تحديد التغير المكاني لملوثات الغلاف الجوي بمكة المكرمة

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Quantifying temporal trends of atmospheric pollutants in Makkah

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Abstract There is a high interest in quantifying temporal trends in air pollutant concentrations as they serve to assess the effects of emission control strategies. In this paper temporal trends (1997 – 2012) in air pollutant concentrations in the City of Makkah, near Al-Haram have been assessed with the help of TheilSen approach and changepoint analysis, which are applicable to both normal and non-normal distributed data and are therefore preferred over parametric statistics. Trend analysis of nitrogen oxides (NOx), nitric oxide (NO), nitrogen dioxide (NO_2), carbon monoxide (CO), sulphur dioxide (SO_2), particulate matter with aerodynamic diameter of 10 micron or less (PM_{10}) and ozone (O_3) was performed not only at mean concentrations but also at several selected quantiles (Minimum, first quartile, median, third quartile and maximum) . TheilSen function revealed significant positive trends in SO_2, NO_2, PM_{10} and ozone concentration and negative trends in CO and NO. Trends vary at various metrics suggesting different behaviour of air pollutants at various quantiles of the distribution. Reasons for the reported temporal trends are discussed. Further work on source apportionment of various emission sources and their temporal trend is required, which will provide further insight into the causes behind the trends of air pollutants and help better manage air pollutant levels in Makkah and elsewhere in the country. **Keywords:** temporal trends, Makkah, Saudi Arabia, air pollutant trends, air quality.

1. Introduction

Increased levels of atmospheric pollutants have adverse effects on human health (Walters and Ayres, 2002; Brunekreef and Holgate, 2002), affect plants growth and reduce crop yields (Bell and Treshow, 2008), and have negative effect on ecosystems (Cape, 2008). Urban ecosystems in large agglomeration and the adjacent surrounding areas are more affected by traffic related air pollutants, such as nitrogen oxides (NOx) and particulate matter. Air pollutants, particularly sulphur dioxide (SO_2) and nitrogen dioxide (NO_2) can lead to the formation of secondary air pollutants, for instance sulphate (SO_{4-2}) and nitrate (NO_{3}-) ions, which add to the adverse impacts of air pollution causing acid rains and visibility problems (Harrison, 2002). In Makkah most of the combustion related air pollutants (e.g., SO_2, NOx, CO, and Hydrocarbons) are predominantly emitted by road traffic (Habeebullah et al., 2012). In addition to their individual environmental and health impact, these may act as precursors leading to the formation of secondary air pollutants, such as ozone (O_3). Particulate matters including PM_{10} (particulate matter with aerodynamic diameter of 10 micron or less) in Makkah are emitted/generated by a wide range of sources. The main emission sources of PM_{10} in Saudi Arabia include road traffic, construction work, resuspension of dust particles, and windblown sand and dust particles (Khodier et al., 2012; Othman et al., 2010; Alharbi, 2009). Air pollutants (e.g., CO, SO_2 and NO_2) levels in Makkah are generally below the air quality standards set for the protection of human health by the Presidency of Meteorology and Environment (PME) of Saudi Arabia, except particulate matters. Due to the arid and hot atmospheric conditions and large exposed sandy deserts, the levels of particulate matter are high and often exceed the air quality limits (Seroji, 2011; Othman et al., 2010; Khodier et al., 2012). Sand and dust storms are common in Saudi Arabia and in the surrounding regions, which add to the atmospheric particulate matter (e.g., Kutiel and Furman, 2003; Barkan et al., 2004; Alharbi et al., 2012).

2

Temporal trend is a relationship between air pollutant concentration and time and is one of the most important and common tasks in the study of air pollution. Trends are calculated to have a general idea as to how the concentrations of air pollutants might have changed over time and/or to establish statistically whether a pollutant has significantly increased (positive trend) or decreased (negative trend) (Carslaw and Ropkins, 2012). There is a high interest in quantifying temporal trends in air pollutant concentrations as they serve to assess the effects of emission control strategies set by national and international organisations (Sicard et al., 2009; Monteiro et al., 2012). In this paper the temporal trends in several major pollutants in Makkah have been analysed, using non-parametric approaches, which are more suitable for air quality data due to their non-normal distribution. The aim is to quantify the temporal trends during the last 15 years or so and assess the impact of strategies for improving the air quality in Makkah implemented by the Kingdom of Saudi Arabia government. No published literature was found on temporal trend in Makkah and in the surrounding areas, therefore this is the first effort to quantify the temporal trends in air pollutant concentrations that could lead to further research in this area.

2. Methodology

This study intends to quantify temporal trends in several air pollutant concentrations in Makkah, Saudi Arabia. Air quality data of NOx (the sum of NO and NO_2), CO, SO_2, ozone and PM_{10} are analysed for about 15 years (1997 to 2012). The data were obtained from AQMS-112 (Figure 1) run by the Presidency of Meteorology and Environment (PME). The site is situated near the Holy Mosque (Al-Haram) as shown in Figure 1, which also shows the map of the surrounding areas, highlighting potential sources of emissions. This is a continuous monitoring site and measures the concentrations of several air pollutants (NOx, NO, NO_2, SO_2, CO, O_3 and PM_{10}) and meteorological variables. A summary of the data is shown in Table 1. As described in details in Munir et al. (2011) that air pollutant concentration is non-normally distributed and therefore non-parametric statistical tests need to be applied for statistical analysis. TheilSen (Theil, 1950; Sen, 1968) is a non-parametric approach and provides consistency between the p value and the uncertainty intervals in the slope and therefore has been preferred over the other test (e.g., Mann-Kendall test and linear regression) (Carslaw and Ropkins, 2012). Furthermore, TheilSen test tends to yield accurate confidence intervals even with non-normal data and non-constant error variance (homoscedasticity) and is resistant to outliers, as it is based on the median of the slopes. TheilSen test calculates slopes between all pairs of points and the median of the slopes is selected as TheilSen estimate, which is taken as the trend of the pollutant for the given period (Carslaw and Ropkins, 2012). Changepoint detection estimates the point where the statistical properties of a sequence of observations (time series) change (Killick and Eckley, 2011). Several algorithms are available for detecting changepoints in a time series data, including Binary Segmentation (Scott and Knott, 1974), Segment Neighbourhoods (Auger and Lawrence, 1989) and Pruned Exact Linear Time (PELT) search algorithm (Killick et al., 2011). In this analysis the latter was preferred due to the fact that it produces exact results quicker than the Segment Neighbourhood and Binary Segmentation algorithms and can be applied to both normally and non-normally distributed data. For details on these methods see Killick and Eckley (2011). Statistical data analysis was carried out in R - programming language (R Development Core Team, 2012) and its packages openair (Carslaw and Ropkins, 2012) and changepoint ((Killick and Eckley, 2011).

3

Table 1. A summary of the various air pollutants ($\mu g/m^3$)

NO2	NO	NOx	O3	CO (mg/m3)	SO2	PM10	Metric
0.00	0.00	0.00	0.00	0.00	0.00	0.00	1q 0

43.24	1.23	54.52	13.73	0.37	2.62	54.00	q 0.1
52.64	6.15	67.68	27.47	0.57	7.85	66.00	q 0.2
60.16	7.38	77.08	35.32	0.73	13.08	77.00	q 0.3
65.80	9.84	84.60	41.20	0.87	15.70	87.00	q 0.4
73.32	12.30	92.12	45.13	0.98	18.31	96.00	q 0.5
78.96	14.76	101.52	51.01	1.10	20.93	108.00	q 0.6
86.48	17.97	110.92	56.90	1.23	23.54	124.00	q 0.7
94.00	22.14	124.08	68.67	1.43	28.78	145.58	q 0.8
109.04	35.67	154.16	84.37	1.83	38.73	192.98	q 0.9
607.24	568.26	1466.40	979.04	48.31	690.62	821.00	q 1
80.10	24.49	119.03	49.26	1.14	20.69	112.83	Mean
95	95	95	96	95	79	88	%Data Capture

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[1] q stands for quantile, where q 0 represent minimum, q 0.5 represent median and q 1 represents maximum concentration. Furthermore, q 0.1 is equivalent to percentile 10 and q 0.2 is equivalent to percentile 20 and so on.

Figure 1.Map of the air quality and meteorological monitoring sites in Makkah (left), and detailed map of the PME site showing various sources of emissions. Blue circle shows construction area and red circles show bus stands.

3. Results and Discussions

Temporal trends in several air pollutant concentrations (PM_{10}, SO_2, CO, NOx, NO, NO_2 and O_3) have been analysed for the last 15 years in Makkah to determine how the concentrations of these air pollutants have changed with time. All pollutants are expressed in µg/m$_3$, except CO which is expressed in mg/m$_3$. Trends in PM_{10} concentrations at mean and various quantiles have been depicted in Figure 2, whereas the trends in other air pollutants are shown in Table 2. Temporal trends in NO and CO are negative, whereas the trends in PM_{10}, SO_2, NO_2, and ozone are positive. Figure 2 shows positive trends in PM_{10} concentrations at mean levels as well as at the selected quantiles (minimum, first quartile, median, third quartile, and maximum). PM_{10} trends are significant at all the selected metrics. The trends are higher for the higher quantiles and vice versa and range from 1.09 to 5.33 µgm$_{-3}$/year for minimum and maximum PM_{10} concentrations, respectively. SO_2, NOx, NO_2 and ozone show positive trends, which are significant at the selected metrics, except NOx which shows generally non-significant trends. NOx is the sum of NO and NO_2 and is affected by the levels of both NO, a primary pollutant emitted by combustion sources mainly road traffic in urban areas, and NO_2, a secondary pollutant formed in the atmosphere by the reaction of NO with ozone ($NO + O_3 \rightarrow NO_2 + O_2$) (e.g., Clapp and Jenkin, 2001; Jenkin, 2004). In Table 2, NO_2 shows positive and NO show negative trends, implying that NOx trends are affected more by NO_2 levels than by NO. NO_2 trend is lowest (1.33 µgm$_{-3}$/year or 1%) and highest (2.15 µgm$_{-3}$/year or 6%) at the maximal and minimal NO_2 concentration, respectively. The same pattern is shown by the NOx trend as well. Predominantly NOx emitted by road traffic is in the form of NO, therefore on roadside locations most of the NOx is present as NO, whereas in areas away from combustion sources predominantly NOx exists in the form of NO_2 due to the conversion of NO into NO_2 in the atmosphere (Munir et al., 2012). NO show negative trend and ranges from -0.46 to -1.53 µgm$_{-3}$/year at minimal and maximal levels, respectively. CO also shows negative trend and ranges from -0.03 to -0.07 mgm$_{-3}$/year. SO_2 has significant positive trend at all selected metrics, showing highest trend at third quartile (1.44 µgm$_{-3}$/year) and lowest at minimal level (0.71 µgm$_{-3}$/year). Similarly ozone concentration shows positive trend at most of the metrics, except at minimal level where trend was non-significant. Highest trend (1.47 µgm$_{-3}$/year) was shown at the maximal ozone level (see Table 2 for details). Negative trends in both NO and CO concentration is a good indicator of improvement in exhaust emission from road traffic, which suggests that despite the increasing number of road vehicles the amount of primary emissions have decreased due to stringent emission policies by the government of Saudi Arabia. Saudi Aramco took proactive initiatives to improve the environmental standards in Saudi Arabia by establishing an environmental master plan, which was endorsed in 2001 by the board of directors (Al-Zahrani, 2010). The plan addressed all contamination sources to the air, soil and water and intended to bring all facilities into compliance with government environmental regulations. Furthermore, unleaded gasoline was introduced in the country in 2001 enabling the use of catalytic exhaust after-treatment systems to reduce the emission of gaseous emissions. Al-Zahrani (2010) has reported a significant reduction in sulphur contents of the gasoline and diesel since 2005, due to strict government regulation to improve air quality in Saudi Arabia. However this is not reflected in SO_2 trends shown in Table 2. The results of the changepoint analysis are shown in Figure 3, which clearly shows that the concentrations of CO and NO have declined after 2001 – 2002, however SO_2 and PM_{10} shows rather increasing levels. Exact reasons for these trends are not known, however due to increasing construction activities and the use of crude oils (which has relatively higher sulphur content) in Makkah might be the possible reasons. Further work is required to confirm this statement.

Positive trends in the levels of NO_2 and ozone are reported by several authors (Cape, 2008; Jenkin, 2008; Derwent el al., 2007; Derwent et al., 2010; Carslaw, 2005; Carslaw et al., 2011). Carslaw et al. (2007) have reported increasing trends in the concentration of NO_2 in the UK and have shown that directly emitted (primary) NO_2 has increased as a fraction of total NO_X from road transport sources. These increases appear to be mostly due to certain after-treatment devices, such as oxidation catalysts and particle filters fitted to diesel vehicles. The gradual increase in average ozone concentration might have been caused by the hemispheric baseline ozone concentration resulting from global-scale effects (Derwent et al., 2007 and Jenkin, 2008). However, Jenkin (2008) and Derwent et al. (2007) have reported decreasing trends in maximum ozone concentrations, which is a disagreement with the trends reported here, which might have been caused by different meteorological conditions and precursor emissions observed in Saudi Arabia. The decreasing trends in peak ozone concentrations in the UK and Europe have been contributed to reductions in the emissions of anthropogenic volatile organic compounds (VOC) and NOx in the EU since the early 1990s (Jenkin, 2008). Kurokawa et al. (2009) reported an increasing ozone trends in Japan and stated that the increasing trend of boundary layer ozone was caused by the recent increase of anthropogenic precursor emissions in East Asia, especially in China. Furthermore,

5

meteorological conditions and titration of ambient ozone by decreasing NO emissions may play an important role in increasing ozone concentrations, particularly in urban areas.

Figure 2. Showing mean and various quantiles trends of PM_{10} concentrations in Makkah from 1997 to 2012.

6

Table 2. Showing trends at mean and various quantiles of the several air pollutants in Makkah during 1997 to 2012. Numbers in parentheses show percent trends, whereas those outside show trend in ugm^{-3}/year. Trends in CO concentrations are given in mgm^{-3}/year.

Minimum	Maximum	Third Quartile	Median	First Quartile	Mean	Pollutants
1.09(2)*	5.33(3)*	4.19(4)*	3.3 (4)*	2.47 (4)*	3.56 (4)*	PM10
0.71(6)*	1.33(4)*	1.44(10)*	1.31(12)*	1.22(18)*	1.22(10)*	SO2
-0.03(-4)*	-0.07(-3)*	-0.04(-3)*	-0.04(-3)*	-0.03(-3)*	-0.04(-3)*	CO
1.61(2.95)*	0(0)ns	0.94(0.93)ns	0.92(1)ns	1.03(1.39)ns	0.66(0.72)ns	NOx
-0.46(-7)*	-1.53(-3)*	-0.52(-3)*	-0.54(-4)*	-0.49(-4)*	-0.62(-4)*	NO
2.15(6)*	1.33(1)ns	1.63(2)*	1.68(3)*	1.7(3)*	1.55(2)*	NO2
0(0)ns	1.47(3)*	1.38(3)*	1.3(3)*	1.36(4)*	1.33(3)*	O3

'*' stands for significant and 'ns' stands for non-significant trend

Figure 3. Changepoint analysis of CO, SO_2, NO and PM_{10} concentration in Makkah from 1997 to 2012.

4. Conclusion

TheilSen function and changepoint analysis indicate increasing trends in ozone, SO_2, NO_2 and PM_{10} concentrations. In contrast, NO and CO have demonstrated decreasing trends over the study periods (1997 – 2012) in Makkah. The positive trend in ozone concentration is most probably due to the regional transport of its precursor and decreasing NO scavenging effect at local levels. Positive trend in NO_2 concentration is probably due to the increasing emission of primary NO_2 from diesel vehicles and certain after-treatment devices, such as oxidation catalysts and particle filters fitted to diesel vehicles. SO_2

7

contents of gasoline and diesel fuels have reduced, however this is not reflected in the atmospheric SO_2 concentration, probably due to the crude oil burning and increasing number of diesel vehicles. PM_{10} concentrations in Makkah have increased significantly probably caused by the increasing construction activities near Al-Haram during the last several years. NO and CO have shown negative trends in spite of the increasing number of vehicles in the city, which might suggest low emission from traffic vehicle as a results of stringent emission regulation. Further work is recommended into the source apportionment and their temporal trends, which will provide further insight into the trends of air pollutants and the factors behind them. **Acknowledgement** We are grateful to the custodian of the Two Holy Mosques Institute for Hajj and Umrah Research, Umm Al-Qura University for funding and to the PME for providing data for this project. We are also thankful to the staff for of the HRI for their help and support.

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8

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