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د. إبراهيم بن عبدالمجيد حسن

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Spatial Distribution and Temporal Variation in ambient ozone and its associated NO_x in Jeddah City atmosphere, Saudi Arabia

I.A.Hassan[1,2*] , Basahi, J.M.[2,3] Ismael, I.M. [2,4]

1. Faculty of Science, Alexandria University, 21526 El Shatby, Alexandria. EGYPT.

2. Centre of Excellency in Environmental Studies (CEES), King Abdulaziz University, 80216, Jeddah 21589..

3.Department of Hydrology & Water resources Management, Faculty of Environment, Meteorology and Arid Land Agriculture, King Abdulaziz University, Jeddah, KSA

4.Department of Chemistry, Faculty of Sceince, King Abdulaziz University, Jeddah

*Corresponding author

ihassan_eg@yahoo.com / iagadallah@kau.edu.sa, Tel: +966599251910; Fax: +96626951674

Abstract: Concentrations of ambient Ozone (O_3) nitrogen oxides (NO_x) were measured continuously for a period of 12 months in the city of Jeddah from December 2011 to December 2012. Meteorological parameters, wind speed, temperature, and relative humidity were monitored as well. Concentrations of ground O_3 were found to be highly dependent on the NO*x* diurnal cycle and wind speed. Nitrogen oxides were found to exceed air quality standard especially in industrial site, while O_3 concentrations were found to exceed 40 ppb, average over 1 h, on more than 24% of the measured days in the rural sites, while they were exceeded 30% in all other areas.

O_3 and NO_x were inversely related; the highest average NO_x concentration (96 ppb) occurred in rural area downwind of a desalination factory while average O_3 concentrations peaked in the rural area upwind of a desalination factory up to 63.5 ppb and 72.6 in another rural area which we considered as background in the present study. The seasonal variations of O_3 were more distinct than NO_x. To the best of our knowledge, this is the first report providing comprehensive background information for air quality in an arid area in the developing world.

Keywords: *Ambient ozone; Nitrogen oxides; Seasonal variation; Air quality; Jeddah*

1. Introduction

Air Quality in Urban areas is affected mainly by photochemical oxidants (Han *et al.*, 2011). Moreover, air pollution is a challenging problem as it would hinder sustainable development all over the globe (Akimoto, 2003; Molina & Molina, 2004). Increased combustion of fossil fuels in the last century is responsible for the progressive change in the atmospheric composition. Air pollutants, such as carbon monoxide (CO), sulfur dioxide (SO_2), nitrogen oxides (NO_x), volatile organic compounds (VOCs), ozone (O3), heavy metals, and respirable particulate matter (PM2.5 and PM10), differ in their chemical composition, reaction properties, emission, time of disintegration and ability to diffuse in long or short distances (Kampa, & Castanas, 2008)
The relation between ozone and its two main precursors, NO_x (NO and NO_2) and volatile organic compounds (VOC), represents one of the major scientific challenges associated with urban air pollution (Sillman, 1999). Tropospheric O_3 is formed in presence of sunlight through the chemical interaction of reactive VOCs NO_x that results from high-temperature combustion of fossil fuels (Nevers, 2000, Pudasainee *et al.*, 2006). Episodes of elevated O_3 are commonly associated with anticyclonic weather (Hassan et al., 1995). The meteorological conditions associated with anticyclones such as high solar radiation, high temperature, low wind speed and low rainfall are favorable for tropospheric O_3 formation, and these are the conditions prevalent in KSA (Presidency of Environment & Meteorology, KSA, unpublished data). In addition to a complex system of chemical reactions, meteorological and topographical factors determine where ozone concentrations will be the highest. Maximum ozone concentrations often occur in locales more distance of the source emissions (Finlayson-Pitts & Pitts, 1997). Moreover, it is well known that the ozone concentrations in ambient air increase with increase in the intensity of radiation and temperature on the clear days (Nishanth *et al.*, 2012). Furthermore, the magnitude of ozone concentration variations is high in clear days than the cloudy days.
NOx is Produced formed in the atmosphere from both natural and anthropogenic sources, involving naturally occurring nitrogen and volatile organic compounds (VOCs) by the action of lightning (Brown *et al.*, 2006). However, NO_x emissions from anthropogenic sources exceed natural sources (Godish, 2004). Increased combustion of fossil fuels and exhaust fumes may be extremely pervasive, causing serious environmental degradation, illnesses and deaths (Ricciardolo *et al.*, 2004; Gurjar et al., 2008; Roberts *et al.*, 2012).
A thorough understanding of the relationships among O_3 and NO_x under various atmospheric conditions is urgently needed to improve our understanding of the chemical coupling among these pollutants (Clapp *et al.*, 2001; Mazzeo *et al.*, 2005; Chou *et al.*, 2006; Costabile and Allegrini, 2007, Nishanth *et al.*, 2012). Seasonal variation effects on gaseous pollutants are of great significance to the life span and cycle of any pollutant in the lower atmosphere (Khan and Salem, 2007)
Saudi Arabia is one of the many arid lands in the world (e.g. Arizona, Nevada, South Africa, Peru, etc.). Very little attention was paid to areas having the same arid environmental condition as Saudi Arabia to monitor and measure the actual air pollution concentration in the atmosphere. Statistical analysis of such data may indicate air quality standards which are drastically different from those of non-arid areas (Sabbak, 1994).
Industrial and vehicular emissions combine to give Jeddah a serious air pollution problem. Thousands of old vehicles crowd the city streets with poor regulation of emission levels, and the city's factories create additional environmental hazards (Kadi, 2009). Concentrations of heavy metals, gaseous air pollutants and particulate emissions far exceed internationally acceptable standards (Al-Jeelani, 2008; Hassan & Basahi, 2013). Although monitoring of ozone is well established in USA and Europe, there is no a comprehensive study showing its seasonal, temporal and diurnal distribution in arid regions. Moreover, there is no enough background information for air quality standards for Jeddah as for the whole kingdom.
This study was undertaken in order to fulfil this gap of knowledge, and to describe the key observation of both diurnal and seasonal variation of ambient O_3 and its precursor NO_x and

their variation with meteorological parameters in Jeddah. Moreover, we attempted in the present study to collect enough and significant data to estimate air quality trends in Jeddah city atmosphere. Different metrological parameters (ambient temperature, relative humidity and wind speed and direction) and their influence on the air quality were also considered in the discussion of the results.

2. **Methodology**

2.1. Study area

Jeddah is a coastal city lies at the Western coast of Saudi Arabia (N 21^0 67', E 39^0 15') 15 meter above sea level. Its population around 3.5 million, it represents one of the largest cities by population and industry in KSA. It has well established traffic networks because it is the maritime port of the kingdom, resulting in high population density and a dense urban and infrastructural network.

There were six monitoring locations chosen along transect representing different levels of urbanization in order to have a comprehensive study of spatial distribution and diurnal variation of O_3 and its precursors. The gradient extended over a distance of approximately 30 km within the boundaries of the city.

2.2. *Ozone monitoring*

Ground levels of O_3 were measured at all stations on a continuous daily basis and the automatic analyzers' response time was 1 min. O_3 concentrations were monitored by ozone automatic analyzers, operated on the principle of photometric detection of the specific absorption of UV light by ozone (O_3 analyzer Model 49i).

2.3 *NO_x monitoring*

Ambient NO_x concentrations were measured simultaneously with chemiluminescence NO - NO_2 - NO_x analyzer Model 42i, in the range of 0–50 ppb to 0 – 100 ppm internal Zero/span Assembly with a detection limit of < 0.4 ppb

2.4 Meteorology of the study areas

Unlike other Saudi Arabian cities, Jeddah retains its warm temperature in winter season, which can range from 25 °C at midnight to 37 °C in the afternoon. Summer temperatures are considered very hot and break the 40 °C mark in the afternoon dropping to 30 °C in the evening. Rain is very rare and usually falls in December and January. Humidity ratio is 45-53%.

2.5. Data analyses

Time series plotting techniques were used to visualize the seasonal, monthly, and diurnal patterns of NO, NO_2, NO_x and O_3 concentrations. Statistical Package for the Social Sciences (SPSS) version 18.0, Statistical Analysis System (SAS) 9.2, and MINITAB 16 software packages were used (Liu et al., 2003).

3. **Results**

3.1. Meteorological conditions

The principal meteorological conditions recorded in the present study were wind speed, wind direction, temperature, relative and humidity in order to evaluate the influence they might have on ozone concentrations.

Monthly mean values of daytime (08:00-20:00) temperature (^{o}C), relative humidity (%), and wind speed (km/h) at the station for January to December 2012 are shown in Figure ١.

It was found Maximum temperature was recorded in August (47.6^{o}C), while the lowest one was recorded during January (21.6^{o}C).

The highest relative humidity was recorded during July (56%), while the lowest one was recorded during January (24%). On the other hand, wind speed was very high during winter season (January - February) where it was 24 km/h) while during summer it showed the minimum speed (11 km/h).

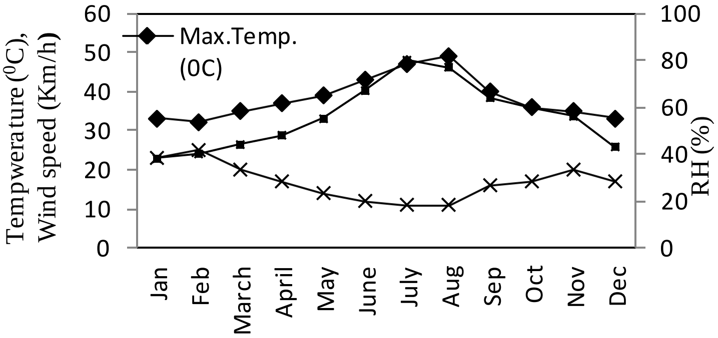

Fig 1. Monthly Mean Values of meteorological conditions averaged between the different monitoring sites within Jeddah during the period of the observation.

3.2. Diurnal variations of O_3 and NO_x

Twelve-hour averages (08:00–20:00 h, KSA Standard Time, NST) ground level ozone concentrations measured are presented in Fig. 1.

During the entire period of study the surface ozone concentration varied from the minimum of 20ppb in city centre and University premises sites in the morning to a maximum of 70 ppb at the rural site. The diurnal cycle of ozone was characterized by the maximum ozone concentration in the afternoon (between 14:00 and 16:00 hr) and minimum ozone concentration in the early hours of the morning (07:00 hr). A gradual decrease was observed in the evening hours (17:00 hr) in the study areas. After sunset, the concentration declining further and reached the lowest level between 20:00 hr and 07:00 hr.

The highest concentrations were recorded in Site 2 (averaged 54.9 ppb) which is a rural site while the lowest concentrations (averaged 25.1 ppb) were recorded in the city centre (site 4) "suburban area" (Fig.2).

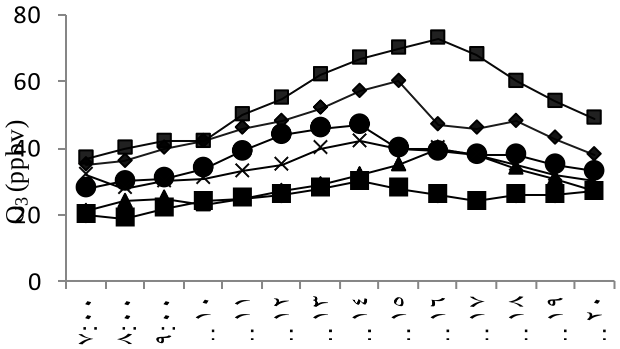

Fig.2. Diurnal variation ozone concentration (ppbv) during daytime (07:00 – 20:00 h) at different monitoring sites.

Diurnal variations in NO, NO_2 and NO_x concentrations are shown in Fig. 4. The Daily average peak emission of NO and photochemical formation of NO_2 and NO_x during the study period occurred at 7:00–9:00 h for sites 1, 5 and 6, 10:00–11:00 h for site 2 and 13:00–15:00 h for site 3, respectively. Site 4 "urban city centre" exhibited morning and evening peaks. The evening peaks occurred between 17:00 and 18:00 h, and it was higher in magnitude than morning peak which occurred between 08:00 and 09:00h.

In general, photochemical formation of ozone in air at a location is influenced by ambient temperature (R2 = 0.0257) and NO_x concentration (-0.372) (Table 2). It was clear that the ozone concentrations rises after 9:00 h and reached to peak value at about 14:00–16:00 h and then after it decreases slowly.

Table 2. Correlation coefficients between hourly averaged ozone Concentration, NO_x and temperature (a) whole time period (08:00–20:00 h) and (b) day light time period (07:00–18:00 h).

	Temperature	Wind speed	O_3	NO_x
(a)				
emperature	1	-0.512	).508	-0.254
Vind speed	-0.512	1	).297	0.071
O_3	0.508	0.297	1	-0.372
NO_x	-0.254	0.071	0.372	1
(b)				
emperature	1	-0.527	).378	-0.257
Vind speed	-0.625	1	).437	-0.108
O_3	0.378	0.437	1	-0.208
NO_x	-0.257	-0.108	0.208	1

3.3. Association of ozone with meteorological parameters and NOx

Figure 3shows that the ozone concentration is at peak when temperature is the maximum which indicates ozone concentration levels are directly related to temperature. There was a significant correlation between temperature and ambient O_3 concentrations (R^2= 0.508, P < 0.01) (Table 2).

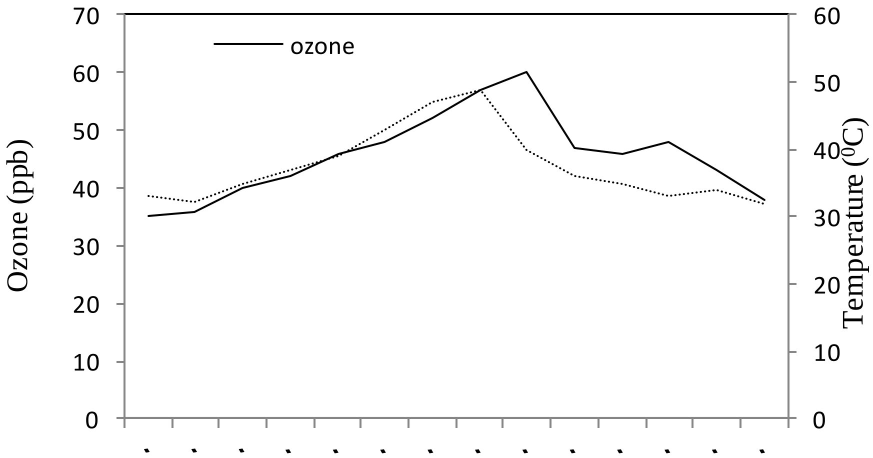

Fig.3 Relationship between ambient levels of ozone and temperature

Photochemical formation of ozone in air at a location is influenced by ambient temperature and NO_x concentration. It was clear that the ozone concentrations rises after 9:00 h and reached to peak value at about 14:00–16:00 h and then after it decreases slowly. The concentrations decrease steadily after sunset. Therefore correlation coefficients between ozone concentration and temperature, as well as ozone precursors were calculated separately for two periods: (i) entire measured time (07:00–20:00 h) and (ii) day light hours (10:00–16:00 h). Correlation coefficients between hourly averaged O_3 concentration and NOx for entire measured period and day light hours were -0.372 and -0.208, respectively. Similarly, correlation coefficient between hourly averaged O_3 concentration and temperature for entire measured time was 0.508. Moreover, multiple correlation coefficient between ozone and meteorological parameters (NOx, and temperature) was 0.74. Statistical details of correlation studies showed that correlation coefficients improved slightly for day light hours.

3.3. Site – to- site variations

Fig. 6 summarises the results for each site for each month of the year. It shows that the daylight-averaged data from site 1 (typically rural area) (51.1 ppb), site 2 (a rural area upwind of a desalination plant) (56.4 ppb), site 3 (downwind of a desalination plant) (30.6 ppb), site 4 (City Centre) (28.5 ppb), site 5 (University premises) (37.7 ppb) and site 6 (Industrial one) (34.1 ppb).

Whereas the local oxidant contributions at the two rural sites are comparable for most of the year, and were higher than the other four sites, the data provide clear evidence that the local source is substantially greater in midsummer (May - August) at all sites.

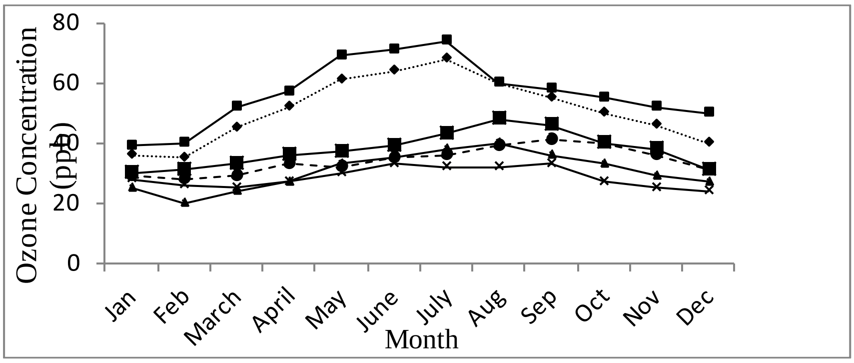

Fig.4. Seasonal variations of monthly average O_3 concentrations (ppb) at different sites.

3.4. Seasonal variations

Figure 5 shows variations in monthly average NO_x and O_3 concentrations. Average O_3 ranged from 11.1 ppb to 36.2 ppb, with wide distribution ranges from June through August, and increased in the winter.

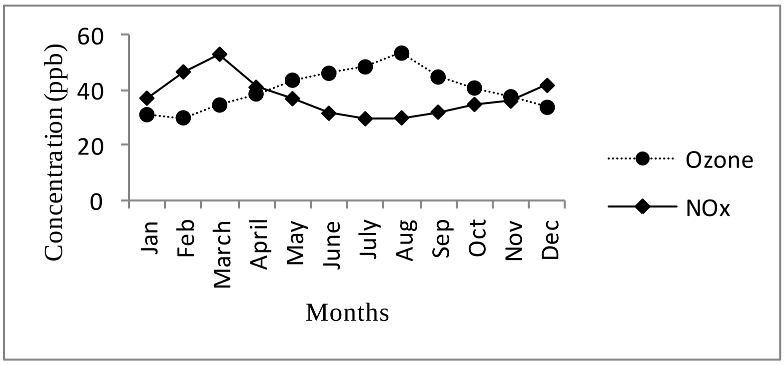

Fig.5. Annual cycle of O_3 and NO_x concentrations (averaged between different sites).

The annual average O_3 concentration was 40.3 ppb (averaged between different sites around Jeddah) while pronounced variations mainly in summer (between May to August). The daily mean O_3 values displayed summer highs with a gradual decline into autumn (Sep. to Nov.) and winter lows (Dec to Feb.).

The monthly NO_x concentrations ranged from 29.7 to 53.1 ppb, while the annual average was 37.9 ppb (averaged between all sites), with a pronounced an increasing trend towards the winter (Dec. to Feb.) to spring (March).

4. DISCUSSION

Air pollution monitoring in Saudi Arabia is extremely limited, moreover, there is a lack of interest and awareness regarding air pollution problems.

O_3 pollution has drawn much attention in Many Asian countries such as Hong Kong, Japan and China, in the last decade (Chan & Yao, 2008). However, O_3 pollution problems in Saudi Arabia and other Gulf countries were much less reported until recently. At present, ozone is measured in six studies in KSA; namely Jeddah (Sabbak, 1994), Makkah (Al-khalaf, 2006; Al-Jeelani, 2008;

2009; Seroji 2010). Dhahran and Jubail (Amin and Husain, 1994) and Riyadh (Al-Dhowalia et al., 1991). However, these studies were on fragmentary occasions.
Jeddah has numerous unregulated sources of particulates and gases (Sabbak, 1994). Most remarkable sources of pollutants are high number of vehicles, brick kilns, dusty roads and small industries. Our Previous study (Hassan & Basahi, 2013) indicated that most part of the city regularly experience total suspended particulates (TSP) and PM10 concentration levels above their acceptable limits set by World Health Organization (WHO) (Khodeir et al., 2012). Observed average ozone concentrations are within WHO guideline values of 60 ppb through most of experimental sites. The concentrations exceeded the guideline value for 8% of the days monitored. Based on EPA classification, during the study period, ozone concentration remained good for most of the sites for about 90% of days, moderate for 6.5% of the days, unhealthy for sensitive groups for 1% of the days for one site. These results are very similar to results of Pudasainee *et al.* (2006), who found similar results in Nepal. Recently, Reddy et al. (2012) stated that O_3 showed a well defined seasonal variation pattern on a diurnal scale with high levels (70.2 ± 6.9 ppbv) during the summer and low (20.0 ± 4.7 ppbv) during the monsoon with an annual mean of 40.7 ± 8.6 ppbv at a semi-arid rural site in Southern India.
The pronounced variations in summer observed in this study were very common in several studies (*Roberts–Semple et al.*2012). Nevertheless, our measurements showed that 12-h average ozone concentration lies in between 11.1 and 56.6 ppb, which are higher than the expected values. However, our results contradict results of Amin & Husain (1994), who reported higher concentrations of O_3 and NO_x in Dhahran in the eastern region of the kingdom. They found mean daily concentrations of these pollutants were 84 and 89 ppb, respectively, while they recorded the maximum hourly concentrations of same pollutants to be 181 and 222 ppb, respectively. One explanation for their high records could be attributed to the fact that they recorded these measurements immediately after Gulf war during the period of oil well fires.
The formation of ozone presents a strong relationship with meteorological conditions. In large urban centers, high emissions of ozone precursors are associated with the burning of fossil fuels by light- and heavy-duty vehicles. The resulting high concentrations of ozone in the atmosphere are harmful to ecosystems (EPA, 1997).
The mid-day peak and low nighttime concentrations of O_3 are typical characteristics of the diurnal cycle of ozone (Pudasainee *et al, 2006;* Han *et al.*, 2011*;* Roberts–Semple, *et al.*, 2012). The ozone concentration slowly rises after the sun rises, attains maximum during daytime and then again decreases until the next morning. This is due to photochemical O_3 formation. The shape and amplitude of ozone cycles are strongly influenced by meteorological conditions (temperature, solar radiation) and prevailing levels of precursors (NOx and HC). In the study area, daily cycle of NO level arising from vehicular emissions and its conversion to NO_2 possessed major impact on the daily cycle of ozone levels.
The diurnal variation of surface ozone is helpful to understand the different processes responsible for ozone formation and destruction at a particular location. It is regulated by chemical and atmospheric dynamic processes (Elampari &Chithambarathanu, 2011).
NOx concentrations increased rapidly during morning hours at the observation sites, which is due to the photochemical processes and emissions-dilution balance of NO_x and O_3, reflecting increased emissions of motor vehicles during the morning rush hours and also from industrial activities, especially at industrial site. During the noon hours, the solar radiation increased greatly and the photochemical processes that produce O_3 dominated, especially after the sunrise. Oxygen atoms produced in the photolysis of NO_2 could react with O_2 and to produce O_3 through the chemical reactions (Reddy et al. 2012).
The average NOx concentrations were much higher at a rural site downwind of a desalination plant, reflecting increased levels of vehicular and factory emissions. Conversely, average O_3 concentrations at this site were lower than others. The negative correlation in hourly

average NO_x and O_3 concentrations suggested that NO_x is the not the only factor contributed to elevated O_3 concentrations (Roberts–Semple, *et al.*2012).

CONCLUSIONS

The results indicate that the diurnal cycle of ozone concentration has a mid-day peak and lower nighttime concentrations. The ozone concentration slowly rises after the sun rises, reaching a maximum during the daytime and then decreases until the next morning. This is due to photochemical O_3 formation. The shape and amplitude of ozone cycles is strongly influenced by meteorological conditions and prevailing levels of precursors (NO_x). In the study areas, the daily cycle of NO concentration arises from vehicular emissions, and its conversion to NO_2, had a major impact on the daily cycle of ozone levels.

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