The OPC allows a rough determination of the PM 10 and PM 2.5 which could be useful parameters to evaluate the state of the atmosphere, at least for health purposes. For implications on climate, other parameters such as the single scattering Albedo and the refractive index of the aerosol should be taken into account, although they need independent measurements. The use of free satellite products, such as the aerosol optical thickness (from MODIS, for

Table 3 Weekly means of PM 2.5 and PM 10 aerosol concentration measurements (cm^{-3})

September 2006–August 2007														
Weeks	1	2	3	4	5	6	7	8	9	10	11	12	13	14
Means														
PM 2.5	104.19	65.35	15.88	113.51	60.20	30.29	119.89	70.41	91.53	51.77	47.12	62.98	57.38	40.93
PM 10	0.077	0.049	0.043	0.070	0.044	0.046	0.066	0.028	0.020	0.032	0.020	0.018	0.033	0.028
Weeks	15	16	17	18	19	20	21	22	23	24	25	26	27	28
Means														
PM 2.5	63.37	48.70	33.97	32.49	45.58	65.62	40.40	73.22	39.73	42.37	130.15	78.44	72.69	111.15
PM 10	0.013	0.054	0.033	0.040	0.026	0.031	0.094	0.016	0.026	0.022	0.051	0.047	0.113	0.060
Weeks	29	30	31	32	33	34	35	36	37	38	39	40	41	42
Means														
PM 2.5	58.36	34.80	71.38	153.04	13.74	28.77	57.13	20.85	138.41	164.0	74.15	46.16	74.60	101.89
PM 10	0.150	0.055	0.079	0.107	0.119	0.067	0.070	0.042	0.249	0.142	0.195	0.148	0.207	0.145
September 2007–August 2008														
Weeks	1	2	3	4	5	6	7	8	9	10	11	12	13	14
Means														
PM 2.5	104.19	65.35	15.88	113.51	60.20	30.29	119.89	70.41	91.53	51.77	47.12	62.98	57.38	40.93
PM 10	0.077	0.049	0.043	0.070	0.044	0.046	0.066	0.028	0.020	0.032	0.020	0.018	0.033	0.028
Weeks	15	16	17	18	19	20	21	22	23	24	25	26	27	28
Means														
PM 2.5	63.37	48.70	33.97	32.49	45.58	65.62	40.40	73.22	39.73	42.37	130.15	78.44	72.69	111.15
PM 10	0.013	0.054	0.033	0.040	0.026	0.031	0.094	0.016	0.026	0.022	0.051	0.047	0.113	0.060
Weeks	29	30	31	32	33	34	35	36	37	38	39	40	41	42
Means														
PM 2.5	58.36	34.80	71.38	153.04	13.74	28.77	57.13	20.85	138.41	164.0	74.15	46.16	74.60	101.89
PM 10	0.150	0.055	0.079	0.107	0.119	0.067	0.070	0.042	0.249	0.142	0.195	0.148	0.207	0.145
Weeks	43	44	45	46	47	48	49							
Means														
PM 2.5	128.53	52.96	65.90	113.99	46.87	115.26	67.43							
PM 10	0.039	0.033	0.103	0.099	0.087	0.139	0.132							

Table 4 Severity states of PM 2.5 and PM 10 aerosol concentration measurements

September 2006–August 2007														
Weeks	1	2	3	4	5	6	7	8	9	10	11	12	13	14
States														
PM 2.5	4	3	1	7	3	4	7	2	4	3	3	5	3	3
PM 10	4	3	4	5	3	4	6	3	3	5	3	4	6	4
Weeks	15	16	17	18	19	20	21	22	23	24	25	26	27	28
States														
PM 2.5	5	1	3	4	5	5	1	7	1	4	7	2	4	6
PM 10	1	7	4	4	3	4	7	2	5	4	7	1	7	2
Weeks	29	30	31	32	33	34	35	36	37	38	39	40	41	42
States														
PM 2.5	4	3	7	7	1	7	7	1	7	4	3	2	6	5
PM 10	7	2	5	5	4	4	4	2	7	3	6	4	5	3
September 2007–August 2008														
Weeks	1	2	3	4	5	6	7	8	9	10	11	12	13	14
States														
PM 2.5	4	5	1	7	4	1	5	1	7	3	4	7	3	3
PM 10	4	4	3	7	2	6	4	4	4	4	7	2	4	4
Weeks	15	16	17	18	19	20	21	22	23	24	25	26	27	28
States														
PM 2.5	5	1	3	4	5	5	1	7	1	4	7	2	4	7
PM 10	1	7	4	4	3	4	7	2	5	4	7	3	6	4
Weeks	29	30	31	32	33	34	35	36	37	38	39	40	41	42
States														
PM 2.5	4	2	4	5	2	4	3	2	7	1	1	4	7	1
PM 10	7	2	7	4	1	6	3	4	1	4	4	4	5	4
Weeks	43	44	45	46	47	48	49							
States														
PM 2.5	7	1	6	6	1	7	1							
PM 10	2	4	7	4	3	7	4							

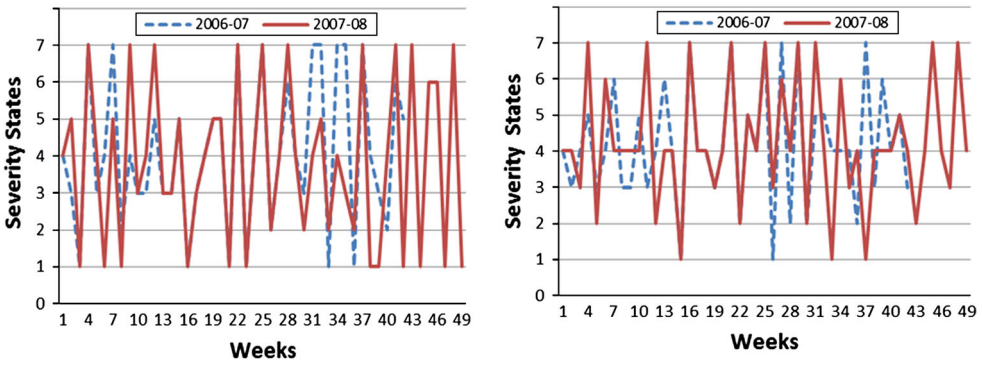

Fig. 7 Severity states of PM 2.5 and PM 10 concentrations

Table 5 Severity state frequencies during 2006–2008

Year	Concentration	States							Total
		1	2	3	4	5	6	7	
September 2006–August 2007	PM 2.5	6	3	9	8	5	2	9	42
	PM 10	2	4	8	13	6	3	6	42
September 2007–August 2008	PM 2.5	12	4	5	10	6	2	10	49
	PM 10	3	5	5	22	2	3	9	49

Table 6 Empirical estimates of severity state probabilities during 2006–2008

Year	Concentrations	States							Total
		1	2	3	4	5	6	7	
September 2006–August 2007	PM 2.5	0.143	0.071	0.214	0.191	0.119	0.048	0.214	1
	PM 10	0.048	0.095	0.191	0.310	0.143	0.071	0.142	1
September 2007–August 2008	PM 2.5	0.245	0.082	0.102	0.204	0.122	0.041	0.204	1
	PM 10	0.061	0.102	0.102	0.449	0.041	0.061	0.184	1

example) or reanalysis data such as the PBL height from GDAS data (NOAA) may be coupled to ground measurements to infer properties of the mixing status of the atmosphere. We intend to undertake such a line of approach in a subsequent study.

Acknowledgments The authors profusely thank the two reviewers for their critical comments which have greatly improved the presentation and content of the manuscript. The Department of Meteorological Services, Gaborone and Department of Physics, University of Botswana are gratefully acknowledged for their assistance with data acquisition.

References

Andronache C, Laakso L, Gronholm T, Philips V (2005) Aerosol particle scavenging during precipitation. Geophys Res Abstr 7:04919

Botswana geographical info (1996). http://www.savenues.com/weather/botswana.htm). Accessed 17 May 2008

Geographical info (1996). http://www.botswanatourism.co.bw/about/geographical-info.html). Accessed 12 May 2008

Gibbons JD, Chakraborti S (2003) Nonparametric statistical inference. Marcel Dekker Inc., New York

Hitchins J, Morawska L, Wolf R, Gilbert D (2000) Concentrations of sub micrometer particles from vehicle emissions near a major road. Atmos Environ 34:51–59

Jayaratne ER, Verma TS (2001) The impact of biomass burning on the environmental aerosol concentration in Gaborone, Botswana. Atmos Environ 35(10):1821–1828

Lohmann U, Feichter J (2005) Global indirect aerosol effects: a review. Atmos Chem Phys 5:715–737. http://www.atmos.chem.phys.net/5/715/2005/

Ma RJ et al (2003) Characteristics and influence of bio smoke on the fine particle ionic composition measured in Asian outflow during the transport and chemical evolution over pacific (TRACE-P) experiment. J Geophys Res 111. doi:10.1020/2002JD003128

Sisterson DL, Johnson SA, Kumar R (1985) The influence of humidity on fine-particle aerosol dynamics and precipitation scavenging, aerosol. Sci Technol 4(3):287–300 (Accessed on 12 Jul 2008)

Stuart A, Ord K (1994) Kendall's advanced theory of statistics, vol 1: distribution theory, 6th edn. Holder Arnold, London

Verma TS, Jayaratne ER (2001) Measurement of aerosol concentrations in Gaborone, Botswana. Botswana Notes Records 32:177–184

Verma TS, Thomas TA (2007) Atmospheric aerosol concentration due to biomass burning. Int J Meterol 32:321

Wallace JM, Hobbs PV (1977) An introductory survey. Atmospheric science. Academic Press, New York

Xuejiao DX, Xiuji ZDW, Liuju ZHT, Fei LI, Xiaoying H, Xueyan B, Tao D (2008) Effects of Southeast Asia biomass burning on aerosols and ozone concentrations over the Pearl River Delta (PRD) region. Atmos Environ 42(36):8493–8501