The International Journal of Engineering and Science (IJES)
|| Volume || 7 || Issue || 9 Ver.II || Pages || PP 64-74 || 2018 ||
ISSN (e): 2319 – 1813 ISSN (p): 23-19 – 1805

Trend Analysis of the Sulfur Dioxide and Particulate Matter Concentrations in the Aegean Region, Turkey

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ABSTRACT
Trend analysis of the concentrations of air pollutant matters is important to assess the long-term increase or decrease. Trend analyses of the concentrations of sulfur dioxide (SO_2) and particulate matter (PM_{10}) in annual and winter season (1990–2009) periods in the Aegean Region cities,Denizli, Izmir, Afyon, Aydin, Kutahya, Manisa, and Usak, were investigated using the methods of linear regression, Mann–Kendall, Sen's, and Spearman's Rho. Linear regression analyses showed that decreasing trends of the SO_2 concentrations were observed in Izmir and Kutahya. The PM_{10} concentrations in winter season also showed downward trend in Izmir. According to the Mann–Kendall, Sen's, and Spearman's Rho tests, decreasing trend was observed for the SO_2 concentrations in Denizli, while the upward trend was seen for the PM_{10} concentrations. Constant reductions in the SO_2 and PM_{10} concentrations were determined in Izmir. Downward trend of the SO_2 concentrations was seen in Afyon. Increasing winter season trend of the SO_2 concentrations was determined in Aydın. There was downward trend of the SO_2 concentrations in Kutahya, while increasing trend was generally seen for the PM_{10} concentrations. Decreasing trend of the SO_2 concentrations was observed in Manisa. Downward trend of the annual SO_2 concentrations was determined in Usak. Probability distribution of the data was identified as lognormal according to the Kolmogrov–Smirnov and Anderson–Darling tests.

KEYWORDS-*Mann–Kendall method, particulate matter, Sen's method, Spearman's Rho method, sulfur dioxide, trend analysis.*

Date of Submission: 06-09-2018 Date of acceptance: 22-09-2018

I. INTRODUCTION

Anthropogenic emissions of SO_2 originate from stationary fuel combustion and industrial processes. Stationary fuel combustion includes all boilers, heaters, and furnaces found in utilities, industry, and commercial and residential establishments [1]. SO_2 is sourced from the oxidation of sulfur contained in fuel as well as from certain industrial processes that utilize sulfur containing compounds. Particulate matter can be emitted from both natural (pollen, sea salt, etc.) and anthropogenic (agriculture, combustion, construction activity, etc.) sources [2]. Particles include solid and liquid substances. Solid and liquid particles form aerosols when dispersed in the atmosphere [3].

Pan and Chen [4] signified that statistical control charts are useful tools to monitor the quality level of environment; such as air quality, industrial pollution, etc. Many techniques are used to determine the statistically significant trends of atmospheric pollution. Linear regression, Mann–Kendall, Sen's, and Spearman's Rho methods are used in the majority of statistical air quality studies. Concentration trends of air pollutants are analyzed to assess the importance of long–term increase or decrease. There are worldwide studies to determine the trends of air pollutants. Guerreiro et al. [5] pointed out that emissions of the main air pollutants in Europe declined in the period 2002–2011. The air pollutant emissions from agriculture and the combustion of fuels for domestic heating has either decreased very little (in the case of agriculture) or not decreased (in the case of domestic fuel combustion). The current state of air quality worldwide indicates that SO_2 maintains a downward trend throughout the world, with the exception of the restrictions on the use of fossil fuels with high sulfur content in developed and many developing countries. During the 1999–2008 period a statistically significant downward trend in the concentrations of SO_2, nitrogen oxides, carbon monoxide (CO), and particulate matter was investigated at most of the urban and urban–background monitoring sites in the Madrid metropolitan area [6].

This study focuses on analyze the trends of the annual and winter season SO_2 and PM_{10}concentrations inDenizli, Izmir, Afyon, Aydin, Kutahya, Manisa, and Usakbetween 1990 and 2009. Trend analyses were made for these parameters using linear regression, Mann–Kendall, Sen's, and Spearman's Rho methods. Additionally, these data sets were evaluated best fit probability distribution analysis.

II. MATERIALS and METHODS

2.1 Study site

Fig. 1 shows the geographical locations of Denizli, Izmir, Afyon, Aydin, Kutahya, Manisa, and Usakin the Aegean Region. The Aegean Region is one of the 7 census defined regions of Turkey. It is located in the west part of the country: bounded by Aegean Sea on the west; Marmara Region on the north; Mediterranean Region on the south and southwest; and the Central Anatolia Region on the east. With its 79.000 square kilometers of land, the Aegean region occupies 11% of the total area of Turkey. The climate of the Aegean Region has a Mediterranean climate at the coast, with hot, dry summers, and mild to cool, wet winters, and a semi–arid continental climate in the interior with hot, dry summers, and cold, snowy winters.

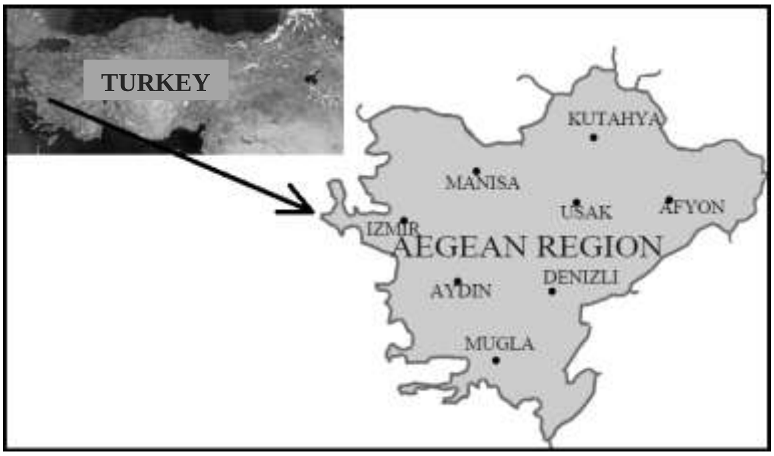

Fig. 1 Geographical location of the cities in the Aegean Region, Turkey

2.2 Trend analysis

The parametric and non–parametric methods were used to determine trends in air pollutants time series. Trend analyses were made for the SO_2 and PM_{10} concentrations using linear regression analysis, Mann–Kendall, Sen's, and Spearman's Rho methods. For this purpose the $SO_2$and PM_{10}concentrations and temperature values during the period from 1990 to 2009 including averages of annual and winter season pattern were used. Yearly concentrations were estimated as the arithmetic mean of monthly concentrations. Monthly average concentrations of SO_2 and PM_{10} were obtained from the initial daily observation network including measurement stations in these cities between 1990 and 2009. Daily concentrations of SO_2 and PM_{10} used in this study were provided from Turkish Statistical Institute, and the Ministry of Environment and Urban Planning Air Quality Monitoring Network. November, December, January, February, and March months are in winter periodin this study. The concentrations in winter season were calculated as the arithmetic average of these months for each period. Evaluation of air pollution in Mugla has not been made because of the lack of data.Due to some technical problems, SO_2 and PM_{10} concentrations could not be measured on some days. Missing values were substituted. For example, if for a particular month, a value was missing, then it was substituted by considering the average of the preceding and succeeding months. Any downward or upward trend was evaluated statistically significant at the 95% confidence level.

2.2.1 Linear regression method

A linear regression method is to check whether there is a significant relation between the variables under consideration. The regression line is used to estimate a slope. The slope indicates the mean temporal change of the studied variable. Positive values of the slope show increasing trends, while negative values of the slope indicate decreasing trends. A linear regression line has an equation of the from

$$y = a + bx \tag{1}$$

where x is the explanatory variable, y is the dependent variable, b is the slope of the line and a is the intercept [7].

2.2.2 Mann–Kendall test

The non–parametric Mann–Kendall test is an efficient statistical tool widely used for analyzing long–term trend of data in a more meaningful way. There are many advantages of using this test. First, it is a non–parametric test and does not require the data to be normally distributed. Second, the test has low sensitivity to abrupt breaks due to inhomogeneous time series [8]. Minimal impressionability from limit values observed in some time series is another advantage of this method [9]. According to this test, the null hypothesis H_0 assumes

that there is no trend (the data is independent and randomly ordered) and this is tested against the alternative hypothesis H_1, which assumes that there is a trend [10].
Mann–Kendall test [11, 12] is based on the correlation between the ranks of a time series and their time order. For a time series X= {x_1, x_2, . . .,x_n}, the test statistic is given by

$$S = \sum_{k=0}^{n-1} \sum_{j=k+1}^{n} \operatorname{sgn}(x_j - x_k) \operatorname{sgn}(x) = \begin{cases} +1, & x > 0 \\ 0, & x = 0 \\ -1, & x < 0 \end{cases} \quad (2)$$

In Equation 2, where n is the length of the data; $x_j$and $x_k$are the data values in time series k and j (j>k), respectively. In cases where the sample size n≥10, the mean and variance are given by

$$E(s) = 0 \quad (3)$$

$$Var(s) = \sigma_s \sqrt{\left(n(n-1)(2n+5) - \sum_{1}^{r} t_i (t_i - 1)(2t_i + 5) \right) / 18} \quad (4)$$

where n is the number of tied groups and t_i denotes the number of ties of extent i. A tied group is a set of sample data having the same value. The standard normal test statistic Z is computed as:

$$Z = \begin{cases} \frac{s-1}{\sigma_s}, & s > 0 \\ 0, & s = 0 \\ \frac{s+1}{\sigma_s}, & s < 0 \end{cases} \quad (5)$$

According to Equation 5, a positive value of Z informs increasing trends while the negative Z informs decreasing trends. Testing of trends is made at a specific α significance level. The significance level of $\alpha = 0.05$ was used in this study. At 5% significance level, the null hypothesis of no trend is rejected if $|Z| > 1.96$ [11, 12].
The annual and winter season Mann–Kendall was calculated to quantify the magnitude of the trend, expressed as a slope (pollutant concentration per unit time).

2.2.3 Sen's method

The non–parametric procedure developed by Sen [13]. The slope estimates of N pairs of data are predicted by Sen's estimator.In Equation 6, x_j and x_k are the data values in time j and k (j>k), respectively.

$$Q_i = \frac{(x_j - x_k)}{j - k} (i = 1, \ldots, N) \quad (6)$$

The N values of Q_i are ranked from smallest to largest and the median of slope or Sen's slope estimator is computed as

$$Q_{med} = \begin{cases} \left[Q_{\frac{(N+1)}{2}} \right] N\, is\, odd \\ \frac{1}{2} \left[Q_{\frac{N}{2}} + Q_{\frac{(N+2)}{2}} \right] N\, is\, even \end{cases} \quad (7)$$

Q_{med} is computed by two sided test and then a true slope can be obtained by the non–parametric test.

2.2.4 Spearman's Rho method

Spearman's Rho test is nonparametric method commonly used to the absence of trends. Its statistic D and the standardized test statistic Z_D are expressed as follows [14, 15]:

$$D = 1 - \frac{6 \sum_{i=1}^{n} (R(X_i) - i)^2}{n(n^2 - 1)} \quad (8)$$

$$Z_D = D \sqrt{\frac{n-2}{1 - D^2}} \quad (9)$$

where$R(X_i)$ is the rank of i_{th} observation X_j in the time series and n is the length of the time series. Positive values of Z_D indicate increasing trends while negative Z_D show decreasing trends. At 5% significance level, the null hypothesis of no trend is rejected if $|Z_D| > 2.08$.

III. RESULTS

3.1 Evaluation of the SO_2, PM_{10} and temperature data

The annual and winter season SO_2 and PM_{10}concentrations and temperature values measured in Denizli, Izmir, Afyon, Aydin, Kutahya, Manisa, and Usak during the study period (1990–2009) were given in Fig. 2.

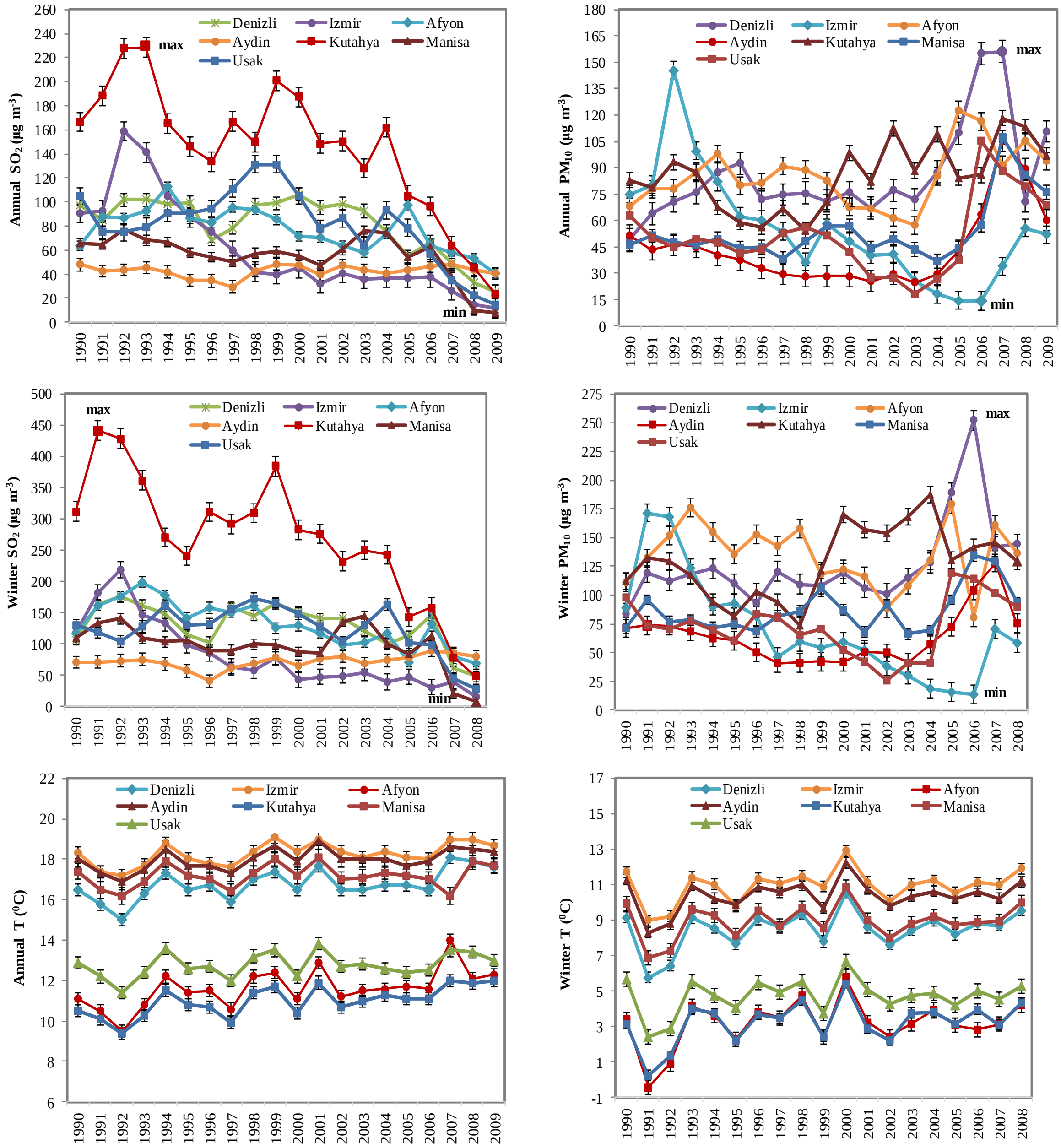

Fig. 2 SO_2 concentrations, PM_{10} concentrations, and temperatures between 1990 and 2009

Ranges of the SO_2 mean annual concentrations were 42.6 ± 4.9 µg m^{-3} for Aydin and 144.2 ± 53.9 µgm^{-3} for Kutahya during the study. These values are above the limit value of the annual SO_2 concentration given in the Regulation on Air Quality Assessment and Management (AQAMR) in Turkey. The highest value of the annual SO_2 concentrations was recorded as 228.8 µg m^{-3} in Kutahya in 1993, while the lowest value of the annual SO_2 concentrations was observed as 8.3 µgm^{-3} in Manisa in 2009. The annual SO_2 concentrations in 2009 were the lowest values during the study period except Aydin. There were noticeable declines in the SO_2 concentrations in all cities since 2007. However, reductions of the SO_2 concentrations in all cities between 1990 and 2006 years were less dramatic.

The PM_{10} mean annual concentrations were ranged from 44.0 ± 21.0 µg m^{-3} to 85.7 ± 27.1 µgm^{-3} for the period of study. These values are above the limit value of the annual PM_{10} concentration given in the AQAMR. During the annual measurement campaigns the maximum PM_{10} concentrations reached 156.1 µgm^{-3}

in Denizli in 2007. The minimum annual PM_{10} concentration was observed as 14.2 µg m^{-3} in Izmir in 2006. Very high annual PM_{10} concentrations were noticed between 2005 and 2007 years in all cities except Izmir.
Very high SO_2 concentrations were recorded in winter season for the study period. Ranges of SO_2 mean concentrations in winter season were 71.6 ± 10.6 µg m^{-3} and 266.5 ± 102.4 µgm^{-3}. These values are above the limit values of the winter season SO_2 concentration given in the AQAMR. High SO_2 concentrations were especially recorded before 2000. SO_2 concentrations recorded during 1991–1992 winter season in Kutahya (441.4 µg m^{-3}) were higher than those recorded in other cities. The concentrations of SO_2 observed in Kutahya were generally high levels except the last two years. The minimum SO_2 concentrations were determined during 2008–2009 winter season for the study period in all cities except Aydin. The lowest value of SO_2 concentrations was signified as 8.8 µgm^{-3} in Manisa during 2008–2009 winter season.

The PM_{10} mean winter season concentrations were ranged from 63.4 ± 22.2 µgm^{-3} to 134.5 ± 26.3 µgm^{-3} for the period of study. During 2006–2007 winter season, the maximum PM_{10} concentrations reached 252.4 µgm^{-3} in Denizli, while the minimum PM_{10} concentrations was determined as 13.4 µg m^{-3} in Izmir. The high PM_{10} concentrations were observed since 2004–2005 winter season in all cities except Izmir.
In the evaluation of the air quality data of Denizli between 1990 and 2009, it was concluded that winter season limits of the AQAMR were exceeded significantly by the SO_2 and PM_{10} concentrations in some periods. Since 1990, the annual long–term World Health Organization standards for SO_2 and PM_{10} have been severely violated in cities at the study area during the heating season (November–March) period.

No significant change in the annual and winter season temperatures in the cities at the study area was observed between 1990 and 2009. Ranges of the annual and winter season average temperatures were 11.0 ± 0.7 ^{0}C (Afyon) to 18.3 ± 0.5 ^{0}C (Izmir) and 3.2 ± 1.2 ^{0}C (Afyon) to 10.9 ± 0.9 ^{0}C (Izmir), respectively. The highest value of the annual temperature was recorded as 19.1 ^{0}C in Izmir in 1999, while the lowest value of the annual temperature was observed as 9.5 ^{0}C in Afyon in 1992. In 1991–1992 winter season, the minimum temperature was observed as –0.42 ^{0}C in Afyon. The maximum temperature value was observed as 12.9 ^{0}C in Izmir in 2000–2001 winter season.

3.2 Statistical relations between the annual and winter season SO_2 and PM_{10} concentrations

The strongest statistical relation between the annual and winter season concentrations of SO_2 was observed for Izmir. The significant relations were also determined between the annual and winter season concentrations of SO_2 for Kutahya, Manisa, and Usak. Variations in the annual and winter season concentrations of PM_{10} in Denizli, Izmir, and Aydin were statistically significant. The highest correlation coefficient was determined between the annual and winter season concentrations of PM_{10} in Izmir. There was no correlation for the annual SO_2–PM_{10} concentrations and winter season SO_2–PM_{10} concentrations except Izmir. The annual SO_2–PM_{10} concentrations and winter season SO_2–PM_{10} concentrations in Izmir varied accordance with each other (Table 1).

Table 1 Regression coefficientsfor the annual and winter season concentrations of SO_2 and PM_{10}

	R^2						
Period	**Denizli**	**Izmir**	**Afyon**	**Aydin**	**Kutahya**	**Manisa**	**Usak**
Annual SO_2 – Winter SO_2	0.56	0.87	0.43	0.48	0.84	0.79	0.80
Annual PM_{10} – Winter PM_{10}	0.60	0.79	0.16	0.77	0.58	0.44	0.54
Annual SO_2 – Annual PM_{10}	0.27	0.74	0.04	0.10	0.11	0.48	0.18
Winter SO_2 – Winter PM_{10}	0.01	0.82	0.07	0.29	0.04	0.12	0.17

Kolmogrov–Smirnov test and Anderson–Darling test were used to determine the suitability of data to probability distribution. The probability distribution was identified as lognormal according to the test results.

3.3 Linear regression analysis

The critical t value at 0.05 level was determined as 2.11. Hypotheses were accepted since t values were below 2.11 fora two sided test. Regression coefficients that greater than 0.60 were considered statistically significant.

Decreasing trends of the annual SO_2 concentrations were seen in Izmir and Kutahya. No annual trend was detected for the SO_2 concentrations in the other cities. The maximum correlation coefficient for the annual concentrations of SO_2 was determined in Izmir. Variations of the annual PM_{10} concentrations were not significant as statistically. No significant statistical relations were found for the annual temperatures (Table 2).

Table 2 Linear regression analysis of the annual SO_2 and PM_{10} concentrations and temperatures

	Denizli	**Izmir**	**Afyon**	**Aydin**	**Kutahya**	**Manisa**	**Usak**
Annual SO_2							
R^2	0.52	0.73	0.39	0.03	0.65	0.37	0.35
a	5964.0	11745.8	3891.8	–226.1	15153.8	3837.0	6295.8
b	–2.94	–0.12	–0.20	0.19	–0.09	–0.19	–0.11
t	–0.54	–1.19	–0.31	0.00	–0.87	–0.28	–0.25
Annual PM_{10}							
R^2	0.37	0.52	0.14	0.16	0.31	0.30	0.08
a	–5609.7	7697.3	–2113.1	–2896.9	–3546.7	–3127.6	– 1979.1
b	2.85	–0.13	0.13	0.11	0.17	0.19	0.08
t	0.37	0.52	0.14	0.16	0.31	0.30	0.08
Annual T							
R^2	0.37	0.30	0.35	0.27	0.47	0.05	0.12
a	–139.2	–85.2	–180.8	–71.9	–161.8	–26.9	–57.3
b	0.08	5.81	3.65	5.92	5.46	2.41	3.33
t	0.29	0.20	0.26	0.16	0.45	0.01	0.04

Linear regression analysis showed that decreasing trends for the winter season SO_2 concentrations were seen in Izmir, Afyon, and Kutahya. No trend was detected for the winter season SO_2 concentrations at the other cities. Decreasing trend of the winter season PM_{10} concentrations was observed in Izmir. Variations of the winter season temperature values were not significant as statistically (Table 3).

Table 3 Linear regression analysis of the winter season SO_2 and PM_{10} concentrations and temperatures

	Denizli	**Izmir**	**Afyon**	**Aydin**	**Kutahya**	**Manisa**	**Usak**
Winter Season SO_2							
R^2	0.31	0.74	0.60	0.25	0.69	0.29	0.23
a	6969.2	16861.9	10241.0	–1861.3	31409.7	6720.8	6676.7
b	–3.42	–0.09	–0.12	0.26	–0.04	–0.09	–0.07
t	–0.20	–1.24	–0.73	0.14	–1.05	–0.18	–0.13
Winter Season PM_{10}							
R^2	0.32	0.60	0.04	0.07	0.26	0.24	0.01
a	–7511.9	12484.9	2005.0	–2124.9	–5509.4	–3333.1	–747.1
b	3.82	–0.10	–0.04	0.07	0.09	0.14	0.02
t	0.22	–0.71	–0.01	0.02	0.15	0.14	0.00
Winter Season T							
R^2	0.13	0.10	0.10	0.08	0.16	0.06	0.04
a	–132.3	–94.7	–149.9	–78.3	167.7	–69.8	–68.4
b	0.07	0.05	0.08	0.04	0.09	0.04	0.04
t	1.61	1.41	1.39	1.21	1.83	1.00	0.87

3.4Mann–Kendall, Spearman's Rho, and Sen's methods

When |Z| value was greater than 1.96, the null hypothesis of no trend was rejected according to the Mann–Kendall test. Hypotheses were accepted since $|Z_D|$ values were above 2.08according to the results of Spearman's Rho test.

The Mann–Kendall Z and Spearman's Rho $Z_D$values of the annual SO_2 concentrations showed the wide ranges from –4.83 to 0.32 and –4.06 to 0.47, respectively. Results of the Mann–Kendall and Spearman's Rho tests confirmed constant reductions in the annual SO_2concentrations across the years with negative slope values observed consistently except Aydin (Table 4).

The Mann–Kendall Z values of the annual PM_{10} concentrations were ranged from –3.80 to 2.70. The Spearman's Rho $Z_D$values of the annual PM_{10} concentrations showed the wide ranges –3.37 to 2.48. Increasing annual trends of the PM_{10} concentrations were observed in Denizli and Kutahya. Constant reductions were taken place in Izmir in the annual PM_{10}concentrations with negative slope of –3.80. No statistically significant annual trend was detected for the PM_{10} concentrations at the other cities (Table 4).

The Mann–Kendall Z and Spearman's Rho $Z_D$values of the annual temperatures ranged from 0.85 to 3.15 and 1.13 to 3.07, respectively. Increasing annual trends of the temperatures were determined in Denizli, Izmir, Afyon, Aydin, and Kutahya(Table 4).

Table 4. Mann–Kendall, Spearman's Rho, and Sen's tests for the annual SO_2 and PM_{10} concentrations and temperatures

	Denizli	Izmir	Afyon	Aydin	Kutahya	Manisa	Usak
Annual SO_2							
Mann–Kendall Z	-3.21	-4.83	-2.92	0.32	-3.73	-2.50	-2.11
Spearman's Rho Z_D	-2.87	-4.06	-2.54	0.47	-3.39	-2.34	-2.16
Sen's slope	-2.702	-4.737	-2.373	0.057	-7.765	-1.622	-2.979
Annual PM_{10}							
Mann–Kendall Z	2.73	-3.80	1.52	-0.16	2.24	1.27	-0.10
Spearman's Rho Z_D	2.48	-3.37	1.74	0.54	2.47	1.41	0.51
Sen's slope	2.730	-3.925	0.868	-0.104	1.927	0.711	-0.260
Annual T							
Mann–Kendall Z	2.67	2.16	2.96	2.06	3.15	0.85	1.24
Spearman's Rho Z_D	2.47	3.04	2.79	2.40	3.07	1.13	1.47
Sen's slope	0.068	0.054	0.100	0.050	0.096	0.025	0.034

The Mann–Kendall Z values of the winter season SO_2 concentrations ranged from –4.69 to 2.56. The Spearman's Rho $Z_D$values of the winter season SO_2 concentrations were taken part in the ranges from –3.96 to 2.54.There were downward trends for the winter season SO_2 concentrations in Denizli, Izmir, Afyon, Kutahya, and Manisa. An increasing trend of the SO_2 concentrations was observed in Aydin in winter season. No statistically significant trend was detected for the winter season SO_2 concentrations in Usak(Table 5).

The Mann–Kendall Z and Spearman's Rho $Z_D$values of the winter season PM_{10} concentrations were –3.85 to 2.03 and –3.27 to 2.32, respectively. No trend of the winter season PM_{10} concentrations was observed in the cities except Izmir and Denizli. Decreasing trend of the PM_{10} concentrations in winter season was apparent with negative slope of –3.85 in Izmir. The winter season PM_{10} concentrations showed a clearly increasing trend in Denizli with positive slope of 2.03. The PM_{10} concentrations in winter season in Kutahya showed upward trend according to the Spearman's Rho method with the slope of 2.32(Table 5).

The Mann–Kendall Z and Spearman's Rho $Z_D$values of the winter season temperatures ranged from 0.85 to 3.15 and 1.13 to 3.07, respectively. No trend was observed for the temperature values in winter season (Table 5).

Table 5. Mann–Kendall, Spearman's Rho, and Sen's tests for the winter season SO_2 and PM_{10} concentrations and temperatures

	Denizli	Izmir	Afyon	Aydin	Kutahya	Manisa	Usak
Winter season SO_2							
Mann–Kendall Z	-2.59	-4.69	-3.43	2.56	-4.13	-2.42	-1.44
Spearman's Rho Z_D	-2.25	-3.96	-3.11	2.54	-3.52	-1.97	-1.47
Sen's slope	-3.400	-7.200	-5.980	0.851	-15.646	-2.200	-2.200
Winter season PM_{10}							
Mann–Kendall Z	2.03	-3.85	-0.63	0.07	1.65	1.61	-0.56
Spearman's Rho Z_D	2.26	-3.27	-0.54	0.51	2.32	1.53	0.27
Sen's slope	2.120	-6.667	-1.343	0.094	2.857	1.469	-1.050
Winter seasonT							
Mann–Kendall Z	0.77	0.91	0.70	0.46	1.51	0.63	0.21
Spearman's Rho Z_D	0.88	0.77	0.66	0.47	1.49	0.54	0.15
Sen's slope	0.024	-0.042	0.041	0.025	0.066	0.030	0.012

3.5 Discussion of the air pollution in the Aegean Region

Fig. 3 shows the long–term trends obtained using the methods of linear regression, Mann–Kendall, Sen's, and Spearman's Rho for the annual and winter season SO_2 and PM_{10}concentrationsinDenizli, Izmir, Afyon, Aydin, Kutahya, Manisa, and Usakduring 1990-2009 period. It can be said that air pollution has components such as regional pollutant sources, anthropogenic sources, and topographic structure. Reasons ofthe air pollution can be listed as low–quality fuel use, topographic structure, unplanned urbanization, industrial facilities, and traffic. Importance level of these reasons differs among cities.

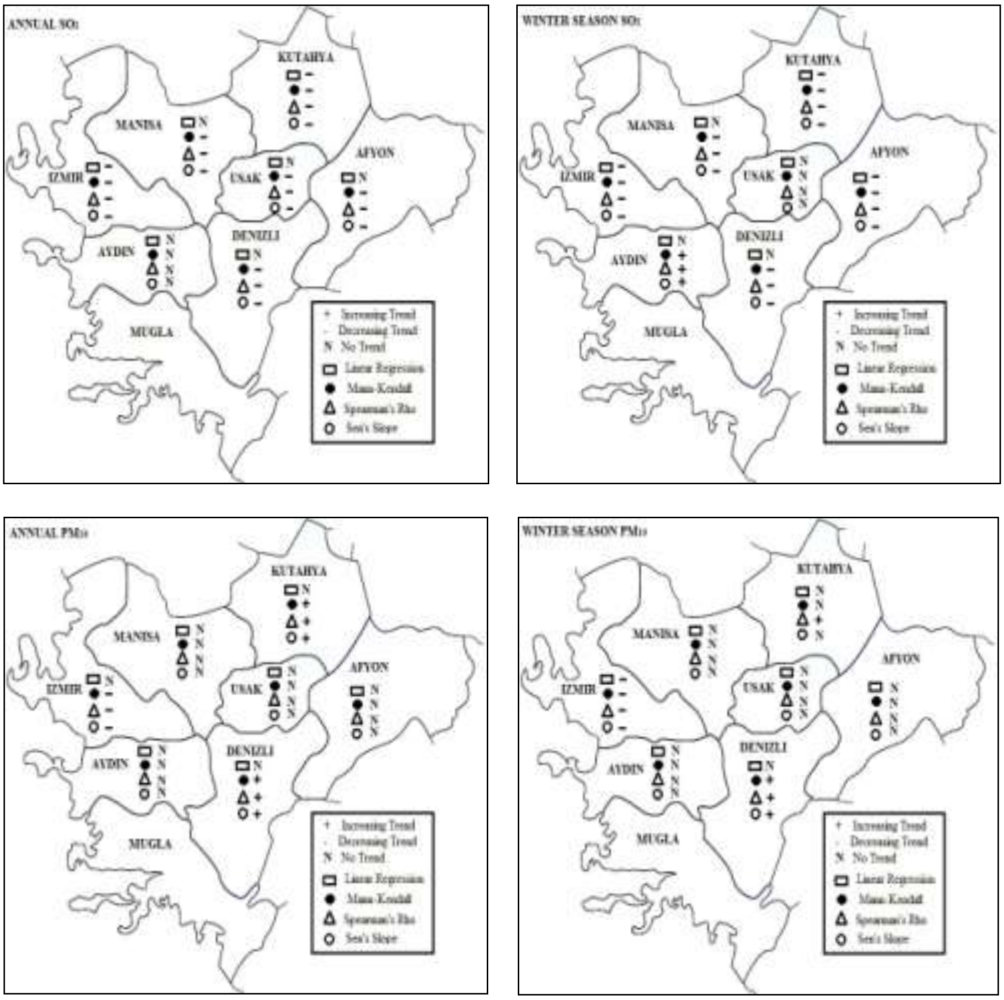

Fig. 3 Long–term trends of the annual and winter season concentrations of SO_2 and PM_{10}between 1990 and 2009

In some parts of the world there are studies that have been carried out in relation to trend analysis of air pollutants. Long–term analysis airborne particulate matter in Yongsan, Korea between 2004 and 2013 indicated a consistently decreasing trend throughout the decadal period, whereas PM_{10} exhibited noticeable decreasing concentrations (23.3%) in the latest years [16]. The seasonality of CO was characterized by wintertime maxima while for methane and non–methane hydrocarbons the highest amount fractions were found in fall in Seoul, Korea between 2004 and 2013. The analysis of their long–term trends based on Mann–Kendall and Sen's methods showed an overall increase of total hydrocarbons and methane, whereas a decreasing trend was observed for CO and non–methane hydrocarbons [17]. The time variability and long–term trends of mean annual levels of $PM_{2.5}$ at Montseny, and various regional background sites in Spain and Europe were studied and interpreted by Cusack et al. [18]. Reductions recorded in $PM_{2.5}$ across Europe were varied year–on–year decreases. Reductions in $PM_{2.5}$ were observed across all stations in Spain and Europe to varying degrees (7–49%). Montseny underwent a statistically significant reduction since measurements began. Similar trends were determined in other regional background sites across Spain. Zhang et al. [19] analyzed the data of SO_2, PM_{10}, CO, nitrous oxides, total suspended particulate, Pb, and benzo[a]pyrene from 1983 to 2007 in Beijing using the Daniel trend test. The results showed that SO_2 and PM_{10} concentrations had significant decreasing trends over the 25 year period. Long–term measurement results of ambient SO_2 and nitric acid at the Mt. Waliguan Observatory, a World Meteorological Organization/Global Atmosphere Watch global baseline station in China was investigated by Lin et al. [20]. Han et al. [21] concluded that the occurrence of haze continuously and rapidly increased in China during the period 1961–2012. The annual–average hazy days for all stations increased from approximately 4 days in 1961 to about 18 days in 2012, with an annual average growth rate of 3%. SO_2 concentrations had a very significant decreasing trend in 1997–2002, but a significant increasing trend in 2003–2009. Zhao et al. [22] determined that daily average PM_{10} concentrations of the 25 cities at the Yangtze River Delta Region, China between 2005 and 2009 ranged from 52.8 to 112.1 μgm^{-3}. PM_{10} levels showed a decreasing trend for the major cities during this period. SO_2 concentrations in Istanbul, Turkey were investigated using linear regression analysis during the heating season from 1985 to 1991. The results indicated that there was increasing trend in the SO_2 concentrations [23]. The study performed in Ostrava, Czech Republic showed that the computational models based on the strong correlations between air pollutants contribute to more cost–effective solutions for air pollution monitoring in cities, optimization of the network of monitoring stations, and the best selection of measuring devices [24].

The annual and winter season SO_2 concentrations in Denizli showed decreasing trends, while increasing trends of the annual and winter season PM_{10} concentrations were observed. Air pollution is an important environmental problem for Denizli. The main sources of this pollution are fuels used for heating purposes, exhaust gases of motor vehicles, and fuels used in the industry. Natural gas was started to be used in industrial facilities in 2005 for the purpose of production and heating in houses in 2006. There is a rapid and continuous increase in the number of vehicles registered to Denizli traffic. The size of air pollution is increasing by distorted urbanization and rapid industrialization day by day. Industrial air pollution in Denizli is sourced from general pollutants originating from stack gases, and organic and inorganic pollutants according to the type and shape of the industry. The sectors that cause air pollution may bemetal, textile, chemical, and cement industries. Denizlicity is surrounded by mountains on three sides. For this reason, polluted air is not dispersed and it is concentrated on the city [25].

Decreasing patterns for the annual and winter season concentrations of SO_2 and PM_{10} were determined during 1990 to 2009 in Izmir. When SO_2 and PM_{10}, which are two important parameters for determining the quality of air in Izmir, are examined, it is observed that the concentration of the pollutants originating from the heating is concentrated in the regions where the settlements are predominant. The districts where the air pollution caused by the heating is the most concentrated are the central districts and air pollution is experienced in Izmir especially when the use of poor quality fuel and the air movements are the lowest. Elbir[26] expressed poor meteorological conditions such as the stable atmospheric stratification, low wind speed, and a ground–based inversion for the efficient mixing of air pollutants occurred during winter season in Izmir. The traffic in the city center can contribute to air pollution. Traffic regulations are made in Izmir to reduce air pollution caused by traffic. Industrial air pollution originates from the districts of Aliaga, Bornova, and Kemalpasa where the industrial facilities are concentrated. Aliaga District is the most prominent with heavy industrial investments in the county where air pollution is the most experienced in Izmir [27]. Sari and Bayram[28] stated that the eastern part of Izmir was the less polluted than other areas in terms of domestic heating emissions (PM_{10}, SO_2, CO, nitrogen dioxide, and volatile organic compounds) during 2008–2009 winter season.

In Afyon, the annual and winter season SO_2 concentrations tend to decrease. No trend was observed for the annual and winter season PM_{10} concentrations. Afyon is among the cities with high concentrations of PM_{10} and SO_2 due to the increase of emissions from the chimneys especially during winter months. The important reasons for the air pollution are the use of topographical structures, combustion systems, and the use of domestic coal. Coal, geothermal energy, heating oil, and wood are used as fuel at the houses. Natural gas was not used for heating in the period of 1990–2009. The diversity of the fuels used for heating together with the incidents of immigration decreased the air quality and the air pollution rate increased [29].

The annual SO_2 concentrations in Aydin showed no trend. Increasing trend of the winter season SO_2 concentrations was observed. No trend was observed for the annual and winter season PM_{10} concentrations. The annual SO_2 concentrations did not change as there was no significant change in sources such as industrial facilities that show air pollutants throughout the year. There was increasing trend in the winter season SO_2 concentrations due to the use of fuel. Coal, fuel oil, and wood are used for heating at the houses in Aydin. During 1990–2009 period natural gas was not used for heating. Aydin is included to second stage polluted cities for SO_2, and PM_{10}. This is not a risk for air pollution because of the geographical structure (due to the fact that the mountains generally extend to the sea and the western–eastern winds from the sea dominate) in Aydin. The industrial establishments in Aydın are not concentrated in the city center. In this respect, the air pollution created by the industry is localized. There are many soil facilities, olive, and olive oil factories in Aydin center. Most of them burn coal at low quality, so these factories also contribute to air pollution. Exhaust gases from vehicles such as cars, minibuses, and buses used in urban transportation are seen as an important element of air pollution [30].

The annual and winter season SO_2 concentrations tend to decrease in Kutahya. The annual PM_{10} concentrations showed increasing trend. There was no trend in the winter season PM_{10} concentrations. Increase in the annual PM_{10} concentrations indicated that PM_{10} pollution sources increase in other seasons. Kutahya is among the first degree polluted cities in terms of air pollution. It has been determined that the SO_2 concentration in Kutahya has reached very high levels especially in winter period. Natural gas usage started in 2009 in Kutahya. In addition to natural gas, solid fuels such as wood, coal are also used for heating purposes. Used low quality (low calorific and high sulfur) coal contributes to SO_2 pollution. Sugar factories in Kutahya and thermal power plants located close to the city center could be played an important role in the increase of concentrations of SO_2 and PM_{10}. Tile factories operating in the summer and using pulverized coal as fuel, bread ovens using fuel oil or coal as fuel in many places in the city center also contribute to SO_2 concentration [31].

The annual and winter season SO_2 concentrations in Manisa showed decreasing trends. There was no trend in the annual and winter season PM_{10} concentrations. The most important reason for the problem of air pollution in winter months in Manisa, which is one of the leading domestic coal production centers of our country, is the use of solid fuel in large quantities for heating purposes. On the other hand, the current air pollution in Manisa is close to the average in Turkey. Among the causes of air pollution in Manisa, topographic structure, meteorological conditions, population density, and unplanned urbanization are important. The Spil Mountain, which rises quite steeply to the south of the city, blocks the air currents, but the inversion phenomenon frequently observed in winter months causes the air pollutants to hang on the city. Thermal power plant located in Soma, Manisa adversely affect the quality of the air. Industrial facilities in the organized industrial region started to use natural gas as of April 2003. In addition, the exhaust gas from vehicles is another source of the air pollution [32].

Decreasing trend of the annual SO_2 concentrations in Usak was observed. No trend was observed for the annual and winter season PM_{10} concentrations, and winter season SO_2 concentrations. It can be considered that the annual SO_2 concentrations showed decreasing trend due to reasons such as decrease or improvement in the sources of air pollution which are continuing throughout the year, such as industrial facilities. Usak is among the first degree polluted cities in terms of air pollution. One of the main reasons for air pollution in winter is the use of low quality fuel. Natural gas is being used in Usak since 2005 for heating purposes. There are two organized industrial districts in Usak. Fuels used in industrial facilities cause air pollution. The Afyon–Izmir highway passes through the center of Usak city. Motor vehicles that use roads linking the inner parts of our country and the western shores lead to the formation of air pollution [33].

IV. CONCLUSION

The long–term trends of the annual and winter season SO_2 and PM_{10}concentrations in the period of 1990 and 2009 in the Aegean Region cities Denizli, Izmir, Afyon, Aydin, Kutahya, Manisa, and Usak were investigated. Linear regression analysis, Mann–Kendall, Sen's, and Spearman's Rho methods were used to determine trends.

Linear regression analysis showed that decreasing trends of the annual SO_2 concentrations were observed in Izmir and Kutahya. The winter season SO_2 concentrations in Izmir, Kutahya, and Afyon showed downward trends. There was no trend for the annual PM_{10} concentrations. In Izmir, the PM_{10} concentrations in winter season showed decreasing trend.

According to the results of the Mann–Kendall, Sen's, and Spearman's Rho tests, the annual SO_2 concentrations had downward trends in all cities except Aydin. Decreasing trends in the winter season SO_2 concentrations were observed in Denizli, Izmir, Afyon, Kutahya, and Manisa. There was upward trend in Aydin. The annual PM_{10} concentrations showed increasing trends in Denizli and Kutahya, and decreasing trend in Izmir. For the winter season PM_{10} concentrations there were upward trend in Denizli, and downward trend in Izmir.

In order to decrease ofSO_2 and PM_{10} concentrations in the cities in the Aegean Region measures such as increasing the use of natural gas, and public transport. Heat insulation must be done in new or existing buildings. Improvement of existing facilities could be provided to reduce air pollution. Emission control systems must be installed. Industrial plants should be monitored continuously.

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SibelCukurluoglu "Trend Analysis of the Sulfur Dioxide and Particulate Matter Concentrations in the Aegean Region, Turkey "The International Journal of Engineering and Science (IJES),), 7.9 (2018): 64-74