Trends of Ambient Concentrations of Gaseous Air Pollutants in Almadinah Al Menawwarah, Central Area, Saudi Arabia

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Abstract. Nitrogen monoxide (NO), nitrogen dioxide (NO₂), nitrogen oxides (NO_x), sulfur dioxide (SO₂), ozone (O₃), carbon monoxide (CO), methane (CH₄) and non-methane hydrocarbons (NMHCs) were measured in Almadinah Al Menawwarah, Saudi Arabia. From Ramadan to Hajj season (October, 2005 - January, 2006) daily average concentrations of NO, NO₂, NO_x, SO₂, O₃, CO, CH₄ and NMHCs, were 0.055, 0.040, 0.095, 0.011, 0.009, 2.82, 3.83 and 1.12 ppm, respectively. The SO₂/NO_x and SO₂/NO₂ concentration ratios were 0.12 and 0.28, respectively, suggesting that the mobile emissions are the predominant sources within the study area. The diurnal variations of NO, NO₂, NO_x, SO₂, CO, NMHCs concentrations were similar and showed two daily peaks, in the morning and late evening. These peaks concentrations are linked to traffic density, boundary layer mixing processes and chemical processes in the atmosphere. The diurnal cycles of O₃ concentrations revealed a uni-modal peak in the mid-day time. The highest concentrations of the measured pollutants were found in Ramadan and Hajj seasons, due to increasing human activities. Significant positive correlation coefficients ($p < 0.05$) were found between NO, NO₂, NO_x, NMHCs and CO, and also between SO₂ and NO, NO₂, NO_x, NMHCs and CH₄. Significant negative correlation coefficients were found between O₃ concentrations and NO₂. These results indicate that an increase in O₃ level is associated with a drop in the concentration of NO₂, CO and NMHCs.

Keywords: Gaseous Air Pollutants, Nitrogen Monoxide, Nitrogen Dioxide, Nitrogen Oxides, Sulfur Dioxide, Ozone, Carbon Monoxide, Methane, Non-Methane Hydrocarbon, Almadinah Al Menawwarah, Saudi Arabia.

Introduction

High rate of industrialization/urbanization process, especially in developing countries, leads to increase in the emission of many types of air pollutants due to fossil fuel combustion. Air pollutants, once emerged from a variety of sources are subject to mixing, dispersion, transport and complex series of chemical interaction and physical transformation processes in urban atmospheres. Nitrogen monoxide (NO), nitrogen dioxide (NO₂), sulfur dioxide (SO₂), carbon monoxide (CO) and volatile organic compounds (VOCs) are the most important groups of air pollutants in the urban atmosphere because they can cause significant risk to human health and play an important role in atmospheric photochemistry. These photochemical reactions transform primary air pollutants into secondary air pollutants. The environmental effects of these secondary pollutants are associated with acidification of precipitation, visibility reduction and have deleterious effects on human health and plants.

Oxides of nitrogen, NO_x (NO₂ and NO), contributes to a wide range of environmental effects including the formation of acid rain, particulate and gaseous nitrate and regional haze, eutrophication of aquatic ecosystems, and elevated ozone(O₃) concentrations with resulting impacts on health and agriculture (Matsumoto and Tanaka, 1996; Matsumoto, *et al.*, 1998; Khoder, 2002 and Mauzerall *et al.*, 2005). NO₂ is consistently associated with adverse health effects at relatively low levels of long-term average exposure(European Commission, 1997; WHO, 2003 and 2005). The major contributor to NO_x concentrations in urban areas is road traffic (Jraiw 2002; Shrestha *et al.*, 2005 and CARB, 2006), and it can be used as a tracer of road traffic emissions in urban areas (Lewne *et al.*, 2004). NO₂ is formed during the combustion of fossil fuel through the oxidation of atmospheric nitrogen and of nitrogen from certain fuels such as coal and oil (WHO, 1997). It originates from both primary and secondary sources (AQEG, 2004). It is mainly a secondary pollutant since in most ambient situations NO is emitted and transformed into NO₂ in the atmosphere through photochemical processes (Jenkin and Clemitshaw, 2000). However, some studies have shown that NO₂ is also emitted as a primary pollutant to a certain extent (PORG, 1997 and Carslaw and Beevers, 2004 and 2005).

The major health impacts of SO_2 include effects on breathing, respiratory illness, weakness of lung defenses, increase in the effects of existing respiratory and cardiovascular disease, and death (Henry and Heinke, 1996 and USEPA 2008). In addition, atmospheric oxidation of SO_2 by hydroxide radical (OH), O_3 and hydrogen peroxide (H_2O_2) contributes to acid deposition and sulfate formation (Finlayson-Pitts and Pitts, 2000 and USEPA, 2008). The major source of SO_2 is the combustion of fossil fuels containing sulfur, in particular from power stations burning coal and heavy fuel oil (Afif *et al.*, 2008). Although road transport is a minor source of SO_2 at the national level, it can be important in some urban areas, especially alongside busy roads (Holgate *et al.*, 1999).

Carbon monoxide plays an important role in tropospheric chemistry, as it serves to produce O_3 and affects O_3 concentrations by its effect on OH and HO_2 concentrations (Badarinath *et al.*, 2007). Atmospheric oxidation of CO leads to O_3 formation or destruction, depending upon the NO concentration (Warneck, 1988). The reaction of CO with OH radicals is the primary removal process from the atmosphere. Through this mechanism, CO acts as a major precursor to photochemical ozone (Crutzen, 1974). Biomass burning, combustion of fossil fuels, and oxidation of hydrocarbons are the major sources of atmospheric CO (Han and Naeher, 2006; Zimmerman *et al.*, 1978; Crutzen *et al.*, 1979; Brewer *et al.*, 1984; Hatakeyama *et al.*, 1991 and CARB, 2006). High concentrations of CO generally occur in areas with heavy traffic (Ali and Athar, 2008). The point sources of CO emission also include industrial processes, non-transportation fuel combustion.

Volatile organic compounds (VOCs), mainly non-methane hydrocarbons (NMHCs), are the key players in the formation of tropospheric O_3 and secondary organic aerosol (Seinfeld and Pandis, 2006). Sources of VOCs to the atmosphere are both anthropogenic and natural emissions. Major anthropogenic VOCs are emitted from traffic-related sources such as vehicle exhaust and gasoline evaporation and non-traffic related sources such as solvent use, natural gas leaks and liquefied petroleum gas (LPG), and industrial processes (Saito *et al.*, 2009).

Ground level O_3 is considered one of the most important photo-oxidants (secondary pollutant) that are produced through a series of photochemical reactions. It is a key precursor of OH radicals which

control the oxidizing power and chemical composition of the atmosphere (Logan *et al.*, 1981 and Thompson, 1992). The level of OH in the atmosphere in turn influences the levels of many primary air pollutants such as CH₄, CO and SO₂ (Poulida *et al.*, 1991). In addition, the elevated O₃ concentrations have a particular concern due to their deleterious effects on public health as well as damages ecosystems, agricultural crops and materials (Brönnimann *et al.*, 2002 and Wang *et al.*, 2003). Ground level O₃ is formed through a series of complex photochemical reactions among its precursors (NO_x, CO and VOCs) in the presence of sunlight and temperature (Seinfeld and Pandis, 1998; Derwent *et al.*, 2003; Abdul-Wahab *et al.*, 2005; Zhang *et al.*, 2004; Tie *et al.*, 2006; Alvim-Ferraz *et al.*, 2006 and khoder, 2009). Once formed, O₃ quickly reacts with NO regenerating NO₂ in the absence of VOCs. The high concentration of VOCs always increases O₃ formation, whereas increasing of NO_x leads to more or less O₃, depending on the prevailing ratio between (VOCs) and (NO_x) (Seinfeld and Pandis, 1998; Barros, 1999; Guicherit and Roemer, 2000; Sadanga *et al.*, 2003 and Pereira *et al.*, 2005). In the presence of VOCs, the oxidation processes of the VOCs play an important role in the oxidation of NO to NO₂ without consumption of O₃.

Air pollution in Saudi Arabia is now recognized to be of significant environmental problem due to the presence of intensive anthropogenic activities. Heavy traffic density in the streets of the urban areas leads to an increase in the emission of O₃ precursors (NO_x and VOCs), and the problem of pollution had been shifted towards the so called photochemical pollutants. The formation of those pollutants in the atmosphere of the urban areas in Saudi Arabia is facilitated by the local climatic conditions (high temperature, intense solar radiation, clear sky).

The assessment of air quality in urban environment, using the available air quality data will help generate information aiding in planning pollution control strategies to keep the pollutants within safe limits. Air quality data is necessary to understand the trends in depth that will lead to more realistic findings and conclusions. Most of the previous studies about air pollution in Saudi Arabia had been carried out in the Holy City of Makkah, focused on the central area near the Holy Mosque and on the Holy places (Mina, Arafat and Al-Taneem areas) (Al-Jeelani, 2009a and b, Al-Jeelani and Ramadhan, 2004 and 2005; Yacob, 2000; Al-Thumali, 1998; Badwi and Al-Hosary, 1993 and Al-Amri and Abu-

Alghat, 1992) and inside the tunnels near the Holy Mosque (Al-Jeelani, 1998; Al-Raddadi, 1996 and Al-Sawas, 1995). There is insufficient or lack of information about the air pollution levels in Almadinah Al Menawwarah atmosphere. Therefore, the present study aims to: (1) Evaluate the levels of NO, NO₂, NO_x, O₃, SO₂, CO, CH₄, and NMHCs from Ramadan to Hajj season (October, 2005 - January, 2006) in Almadinah Al Menawwarah, (2) study of the diurnal variations of the measured gases and factors affecting them, and (3) investigate the relationships between the measured gases.

Materials and Methods

Sampling Site

Al Madinah Al Menawwarah is a city in the Hejaz region of western Saudi Arabia. It is the second holiest city in Islam and it has a population of more than 1,300,000 people. It is located at about 340 km north of Makkah and ≈ 190 km from the Red Sea coast at a height of ≈ 600 m from the mean sea level. The area of Almadinah is ≈ 590 km², the area occupied by the city's urban area is ≈ 293 km². The area outside the urban area consists of mountains and valleys and slopes of Seoul and desert land and agricultural activities and parts of the network of highways. The climate of Almadinah is generally dry and characterized by high temperatures ranging from 28-42 C⁰ in the summer and 11-24 C⁰ in the winter. The relative humidity is low throughout the year except for rainfall periods and averaging $\approx 22\%$. The average annual rainfall is ≈ 3.94 mm and the city is exposed to northwesterly and westerly winds in general. The wind speed ranges between 9.26 – 14.8 km / h.

Air Sampling

Air quality monitoring mobile station (Environment SA, France) was used to conduct the measurements of pollutants including: NO, NO₂, NO_x, O₃, CO, SO₂, NMHCs and CH₄. Calibration was done daily and no data was recorded during that period. Average hourly measurements were taken in the period from Ramadan to Hajj season (October, 2005 - January, 2006). To study the effect of traffic density on the air quality of the study area, the data have been classified into seven periods: October 8th – November 2nd, 2005 (A), 3rd - 12th November, 2005 (B), November 13th - 26th, 2005 - November, 2005 (C), 20th – 31st December, 2005 (D), 1st

- 8thJanuary, 2006 (E), 9th -14thJanuary, 2006 (F), and 15th- 30thJanuary, 2006.

Statistical Analysis

The correlation coefficient (r) and the correlation significant t -test were determined using the alternative method of calculation (Gregory, 1963).

Results and Discussion

Air Quality at the Study Area during the Period of Study

Presented in Fig. 1 the daily average concentrations ranged from 0.020 to 0.124 ppm with an average value of 0.055 ppm for NO and from 0.018 to 0.070 ppm with an average value of 0.040 ppm for NO₂. The daily average concentrations of NO_x (NO + NO₂) ranged from 0.040 to 0.187 ppm with a mean value of 0.095 ppm. In the current study the average NO₂ concentration (0.055 ppm) was lower than those found in the city centre (Ramsis square) of Cairo, Egypt (0.117, 0.137, 0.100 and 0.089 ppm in the autumn, winter, spring and summer seasons, respectively, (Khoder, 2004) and in Dokki, Giza, Egypt (0.098 ppm) in the winter and 0.070 ppm in the summer season, Khoder, 2002). On the other hand, the average NO₂ concentration in the present study was higher than those found in Beijing (0.038 ppm), (Wang *et al.*, 2008) and in Al-Taneem area in the Holy Makkah city, Saudi Arabia (0.018 ppm), (Al-Jeelani, 2009a), in Germany (0.018 ppm), the Netherlands (0.020 ppm), Rotterdam (0.021 ppm) and Sweden (0.015 ppm), (Lewne *et al.*, 2004), in Kolkata, India (0.017 ppm), (Gupta *et al.*, 2008) and in the city of Eskisehir, Turkey (0.016 ppm) in winter and (0.008 ppm) in summer, (Özden *et al.*, 2008).

Fig. 1. The daily average concentrations of NO, NO₂, NO_x, O₃ and SO₂ during the period of study.

The average concentrations of NO_x (0.095 ppm) in the present study was lower than that found in holy Makkah city, Saudi Arabia, during Hajj season 1425 H (0.15 ppm), (Al-Jeelani, 2009b) and much higher than those found in Al-Taneem area in the Holy Makkah city, Saudi Arabia (0.033 ppm), (Al-Jeelani, 2009a) and in Mena area during Hajj season 1424 H (0.078 ppm), (Al-Jeelani, 2009).

The daily average concentrations of SO_2 (0.011 ppm) (Fig. 1) was nearly similar to those found in Al-Taneem area in the Holy Makkah city, Saudi Arabia (0.010 ppm), (Al-Jeelani, 2009a) and in Ankara, Turkey (0.011 ppm), (DRSCH, 2006). On the other hand, the average concentrations of SO_2 in the present study was lower than those found in Dokki, Giza, Egypt (0.048 ppm) in winter and (0.032 ppm) in summer, (Khoder, 2002), in Dhaka, Bangladesh (0.045 ppm) (Gurjar *et al.*, 2008) and in China (0.114 ppm), (Lei *et al.*, 1997) and relatively lower than those found in Mexico City, Mexico (0.018 ppm), (Gurjar *et al.*, 2008) and in Eskisehir, Turkey (0.018 ppm) (Özden *et al.*, 2008) in Izmir, Turkey (0.015 ppm), (DRSCH, 2006), in the Holy Makkah city, Saudi Arabia during Hajj season 1425 H (0.016 ppm) (Al-Jeelani, 2009b) and in Mena area during Hajj season 1424 H (0.015 ppm), (Al-Jeelani, 2009). On the other hand, the average concentration of SO_2 in the current study was much higher than those found in Beirut, Lebanon (0.003 ppm), (Afif *et al.*, 2008), in Roma, Italy (0.006 ppm) (Avino and Manigrasso, 2008), in Pamplona, Spain (0.003 ppm) (Zabalza *et al.*, 2007) in Thurroch, UK (0.002 ppm), (UK Air Quality Archive, 2008) and relatively higher than that found in New York, USA (0.009 ppm) (USEPA, 2008) and in Beijing (0.009 ppm) (Wang *et al.*, 2008).

The daily average concentrations of O_3 (0.009 ppm) in this study were much lower than those found in Giza (Haram), Egypt, during the winter (0.030 ppm), spring (0.048 ppm), summer (0.064 ppm) and autumn (0.043 ppm), (Khoder, 2009), and in the city centre of Cairo, Egypt, (0.040, 0.058, 0.079, 0.051 ppm) during the corresponding seasons, respectively (Khoder, 2004), in Eskisehir, Turkey (0.019 and 0.027 ppm) during the winter (0.019 ppm) and in summer (0.027 ppm), (Özden *et al.*, 2008) and in Al-Taneem area in the Holy Makkah city, Saudi Arabia (0.021 ppm), (Al-Jeelani, 2009a). In contrast SO_2 average concentration was higher than that found in the Holy Makkah city, Saudi Arabia during Hajj season 1425 H (0.004 ppm), (Al-Jeelani, 2009b).

The daily average concentrations of CO, CH₄ and NMHCs during the period of study are graphically presented in Fig. 2. The daily average concentrations of CO ranged from (1.18 to 4.82 ppm) with an average of (2.82 ppm). This average concentration was higher than those found in Beijing (1.97 ppm) (Wang *et al.*, 2008) in Al-Taneem area in the Holy Makkah city, Saudi Arabia (1.39 ppm) (Al-Jeelani, 2009a), and nearly similar to that found in Mena area during Hajj season 1424 H (2.45 ppm, Al-Jeelani, 2009). On the other hand, the daily average concentration of CO in this study was lower than that found in Holy Makkah city, Saudi Arabia during Hajj season 1425 H (11.30 ppm, Al-Jeelani, 2009b).

Fig. 2. The daily average concentrations of CO, CH₄ and NMHCs during the period of study.

The daily average concentrations of CH₄ and NMHCs in the present study were 3.83 and 1.12 ppm, respectively (Fig. 2). The daily average concentrations of CH₄ (3.83 ppm) and NMHC (1.12 ppm) were higher than those found in Al-Taneem area in the Holy Makkah city, Saudi Arabia (2.15 ppm) for CH₄ and (0.39 ppm) for NMHCs (Al-Jeelani, 2009a). In addition, the daily average concentrations of NMHCs in this study was higher than those found in Nagoya, Japan (0.028 ppm) (Saito *et al.*, 2009), London, UK (0.030 ppm) (Derwent *et al.*, 2000), Lille, France (0.022 ppm), (Borbon *et al.*, 2002), Seoul, Korea (0.052 ppm), (Na and Kim, 2001) and Dallas, USA (0.041 ppm), (Qin *et al.*, 2007).

Mobile emission sources are predominantly characterized by high NO_x and CO concentrations, and point emission sources by high SO₂ and NO_x. So, the mobile sources have relatively low SO₂/NO_x ratios (Aneja *et al.*, 2001). In the current study, the SO₂/NO_x and SO₂/NO₂ concentration ratios were 0.12 and 0.28 during the period of study, respectively, suggesting that the mobile emissions are the predominant source within the study area. The SO₂/NO₂ concentration ratio at the

study area during the period of study was lower than those found in Kolkata, India (ranged from 0.38 - 0.41) (Gupta *et al.*, 2008).

Diurnal Variations of NO, NO₂, NO_x, O₃, NMHC, CO, CH₄ and SO₂ Concentrations

The hourly variations of air pollutants provide valuable information on their sources, transport and chemical formation/destruction. Figure 3 shows the diurnal variations in NO, NO₂, NO_x and NMHCs concentrations during the period of study. The hourly concentrations of NO increased from 06.00 to 08.00 h, and then decreased in mid-day time. After that time, the concentration increased again to give a second peak in the late evening. The morning peak (0.062 ppm at 08.00 h) was lower in magnitude than that of the late evening one (0.089 ppm at 21.00 h). The trend of NO₂ hourly concentrations was similar to that of NO concentrations, with the morning peak was achieved at 08.00 h (0.043 ppm) and late evening peak at 20.00 – 21.00 h (0.052 ppm). The diurnal variations of NO_x concentrations was also similar to the trend of NO and NO₂, with double peaks, in the morning at 08.00 h (0.110 ppm) and late evening at 21.00 h (0.141 ppm). These double peak patterns of NO, NO₂ and NO_x may be the typical signature of urban influence boundary layer processes, surface wind pattern, emissions, transportation/work cycle and chemical processes. During the morning time, the increase in the emission rate from traffic, accompanied with a poorer dispersive conditions and lower photochemical reaction lead to accumulate the concentrations of air pollutants. Latha and Badarinath (2003) reported that high levels of NO_x during the morning and late evening can be attributed to combinations of anthropogenic emissions, boundary layer processes, chemistry as well as local sources and wind patterns. In the present study, the low concentrations of NO and NO₂ during the mid-day time may be due to the high dispersion and high dilution conditions, under the effect of high temperature, which increases the thermal turbulence currents. Moreover, the high temperature and solar radiation intensity during the mid-day time lead to increase the photochemical reactions and consequently increasing the chemical loss of those pollutants.

Fig. 3. Diurnal variations in NO, NO₂, NO_x and NMHCs concentrations during the period of study.

This leads to decrease the NO_x concentrations in the mid-day time. This corresponds with Rao *et al.* (2002a & b) who attributed that the variations in NO_x to variations in boundary layer mixing processes, chemistry, anthropogenic emissions, and local surface wind patterns. During the late evening and nighttime, the boundary layer descends and remains low until early morning, thereby resisting the mixing of anthropogenic emissions with the upper layer. Hence, pollutants get trapped in a shallow surface layer resulting in raising levels of NO, NO₂ and NO_x in the current study (Fig. 3).

It can be noticed that, the trend of NMHCs concentrations was similar to those of NO, NO₂ and NO_x concentrations (Fig. 3), whereas it was inverse with O₃ concentrations. The morning peak (0.99 ppm at 08.00 h) was lower in magnitude than that of the late evening peak (2.07 ppm at 21.00 h). This two-peak pattern had also been observed in many other sites, such as Changchun, China (Liu *et al.*, 2000); Taichung, Taiwan (Yang *et al.*, 2005); and Bilbao, Spain (Durana *et al.*, 2006). The pattern suggested that the dominant effect in the early morning and late evening was of traffic density, that resulted in the increase of exhaust from vehicles. The mid-day time minimum concentrations of NMHCs may be attributed to dilution processes induced by increasing in the mixing depth, reduced emissions and its depletion due to photochemical reactions. The major sink of hydrocarbons is the OH radical which reaches its peak at noon due to the high solar radiation and temperature.

The diurnal variations of O₃ concentrations during the period of study are graphically presented in Figure 3. O₃ showed a uni-peak pattern with the highest levels during the daytime and the lowest levels in the nighttime, with a uni-modal O₃ peak during the daytime. This is

attributed to higher temperature, and solar radiation intensity during the daytime, which are the favorable conditions to power the photochemical reactions. Minimum concentrations of O_3 appear in the nighttime and early morning hours (near sunrise) due to the combined effects of chemical loss by NO and NO_2 species and suppressed boundary layer mixing processes. During the morning, concentrations of NO_x are high, which leads to destruction of O_3 (Badarinath *et al.*, 2007). In the present study, the O_3 concentration increased after the sun rises and reached its maximum levels at 12.00 h, with a mean value of 0.021 ppm. After that time, O_3 concentration decreased again. Variation in O_3 concentration had a tendency to follow the solar radiation intensity, resulting in higher O_3 levels during the daylight periods. This higher level in O_3 concentration during the daylight periods is attributed to the photochemical processes of O_3 formation. On the other hand, as the sun goes down in the evening and nighttime, the photochemical processing of O_3 is halted due to the absence of the photochemical reactions, and the O_3 that remains in the atmosphere is then consumed by deposition and/or reaction with NO which acts as a sink for O_3 .

Diurnal variations of CO, SO_2 and CH_4 concentrations during the period of study are graphically presented in Fig. 4. They showed double peaks patterns. The hourly concentrations of CO increased from 06.00 to 08.00 h, and then decreased in the mid-day time. After that time, the concentration increased again to give a second peak in the late evening. The morning peak (2.85 ppm at 08.00 h) was lower in magnitude than that of the late evening peak (4.91 ppm at 21.00 h). These double peak patterns may be the typical signature of urban influence. The diurnal cycle of CO is related to the transportation/work cycle, metrological conditions and photochemical reactions. The boundary layer in the late evening descends and remains low until early morning, thereby resisting the mixing of anthropogenic emissions with the upper layer. This will lead to increase the accumulation of CO in a shallow surface layer and consequently, the level of CO in that time is increased. During morning and late evening, the highest levels of CO may be attributed to combinations of anthropogenic emissions, boundary layer processes, chemistry as well as local sources and wind patterns (Latha and Badarinath, 2003). During nighttime, formaldehyde (HCHO) accumulates while during the morning its chemical reaction and photo dissociation results in the production of CO (Badarinath *et al.*, 2007). In the present

study, the observed diurnal cycle of CO was inversed of that of O₃ (Fig. 1). The reaction of OH is the primary mechanism for the removal of CO from the atmosphere. Major chemical loss of CO is by reaction with OH radical during daytime, producing ozone (Badarinath *et al.*, 2007). In the present study, the comparison of the diurnal cycles of O₃ and CO, indicated that the depletion of O₃ and the enhancement of CO during the non-photochemistry hours (morning and late evening) came from the O₃ titration when mixed with urban emissions and simultaneously the anthropogenic release of CO.

Fig. 4. Diurnal variations of CO, SO₂ and CH₄ concentrations during the period of study.

The hourly concentrations of SO₂ generally increased from 06.00 to 11.00 h, and decreased in the mid-day time. After that time, the concentration increased again to give the main peak in the evening (5 pm). The concentrations of SO₂ in the early morning, late evening and night exceed their daylight concentrations due to the presence of temperature inversions during these periods and the absence of photochemical reactions. The relatively lower daytime concentrations of SO₂ would be attributed to the difference in emissions, relatively higher oxidation of SO₂ to sulfate, under the effect of solar radiation, (Harrison and Perry, 1986). Khoder's (2002) results show that the oxidation processes and conversion of SO₂ to sulfate depend on the photochemical oxidation and that the sulfur conversion ratio increases with increasing ozone concentration. In the present study, there was no diurnal variation in hourly CH₄ concentrations during the periods of study. The hourly concentrations of CH₄ ranged from 3.65 ppm at 14.00 h to 3.91 ppm at 22.00 h.

The results of daily (24-h) minimum, maximum and average concentrations of NO₂, O₃, SO₂ and CO were compared with the available air quality standards in different countries (Table 1). The

concentrations of these pollutants did not exceed the available standards. Although, the studied area does not suffer from NO₂, O₃, SO₂ and CO pollution and no health risk, comprehensive, long-term air quality management programs are needed in order to keep air quality in a good condition.

Table 1. Air quality standards in different countries (Al-Jeelani, 2009a).

Standards (ppm)	CO	NO ₂	O ₃	SO ₂
NAAQSs (USA) 1-h average	35		0.12	
NAAQSs (USA) 24-h average				0.14
NAAQSs (Canada) Desirable 1-h average	13.1		0.051	0.172
NAAQSs (Canada) Desirable 24-h average			0.015	0.057
NAAQSs (Canada) Acceptable 1-h average	30.6	0.213	0.082	0.334
NAAQSs (Canada) Acceptable 24-h average		0.106		0.115
WHO 1-h average	26		0.05-0.10	0.13
WHO 24-h average				0.038-0.058
Alberta Guidelines 1-h average	13	0.21	0.082	0.17
Alberta Guidelines 24-h average		0.11	0.025	0.06
NAAQSs (Korea) 1-h average	25	0.15	0.1	0.15
NAAQSs (Korea) 24-h average		0.8	0.06	0.05
PME (MEPA) 1-h average	35	0.35	0.15	0.28
PME (MEPA) 24-h average				0.14

Fig. 5. Comparison of diurnal variations of NO, NO₂, NO_x, NMHCs, O₃, CH₄, CO and SO₂ concentrations during the periods of October 8, 2005 – November 2, 2005 (A), November 3, 2005 - November 12, 2005 (B), November 13, 2005 - November 26, 2005(C), December 20, 2005 – December 31, 2005 (D), January 1, 2006 - January 8, 2006 (E), January 9, 2006-January 14, 2006 (F), and January 15, 2006- January 30, 2006 (G).

Comparison of Diurnal Variations of the Measured Pollutants during Different Time Periods

The diurnal variations of NO, NO₂, NO_x, O₃, NMHCs, CH₄, CO and SO₂ during different periods are graphically presented in Fig. 5. It can be noticed that, a similar behavior trends in hourly concentrations were observed for the different periods of times, with the morning and late evening peaks. The highest hourly concentrations were found in 15th-30th January 2006, for NO, 8th October - 2nd November 2005, for NO₂, and 1st - 8th January for NO_x. In addition, the highest hourly concentrations were found in 8th October – 2nd November 2005 for NMHCs, 8th October – 2nd November 2005, for CO and 1st - 8th, 15th-30th January for SO₂. With regard to the diurnal variations of O₃ during different periods of study (Fig. 5), a similar behavior trends in O₃ hourly concentrations was observed for all different periods of times, with a uni-modal peak during the daytime. The highest hourly concentrations of O₃ were found in 9th-14th January 2006. For CH₄, did not show diurnal variation during the different periods of study (Fig. 5), with the highest concentrations in 15th-30th January 2006, followed by 8th October – 2nd November 2005 and 3rd – 12th November 2005.

The daily average concentrations were 0.052, 0.045, 0.044, 0.049, 0.078, 0.050 and 0.069 ppm for NO, 0.047, 0.041, 0.035, 0.039, 0.038, 0.032 and 0.037 ppm for NO₂, 0.098, 0.086, 0.079, 0.087, 0.115, 0.081 and 0.105 ppm for NO_x, 3.28, 3.30, 2.74, 2.79, 2.67, 2.41 and 2.10 ppm for CO and 0.012, 0.010, 0.009, 0.010, 0.014, 0.007 and 0.014 ppm for SO₂ during the periods of October 8 – November 2, 2005, November 3 – November 12, 2005, November 13 – November 26, 2005, December 20 – December 31, 2005, January 1 – January 8, 2006, January 9 – January 14, 2006 and January 15 – January 30, 2006, respectively. In addition, the daily average concentrations of O₃ were 0.006 ppm in October 8 – November 2, 2005, 0.009 ppm in November 3 – November 12, 2005, 0.009 ppm in November 13 – November 26, 2005, 0.007 ppm in December 20 – December 31, 2005, 0.013 ppm in January 9 – January 14, 2006 and 0.011 ppm in January 15 – January 30, 2006. With regard to CH₄ and NMHC, the daily average concentrations were 3.89, 3.76, 3.68, 3.69, 3.65 and 4.100 ppm for CH₄ and 1.30, 1.06, 1.17, 1.02, 0.95 and 1.06 ppm for NMHCs during the corresponding periods respectively. Generally, the highest concentration levels of the measured pollutants in the study area were observed during Ramadan (8 October-2 November 2005) and Hajj (1-8 and 15-30 January 2006) days. This may be attributed to the high emission of pollutants from heavy traffic during Ramadan and Hajj days.

Correlation within the Measured Air Pollutants During the Period of Study

Correlation coefficients within the measurement of air pollutants are shown in Table 2. Statistically significant correlation coefficients ($P < 0.05$) are highlighted in bold. Significant positive correlation coefficients were found between NO, NO₂, NO_x, NMHCs and CO. SO₂ concentrations were positively correlated with NO, NO₂, NO_x, NMHCs and CH₄. These results indicated that these pollutants originate from similar or co-located sources. Significant negative correlation coefficients were found between O₃ concentrations and NO₂, since NO₂ is an important precursor for O₃, which is constantly consumed by the photochemical reactions and produced lots of O₃. Negative correlation coefficients were found between O₃ concentration and NO, NO₂ as well as NO_x (Abdul-Wahab *et al.*, 2005; Pudasainee *et al.*, 2006 and khoder, 2009), these results confirmed the finding of the present study.

Table 2. Correlation coefficients among different pollutants during the period of study.

	NO	NO ₂	NO _x	SO ₂	O ₃	CH ₄	NMHCs	CO
NO	1.00	0.55	0.96	0.77	-0.02	0.29	0.39	0.25
NO ₂		1.00	0.76	0.62	-0.35	0.20	0.59	0.64
NO _x			1.00	0.81	-0.14	0.29	0.50	0.41
SO ₂				1.00	-0.02	0.40	0.22	0.04
O ₃					1.00	0.41	-0.58	-0.43
CH ₄						1.00	0.16	-0.11
NMHCs							1.00	0.76
CO								1.00

O₃ concentrations in the present study were negatively correlated with CO and NMHCs, indicating that the reaction with OH is the primary mechanism for the removal of these pollutants from the atmosphere. Major chemical loss of CO in the atmosphere is by reaction with OH radical during daytime, producing ozone (Badarinath *et al.*, 2007).

Conclusion

The SO₂/NO_x and SO₂/NO₂ concentration ratios suggested that the mobile emissions were the predominant source within the study area. The diurnal variations in NO, NO₂, NO_x, SO₂, CO, NMHCs concentrations had similar trend patterns and inverse of that of O₃. The relatively higher traffic density during Ramadan and Hajj seasons lead to increase in the traffic emission, and consequently the concentrations of measured pollutants were increased. The significant positive correlation coefficients between NO, NO₂, NO_x, NMHCs and CO, and also between SO₂ and NO, NO₂, NO_x, NMHCs and CH₄ indicating that these pollutants originate from similar or co-located sources. The concentrations of O₃ were negatively correlated with NO₂, CO and NMHCs. The level of 1-h and daily (24-h) average concentrations of NO₂, O₃, SO₂ and CO did not exceed the national and international available air quality standards. It is recommended a comprehensive and long-term air quality management programmes are needed to ensure that tolerable levels of pollution are not exceeded and that the population exposure is reduced to acceptable levels.

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دراسة حركة واتجاهات تركيزات الملوثات الهوائية الغازية في أجواء المدينة المنورة، المنطقة المركزية، المملكة العربية السعودية

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المستخلص. تم قياس تركيزات أول أكسيد النيتروجين، وثاني أكسيد النيتروجين، وأكاسيد النيتروجين، وثاني أكسيد الكبريت، والأوزون، وأول أكسيد الكربون، والميثان، والهيدروكربونات غير الميثان خلال الفترة من رمضان إلى موسم فريضة الحج (أكتوبر ٢٠٠٥ - يناير ٢٠٠٦) في المنطقة الحضرية بالمدينة المنورة، المملكة العربية السعودية. أوضحت الدراسة أن متوسط تركيزات غازات أول أكسيد النيتروجين، وثاني أكسيد النيتروجين، وأكاسيد النيتروجين، وثاني أكسيد الكبريت، والأوزون، وأول أكسيد الكربون، والميثان والهيدروكربونات غير الميثان كانت ٠,٠٥٥، و٠,٠٤٠، و٠,٠٩٥، و٠,٠١١، و٠,٠٠٩، و٢,٨٢، و٣,٨٣، و١,١٢ جزء في المليون أثناء فترة الدراسة على التوالي. وكانت نسب تركيز غاز ثاني أكسيد الكبريت إلى تركيز أكسيد النيتروجين، وتركيز غاز ثاني أكسيد الكبريت إلى تركيز غاز ثاني أكسيد النيتروجين هي ٠,١٢، و٠,٢٨ على التوالي، وبديل ذلك على أن الانبعاثات الناتجة من المصادر المتحركة (انبعاثات السيارات) هي السائدة في منطقة الدراسة. وجد أن الاختلاف في تركيزات غازات أول أكسيد النيتروجين، وثاني أكسيد النيتروجين، وأكاسيد النيتروجين، وثاني أكسيد الكبريت، وأول أكسيد

الكربون، والهيدروكربونات غير الميثان خلال ساعات اليوم أثناء فترة الدراسة كانت متشابهة، حيث لوحظ أن أعلى تركيزات لهذه الغازات قد سجلت خلال فترتين من اليوم، إحداها في وقت الصباح، والأخرى في وقت متأخر من المساء، نتيجة لارتباطهما بكثافة حركة المرور والعمليات الكيميائية في الجو. كذلك وجد أن أعلى تركيز لغاز الأوزون قد وجد في فترة واحدة من اليوم (أثناء الظهيرة)، وذلك نتيجة لتكونه نتيجة للتفاعلات الضوئية كيميائية أثناء هذه الفترة. لوحظ أن أعلى تركيز للملوثات المدروسة في منطقة الدراسة كانت أثناء شهر رمضان وموسم فريضة الحج، وذلك نتيجة لزيادة انبعاث هذه الملوثات من الزيادة الشديدة لوسائل النقل المستعملة أثناء شهر رمضان وموسم فريضة الحج. وجدت علاقات موجبة معنوية بين تركيزات غازات أول أكسيد النيتروجين، وثاني أكسيد النيتروجين، وأكاسيد النيتروجين، والهيدروكربونات غير الميثان، وأول أكسيد الكربون، وكذلك بين تركيز غاز ثاني أكسيد الكبريت، وغازات أول أكسيد النيتروجين، وثاني أكسيد النيتروجين، وأكاسيد النيتروجين، والهيدروكربونات غير الميثان، والميثان أثناء فترة الدراسة، وهذا يُشير إلى أن هذه الملوثات تنشأ من مصادر مماثلة أو مصادر مختلطة. وجدت علاقات سالبة معنوية بين تركيزات غاز الأوزون وثاني أكسيد النيتروجين، وهذا يدل على أن الزيادة في مستوى تركيز غاز الأوزون مرتبط بالهبوط في تركيز غاز ثاني أكسيد النيتروجين.

الكلمات الدالة: ملوثات الهواء الغازية، أول أكسيد النيتروجين، ثاني أكسيد النيتروجين، أكاسيد النيتروجين، ثاني أكسيد الكبريت، الأوزون، أول أكسيد الكربون، الميثان، الهيدروكربونات غير الميثان، المدينة المنورة، المملكة العربية السعودية.