HISTORICAL STUDY OF POLLUTANTS EMISSION OVER EGYPT USING ACCMIP DATA

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ABSTRACT

Statistical analysis for the emissions of some pollutants over Egypt during the period from 1850 to 2000 using the ACCMIP (Atmospheric Chemistry and Climate Model Intercomparison Project) history emission inventory dataset has been done. The seasonal and annual variations of the following compounds: Black Carbon (BC), Carbon monoxide (CO), Propane (C_3H_8), Formaldehyde (HCHO), Nitrogen Oxides (NO_X), Toluene (C_7H_8), Ethylene (C_2H_4), Ammonia (NH_3), Propene (C_3H_6), Sulfur Dioxide (SO_2) and Organic Carbon (OC) have been examined. It is found that these pollutants are intense over Nile Delta and Greater Cairo (30.25º - 31.75º E and 29.25º - 31.25º N); it produces mainly from human activities. The annual analysis of these pollutants illustrates that four of them (C_3H_8, HCHO, SO_2 and NO_X) have the same behavior during the period of study, where an obvious increase appears from about 1930 to 2000. Also, there are four pollutants (BC, C_2H_4, C_3H_6 and OC) that have the same behavior, where an evident increase appears from 1830 to 2000. The higher values of the coefficient of variation (COV) appears with the seasonal and annual time series of CH_2O and C_3H_8 while the lower values of COV were found with the time series of C_2H_4 and C_3H_6. The trend analyses conclude that an obvious increase of all pollutants emissions except with NH_3 where a negative trend appears with the seasonal and annual time series during the period from 1930 to 2000.

Keywords: ACCMIP inventory; Air pollution; VOCs emission sources; Nile Delta and Greater Cairo.

1. INTRODUCTION

Currently, poor air quality it renowned as the most demanding problems in urban areas with very hurtful impacts on human health and the environment [1]. Climate change is affecting human health, where the climate stays to change; the hazards to human health will rise, exacerbating existing live and environmental threats and making new challenges [2]. Air pollution effects are mostly due to high levels of ozone (O_3), Volatile Organic Compounds (VOCs) and particulate matter (PM) in the atmosphere [1]. The pollutants classified according to sources to natural (volcanic emissions and ocean sea salt);and anthropogenic e.g. industrial emission, soil emission from agriculture activities, dust and biomass burning [3; 4; 5; 6]. The anthropogenic emissions are known as the highest contributors to air pollution; however the weather occurring at diverse scales contributes to the increase the

environmental damages [6]. The growth of the urban population is extra rapid in developing countries, that affected by pollution and climate. Modern studies have advised that climate change will have important influences on weather and urban air pollution [8; 9]. Changes in climate affect the average weather conditions, where atmospheric aerosols are liquid, solid, or mixed particles suspended in the atmosphere, with very mutable chemical composition and size [10]. Their presence is often directly observable by the human eye as haze in many industrial regions. Atmospheric aerosol particles may be emitted directly as particles (primary sources) or formed from gaseous precursors (secondary sources) via the aforementioned gas-particle conversion of SO_2, NOx, and VOC. The anthropogenic aerosols concentrations have evidently increased since the Industrial revolution start and human activity growth [11]. Inorganic primary aerosols are quite large (> 1µm) and initiate from sea salt, volcanoes

mineral and dust; while the BC and OC aerosols are generally < 1 µm [12]. The so-called superfine particles (less than about 0.1µm) mainly arise from gas-to-particle conversion whereby gases such as sulfur dioxide (SO₂), nitric oxide and nitrogen dioxide (NOx) and volatile organic carbon (VOC) are oxidized and condensed [13]. The interactions of Aerosol and clouds permit an effect of aerosols on the number and mass of cloud droplets [14]. The so-called coarse particles (> 1 µm) are usually produced mechanically (e.g. wind blowing over the dusty area, evaporation of sea salt). Fine mode aerosols generally have atmospheric lifetimes of days to about a week [15]; can be extremely variable in the spatial and temporal distributions. Through aerosols lifetime in the atmosphere, it has a great impact on the Atmospheric processes [16; 17; 3], e. g. air quality, cloud formation, human health and visibility. Such a large perturbation of the global and regional aerosols from the anthropogenic sources is a major climatic and environmental concern [18].

The Nile Delta (ND) and Greater Cairo (GC), is one of many regions in the world that suffer from air pollution problems. The GC is one of the highest densely populated areas with a population of larger than 15 million people [19]. The massive motorized traffic as well as several factories located in the ND and GC play a major role in increasing air pollution in the region [20]. Moreover, the black cloud (BCL) phenomenon produced by smoke advection generated from burning rice straw after harvest in ND was observed in GC during harvest season when temperature inversions helped intensify the situation [20; 21]. Prior studies describe that BCL over ND and its vicinity may affect regional climate through radiation scattering or absorption [22; 23; 24; 25] and explained that BCL may influence plant growth in ND by changing the amount of solar radiation existing for photosynthesis. Abdul-Wahab et al. (2011) and El-Metwally et al. (2011) suggested that the aerosols can be classified according to

pre-sources into three main sections formed by 1) the diurnal activities, 2) desert dust and 3) the agricultural activities in the ND [4; 5]. These studies illustrate that the higher aerosol concentrations are found in July month due to its higher stability of the atmospheric condition. Wheida et al. (2017) exposed that the peak of the aerosol concentrations is generally due to the advection of dust towards the GC from surrounding deserts or the agricultural biomass burning in the ND. Additionally, the impervious BCL over Cairo and its surrounding regions might affect regional climate during the scattering or absorption of solar radiation [23]. The change in incoming solar radiation affects the growth of plants in the Nile Delta, also, high aerosols concentration loads are likely to influence the water cycle by suppressing precipitations [29; 24]. Several studies have been conducted to understand the main reasons for the increased pollution levels in Cairo using ground-based and satellite air quality data [30; 31; 32; 33; 34]. However, the genesis of the fall episodes (the black cloud season) is still under discussion and needs more investigations like the present study. ***This work aimed to study the major aerosol species emissions over Egypt for the period from 1850 to 2000 using the ACCMIP emission inventory.***

2. Data and methodology

The Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) monthly mean anthropogenic pollutant emissions data for 11 species with 0.5° x 0.5° horizontal resolution during the historical period 1850 - 2000 are used in this study. The chosen compounds are Black Carbon (BC), carbon monoxide (CO), Propane (C_3H_8), Formaldehyde (HCHO), nitrogen oxides (NOx), Toluene (C_7H_8), Ethene (C_2H_4), ammonia (NH₃), Propene (C_3H_6), Sulfur dioxide (SO₂) and Organic carbon (OC). These data are downloaded from the ECCAD site (http://accmip-emis.iek.fz-juelich.de/data/accmip/). The statistical methods used in our study are:

A) Bartlett test: in this study we examine the data homogeneity when values of Gaussian distribution are considered. This method is simply expert by dividing the concidered time series into K equal subperiod (k ≥2). The sample variance S_k^2 is calculated by the following relation [35]:

$$S_k^2 = \frac{1}{n}\left(\sum x_i^2 - \frac{1}{n}\left(\sum x_i\right)^2\right) \qquad (1)$$

wherever the summaries range during the n values of the series, Σi in the subperiod K. Available data, a period of 150 years, we divide it into three subperiod (K=3), each subperiod having 50 years (n=50). The estimated ratio between the maximum and minimum values $\left(S_{max}^2 / S_{min}^2\right)$ is compared with the significance value it corresponding to n and K [36] in order to determine the percentage value of significance.

B) coefficient of variation (COV): to estimate and analyze the seasonal and annual variability the coefficient of variation (COV) as in the following equation.

$$COV = 100 * SD / \mu \qquad (2)$$

Where SD is the standard deviation and μ is the temporal mean for N years.

On the other hand, different statistical methods were applied to study the trend and fluctuations of the pollutant emissions over the studied area.

C) The trend analysis: we compute the trend by different methods, i) Least Square method [37], ii) non-parametric Mann-Kendall (M-K) rank correlation test [38; 39] is used to detect any possible trend in emissions series as well as to test the significance of the trend, iii) Gaussian low-pass filter [35] and iv) Binomial low-pass filter [35; 39].

The cumulative seasonal means (Equation 3) is used [40] to calculate the decadal and inter-decadal fluctuations or "persistence" in the behavior of the selected species.

$$y_j = \frac{1}{j}\sum_{i=1}^{j} x_i, \qquad j = 1,2,..............,N. \qquad (3)$$

Where x_i is the total amount of aerosols emissions and N is the number of years of data used.

4- Abrupt change test: the abrupt change procedure (Mann-Whitney test for step trend) which is given in Equation 4.

$$Z_c = \left[\sum_{t=1}^{n1} r(x_i) - n_1(n_1 + n_2 + 1)/2\right] \Big/ \left[n_1 n_2(n_1 + n_2 + 1)/12\right]^{1/2} \qquad (4)$$

Where r(xi) is the rank of the observations. Given the data vector $X = (x_1, x_2, \ldots\ldots, x_n)$, partition X so that $Y = (x_1, x_2, \ldots\ldots, x_{n1})$ and $Z = (x_{n1+1}, x_{n1+2}, \ldots\ldots\ldots, x_{n1+n2})$. The null hypothesis H0 is accepted if $-Z_{1-\frac{\alpha}{2}} \le Z_c \le Z_{1-\frac{\alpha}{2}}$, where $\pm Z_{1-\frac{\alpha}{2}}$ are the $1 - \frac{\alpha}{2}$ quintiles of the standard normal distribution corresponding to the given significance level α for the test.

4. RESULTS

4.1 Homogeneity of the data

The homogeneity analysis of data is an important test to improve the ability of data to use and is essential in climatology. Homogeneity is manifested otherwise in different climatic elements. The climatic elements' values could be the method of estimating annual and seasonal averages. The human effects and changes can be lead to the local environment change, produce for values and non-homogeneity in historical records of climatic data. Homogeneity of pollutants emission over the study area has been examined by means of Bartlet test [35]. The available data period is 152 years, we divided each monthly time series into three subperiod (K=3), each subperiod contains 50 years (N=50), and we found the corresponding 95% significant point is 8.38. Figure 1 shows the Bartlet test (short-

cut) result for the annual time series of our pollutants emission. The annual time series of our pollutants emission seems to be homogeneous regarding the ratio (S_{max}^2 / S_{min}^2) Bartlet test with 95% significance point as shown by Mitchell et al., (1966).

4.2 Spatial distribution of emissions

The spatial distribution of the annual mean emissions from the ACCMIP reanalysis dataset over Egypt is illustrated in Figure 2. It is evident that, the anthropogenic pollutants emissions generally occur along with and over the northern part of the River Nile and Delta region (nearly from 25° N up to 31° N). Furthermore, the highest anthropogenic pollutants emissions are detected over the Nile Delta region (red box in Figure 2) particularly over Greater Cairo (GC) which agrees with Aboel Fetouh et al. (2013), as the GC area in Egypt is the largest city in northern Africa and one of the 15 largest cities in the world with a population of about 15 million people [44]. According to the Central Agency for Public Mobilization and Statistics [44], the total number of licensed vehicles in Cairo governorate were out of 2.3 million vehicles from 8.6 million vehicles overall Egypt at the end of December 2015. This dense population, human activates and great vehicle number in the Cairo governorate causes high anthropogenic emissions from transportation, industry, and waste burning. So, in this study we focused our attention on emissions over the Nile Delta region including Cairo city. Table 1 explains the climatic average ranges for ACCMIP emissions (based on Figure 2) and their principal sources.

4.2 Annual pollutants emissions

Figure 3 shows the annual values of the pollutants emission throughout the period of study. It is clear that CO record the highest concentration of emission (65.9 – 1490.6) kg m-2 s-1*10-12, while HCHO it records a lower values of emissions (0.23 – 21.3) kg m-2 s-1 *10-12. The annual averages of pollutants

emission; BC, OC, C_2H_4, C_3H_8, NH_3, NO_X, C_7H_8 and SO_2 are record, (0.97 – 15.32), (4.26 – 22.06), (1.47 – 12.02), (0.25 – 19.7), (7.95 – 9.65), (0.89 – 131.64), (0.47 – 19.01) and (0.4 – 300) x10-12 kg m-2 s-1, respectively. In addition, Figure 3 shows the change in the trend behavior of pollutants emission. The trend of pollutants emission: NO_X, HCHO, C_7H_8, C_3H_8 and SO_2 have five periods, the first one from begining to about 1930 while the trend is constant, the second period from 1930 to 1960 for HCHO, C_7H_8 and C_3H_8, but from 1930 to 1970 with a weak increasing, the third period from 1960 to 1970 for HCHO, C_7H_8 and C_3H_8 with a strong increase, except NO_X and SO_2; the NO_X continues to increase by the same behavior up to 1970, and SO_2 have constant values from 1960 up to 1970, the four period from 1970 to 1980 with decreasing for C_3H_8, and HCHO and increasing values for NO_X and strong increasing for SO_2 from 1970 up to 1990, the five periods from 1980 to 2000 for C_3H_8, HCHO, C_7H_8, but SO_2 decreases from 1990 up to 2000. BC, OC, C_2H_4 and C_3H_6 hase increases gradually from the beginning of the available period up to the year 1890, the rate of increase is accelerating during a period 1890 - 2000 for CO and BC, but the last ten years for C_2H_4 and C_3H_6 are constant. A sharp increase of OC appears in the period 1940 - 2000. The C_3H_8, HCHO, NOx and C_7H_8 annual values have the same behavior throughout the period of study, where there is an increase in its values from 1990 up to 2000. The values of BC from 1940 to 1960 decreases gradually and increase during the period from 1960 to 1980, followed by a significant increase up to 2000. The annual values of NH_3 nearly a stable behavior from beginning the period up to 1940, these annual values are nearly constant followed by a sharp increase up to 1950, and sharp decrease up to 1960 directly. After this jumping, the trend of NH_3 is decreasing up to 1980, and constant up to end the period. The behavior of the chemical species CO experiences an obvious increase throughout the available data period (1850-2000), this increase

becomes very slowly during the period 1850 to 1930. The OC is increasing from beginning the period up to 1940, followed by a decrease period up to 1960, and an increase from 1960 up to 2000.

4.3 Coefficient of variation (COV)

The COV of the annual and seasonal pollutants emission is displayed in Figure 4. The higher values of COV appear with the annual and seasonal time series of HCHO, C_3H_8, SO_2 and NH_3. The second higher values of COV are for NO_X, BC, C_7H_8, CO and OC for all seasons and annual time series. The lower values of COV for the seasonal and annual time series are for C_3H_6 and C_2H_4. In general, the COV for NO_X, BC, C_7H_8, CO and OC in spring are lower than those corresponding in winter, summer and autumn. The value of COV for all species in winter, summer and autumn is equaled. The COV of C_3H_6 and C_2H_4 seems to be constant for all seasons. According to Table 2 we noted that the COV during the long period (1850-2000) has a largest values for all pollutants.

4.4 Trend analysis

The annual values of pollutants emission time series were investigated to determine their trends. The studies of trends were performed by means of both a simple and a sophisticated tools. In our study we will use two methods the first one is the Least square method of first order (linear regression), while the second method is Mann-Kendall rank statistical test. The evaluation of the trend is based on the M–K rank statistical test. M–K rank statistics, which make no supposition about probability distribution for the original data, are tested for significance using a standard distribution. The spatial distribution pattern is not complex, where the resultant statistics of the M–K test give both negative and positive trends. Figure 5 shows the M–K statistics for the pollutants emission. The values of M–K trend test (u) were computed according to Sneyers (1990).

Figures 5a-e show the seasonal and annual values of the pollutants emission trend by the Mann-Kendall rank statistical test, while Figure 5f illustrates trend vales by least-square test for annual pollutants emission. Generally, Positive trends are observed for all species pollutants emissions. This means that the pollutants emissions over the area of study are increasing continuously. The higher trend values of a trend are observed for the emission HCHO, C_3H_8, SO_2 and NH_3 for the annual and seasonal. The lower trend values of a trend are observed for the emission of C_2H_4 and C_3H_6 for the annual and seasonal. The trend analysis by the least-square shows that all pollutants have a positive annual trend. The CO has the largest values, while C_3H_6 have lower values of the annual least square trend. The behavior of negative or positive annual trends by the Mann-Kendall rank statistical test is very consistent with the corresponding values from the least square method. But the trend analysis during 1930-2000 illustrate that the NH_3 has a negative trend (Table 2).

4.5 Cumulative annual mean

In this section, the pollutants long-term variability behavior of the annual values is analyzed with respect to the time variations of annual aerosols emissions. To visualize the vacillations in the annual pollutants emission behavior, we use the cumulative means, because they favorably expose time-varying assemblies in time-series. Furthermore, the cumulative means have a smoothing result, alike to low-pass filters [45]. Persistent phases of irregular increase or decrease of the pollutants emissions, which vary in length, are familiar in the time series of the annual pollutants emission. Figures 6 and 7 illustrates the pollutants emission behavior during two periods 1850-2000 and 1930-2000. Careful examination of the results provided for the pollutants emission leads to the following main features:-

- The CAM of the BC emissions has a negative trend in emissions during the first

period (1850- 1930). This period followed by a positive trend up to the end of the available data (2000).

- The CAM of the OC emissions has a negative trend in emissions during the first period (1850- 1920). This period followed by a positive trend up to the end of the available data (2000). OC emissions tend to be fixed after the year 1940 to 1990 and increase from 1990 to 2000.

- The CAM of the CO, C_2H_4 and C_3H_6 emissions have the same behavior throughout the available period, where they experience a negative trend in emissions during the first period (1850- 1940), followed by positive trend up to the end of the available data (2000).

- The CAM of the NH_3 emissions has a negative trend in emissions during the first period (1850- 1945). This period followed by a positive trend up to the end of the available data (2000). NH_3 emissions tend to be strong increasing after the year 1950 to 1960, and low increase from 1960 to 1970, but are decreasing from 1970 to 2000.

- Where NO_X results appear to be similar, where the behavior of CAM time series can be divided into two periods, the first extending from the beginning of the data to 1955 illustrate a negative trend, while the second period illustrates a positive emissions trend (1955-2000).

- A negative trend of C_7H_8 and SO_2 occurred during the period of 1850 to 1960, followed by a positive trend up to the year 2000.

-Negative trend values of C_3H_8 and HCHO were the dominant features during the period 1850 to 1965. This period was followed by a positive trend up to the year 2000. This means that emissions of these pollutants are increasing, due to population growth and increasing the number of vehicles, and increase the amount of fuel burnt [46; 47].

- Moreover, Figure 7 illustrate that all pollutants emission behavior is changed around the year 1970 (before or after it by about three years) except for the OC and NH_3 thire are have complex behavior.

4.6 Abrupt change analysis

The results of the Manne Whitney test for the abrupt change of the time series of the eleven pollutants emission for 70 years (from 1930 to 2000) at the 0.1 significance level are illustrated in Figure 8. Figure 8a shows the departure curves of the time series of the CO and illustrates that it has a 9 periods, the first two periods are negative (1951-1959 and 1960-1967) and other periods are positive (1968-1974, 1975-1981, 1982-1987, 1988-1994 and 1995-2000). The Manne Whitney test detected 9 periods abrupt changes in the time series of C_3H_6 the first period is negative (1930-1939) while the other periods are (1940-1947, 1948-1955, 1956-1962, 1963-1969, 1970-1975, 1976-1982, 1983-1989 and 1990-2000) as shown in Figure 8b. The Manne Whitney test detected eight periods abrupt changes in the time series of C_2H_4 the first and second periods are negative (1930-1940 and 1941-1948) while the other periods are (1949-1956, 1957-1963, 1964-1970, 1971-1976, 1977-1983 and 1984-2000) as shown in Figure 8c. Figure 8d shows the Manne Whitney test detected seven periods abrupt changes in the time series of NO_X, where the first and second periods are negative (1930-1947 and 1948-1960) while the other periods are (1961-1970, 1971-1976,1977-1983, 1984-1990 and 1991-2000). Figure 8e and f show the departure curves of the time series of the BC and C_7H_8 emission that found the six obvious periods in the study area for the past 70 years, the first one is the decrease period of 1930-1963. The other five periods are the increase periods, these periods are 1964-1973, 1974-1981, 1982-1987, 1988-1993 and 1994-2000, respectively. While the C_3H_8 and HCHO have the same departure curves fluctuate and have five periods; the first one is negative (1930-1959) but the other periods are positive (1960-

1965, 1966-1984, 1985-1991 and 1994-2000; respectively) as shown in Figure 8g and h. Moreover the Whitney test detected 7 periods abrupt changes in the time series of OC illustrates in Figure 8i; the third and fourth periods are negative (1952-1975 and 1976-1982) but the other periods are positive (1930-1944, 1945-1951, 1983-1988, 1989-1995 and 1996-2000). The SO_2 Whitney test detected 7 periods abrupt changes in the time series illustrates in Figure 8j; the first and second periods are negative (1930-1942 and 1943-1952) while the other periods are (1953-1971, 1972-1978, 1979-1984, 1985-1996 and 1997-2000). Finally the NH_3 Whitney test detected 9 periods abrupt changes in the time series illustrates in Figure 8k; the first and second periods are negative (1930-1941 and 1942-1950) while the other periods are (1951-1959, 1960-1967, 1968-1974, 1975-1981, 1982-1987, 1988-1994 and 1995-2000).

5. CONCLUSION

In this work, a statistical study for the major aerosol species emissions over Egypt for the period from 1850 to 2000 using the ACCMIP emission inventory hase been done. The annual and seasonal variations of the pollutants: Black Carbon (BC), Carbon monoxide (CO), Propane (C_3H_8), Formaldehyde (HCHO), Nitrogen Oxides (NOx), Toluene (C_7H_8), Ethene (C_2H_4), Ammonia (NH_3), Propene (C_3H_6), Sulfur dioxide (SO_2) and Organic Carbon (OC) have been examined. The most significant findings are as follows:

- The pollutants emissions increase with increasing human activities, where the population density had increased by about fourteen-fold since 1850-2000.
- The main period divided into two sub-periods; the first one from the beginning to about 1929 with the low rate of increase; the second initiated from 1930 up to final with the high rate of increase due to increasing the agriculture and industrial activities.
- The higher values of coefficient of variation

(COV) appear with time series of HCHO, SO_2, C3H8 and NH_3 while the lowest values were found in the time series of NO_X, BC, C_7H_8, OC, C_2H_6, and C_2H_4.

- The trend analyses indicate that an obvious increase in all pollutants emissions in the 1850-2000 period.
- The trend analysis during 1850-1929 period shows that all pollutants emission has a positive trend, where during the 1930-2000 period all pollutants emission have a positive trend except NH_3hase a negative trend in annual time series.
- The results of the cumulative annual mean show that, all pollutants emissions have a low growth rate during the period 1850-1929 and have a high growth rate during the period 1930-2000. This means that emissions of these pollutants were increased due to population growth and increasing the number of vehicles, and increase the amount of fuel burnt.
- The Manne Whitney test detected many abrupt changes in the time series of pollutants due to the period it's along and high changing in human activity; these periods discussed in Table 3.

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