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Primary Pollutants and Potential Photochemical Smog Formation in Makkah, Saudi Arabia

الملوثات الأولية واحتمالات تكون الضباب الكيموضوئي بأجواء مكة المكرمة، المملكة العربية السعودية

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Abstract: This study was conducted in Mina Valley and the central district of the holy city, Makkah, during the pilgrimage (Hajj) season of 1424 Hijri (2004). During this season, more than 2.5 million people gathered in Makkah and Mina Valley to perform the Hajj rituals. Two Mobile air pollution laboratories were used to monitor NO, NO_2, NO_x, non-methane hydrocarbons and ozone (O_3) in the atmosphere in Mina and Makkah. Instruments were calibrated periodically against standard gases. The present investigation showed clearly an ideal diurnal cycle of local ozone formation. Although the intensity of the incoming UV radiation was the lowest compared with other months of the year, recorded ozone levels approached the maximum allowable levels of 150 $\mu g/m^3$ in Mina, and exceeded 160 $\mu g/m^3$ in Makkah during the pilgrimage period. The problem was intensified by the high recorded levels of NOx, sometimes reaching more than 800 $\mu g/m^3$, 1h average, coupled with 1h average concentration of about 3 ppm non-methane hydrocarbons. Furthermore, the average maximum hourly ozone concentrations increased gradually from less than 60 $\mu g/m^3$ during February to reach more than 200 $\mu g/m^3$ (as an indication of smog formation) during some days of May. This coincides with the increase in the intensity of the incoming UV radiation reaching its maximum level in May. Consequently, it can be concluded that Makkah may face severe air pollution episodes when the pilgrimage season shifts to the summer months in the next few years. This may pose acute health problems for elderly people and those with respiratory health problems. Good air quality and transportation management as well as the use of alternative clean fuel are highly recommended.

Keywords: *Primary pollutants, O_3, NO_x, Photochemical smog, UV, Mina valley.*

المستخلص: أجريت هذه الدراسة خلال موسم حج 1424هـ (2004) بمكة المكرمة ووادي منى، حيث يتجمع فيهما أكثر من 2.5 مليون نسمة وينتج عن ذلك ضغوطات على البيئة الهوائية نظرا للانبعاثات الكثيفة لملوثات الهواء من عوادم السيارات ووسائل حرق الوقود الأخرى. لقد تم متابعة التغير في قياسات تركيزات الملوثات الأولية وخاصة أكاسيد النيتروجين والهيدروكربونات والتفاعلات الجوية المؤدية لتكوين غاز الأوزون تحت تأثير الأشعة فوق البنفسجية. وقد تمت هذه القياسات باستخدام معملين متنقلين للرصد المستمر لملوثات الهواء تمت معايرتهما بصفة دورية كل 24 ساعة باستخدام الغازات العيارية للتأكد من سلامة ومصداقية القراءات. أوضحت الدراسة أن غاز الأوزون يتكون محليا بوادي منى والمنطقة المركزية، وبدورة نموذجية للتفاعلات الجوية المؤدية لتكون الملوثات المؤكسدة وبالتالي الضباب الكيموضوئي. وبالرغم من توافق هذين الموسمين مع فصل الشتاء حيث كانت شدة الأشعة فوق البنفسجية في أدنى مستوياتها فقد رصدت تركيزات لغاز الأوزون تقترب من الحدود القصوى المسموح بها وهي 150 ميكروجرام/م^3 (متوسط ساعة) بوادي منى وتجاوزت 160 ميكروجرام/م3 بالمنطقة المركزية بمكة المكرمة أثناء موسم

الحج. وقد ساهم في هذه المشكلة ارتفاع تركيز أكاسيد النيتروجين لأكثر من 800 ميكروجرام/م^3 كمتوسط لمدة ساعة ووصول تركيزات والهيدروكربونات غير الميثان إلى ما يقارب 3 جزء في المليون كمتوسط لمدة ساعة، هذا على الرغم من أن أقصى تركيز للأوزون بمكة المكرمة لمدة ساعة خلال فبراير 2005 خارج موسم الحج، لم يتجاوز 60 ميكروجرام/م3. وتجدر الإشارة إلى أن تركيزات غاز الأوزون قد تزايدت تدريجيا لتتجاوز 200 ميكروجرام/م^3 كمتوسط لمدة ساعة (مؤشر تكون الضباب الكيموضوئي) خلال بعض أيام شهر مايو حيث وصلت شدة الأشعة فوق البنفسجية إلى أقصى قيمة سجلت خلال العام، ولذلك فإن احتمالات تكون الضباب الكيموضوئي في أجواء مكة المكرمة بعد عدة أعوام عند حلول موسم الحج خلال شهر مايو وشهور الصيف قد تصل إلى حدود الحالات الحرجة لتلوث الهواء، بسبب توافق الزيادة الحادة للملوثات الأولية المنبعثة من وسائل النقل مع تواجد أقصى طاقة ممكنة لاستمرارية التفاعل بهواء مكة المكرمة و المستمدة من الأشعة فوق البنفسجية. وانطلاقا من هذه النتائج يوصى بضرورة الإسراع في اتخاذ الخطوات اللازمة لخفض الإنبعاثات من عوادم السيارات واستخدام وسائل النقل البديلة والوقود النظيف مع وجود برنامج متكامل لإدارة نوعية الهواء وإدارة الأزمات بمكة المكرمة واستمرارية متابعة نوعية الهواء، خاصة أثناء مواسم الحج.

كلمات مدخلية : *الملوثات الأولية، غاز الأوزون، أكاسيد النيتروجين، الضباب الكيموضوئي، الأشعة فوق البنفسجية، وادي منى.*

Introduction

Makkah, Saudi Arabia, is the holy city for Muslims. Mina Valley lies just outside Makkah, and is situated 7 km to the east of the city centre and the Holy Mosque (Al-Masjid Al-Haram). During the Muslims pilgrimage period every year, Muslims start to gather in Makkah on the beginning of Zul-Hijah (the last month of Muslims lunar calendar year). All pilgrims (more than 2.5 millions) move on the 8th of Zul-Hijah to Mina Valley. They may stay overnight at the valley for one night before they proceed to Arafat (about 12 km to the south) on the morning of the 9 Zul-Hijah (Al-Wakffah Day). On the early morning of 10th of Zul-Hijah, all pilgrims return to Mina, where they spend 3-4 days and sleep in tents, which has an area of approximately 4 km^2, with each pilgrim has an area of less than 2 m^2.

Sources of air pollution are mainly fuel combustion manifested by auto-exhaust. Means of transportation are mainly buses, minibuses, small vehicles and lorries. The central area of Makkah is characterized by a very highly dense population, high buildings, narrow streets, and congested traffic flow. Congestion and the high rate of pollutants emission in such valleys of small areas, coupled with the predominant weather condition of high temperature, lack of rainfall, prevailing one wind direction, low wind speeds and the potentiality of thermal inversions make the area an ideal situation for the accumulation of air pollutants and the formation of photochemical smog. Atmospheric photochemical reactions involving the auto-exhaust emitted primary air pollutants with reference to NOx and hydrocarbons under the effect of the incoming solar radiation to produce ozone and other health deleterious compounds have been extensively studied in the last decades in USA and European cities (Borchent, 2004; Rombout, *et al.,* 1986; USEPA, 1996; Van Aalst, 1989; Zalewsky, 1992). However, there is a lack of information on the subject in other countries under subtropical climatic conditions, such as Saudi Arabia. Furthermore, the problem is of interest in Makkah and Mina valley due to the intensified auto-exhaust emissions during Hajj period. This is due to the location of the central area of Makkah and Mina in small valleys surrounded by mountains and the predominant weather conditions favoring accumulation of air pollutants and the consequent photochemical atmospheric reaction. The problem is of concern when scavenging mechanisms fail to dilute pollutants, pilgrims may face an acute health problems.

The present work is the first outcome of a research project conducted to investigate air quality in Makkah and Mina and factors affecting pollutant concentrations during the gathering of the pilgrims. This work aims at studying the potentiality of atmospheric photochemical reactions leading to the production of high levels of ozone in the atmosphere of Mina and Makkah and the consequent formation of photochemical smog, and to investigate the required plans for air quality management under such conditions. The area of research is unique in that it represents a limited area under subtropical conditions with typical topographical features for pollutant accumulation, and therefore, can have a wide interest for air pollution studies.

METHODOLOGY

Air pollution monitoring program was conducted in Makkah and Mina Valley to monitor NO, NO_2, NO_x, non-methane hydrocarbons (NMHCs) and ozone, as well as weather elements during the pilgrimage period of 2004 G (1424H). Air pollutants were monitored at 2 locations in Mina Valley during Zul-Hijah 1424 (January 24 - February 24, 2004). Site 1 was located at the middle of the valley, whereas site 2 was located at a distance of about 40 m from a traffic busy road. One of these stations was moved to monitor air pollutants in Makkah city centre close to the holy mosque from 10 February to May 2004. The used mobile laboratory is equipped with air sampler and continuous air pollution monitors. Air was sampled at a level of 4 m above the ground surface. Hydrocarbon analyzers equipped with FID were used to monitor non-methane hydrocarbons, nitrogen oxides, NO and NO_2 were monitored using Chemiluminescent nitrogen oxides Analyzers and ozone was monitored using UV absorption ozone analyzers (Horiba Co. Japan). Stations were also equipped with data loggers and computers programmed for data processing. Nitrogen oxides and non-methane hydrocarbons were periodically calibrated every 24 h using gas cylinders of nitrogen dioxide and propane, respectively. Ozone analyzer was also calibrated every 24 h through an internal calibrator producing a standard known concentration of ozone.

RESULTS AND DISCUSSION

Nitrogen Oxides

Tables 1 and 2 show the concentrations of NO, NO_2, NO_x and non-methane hydrocarbons monitored in Mina Valley. These data show that NOx in the air of monitoring site 2 close to a busy road and a traffic bridge, peaked to high levels reaching 926 $\mu g/m^3$ and 830 $\mu g/m^3$, 1h average, during peak time of pilgrims massive movements on the 9th and 10th of Zul-Hijah, respectively (January 31st . and February 1st, 2004). Lower values of 142 and 114 $\mu g/m^3$ hourly peak concentration were recorded in the middle of the Valley at monitoring site 1 during rush hours of the same days. It should be noted that the levels of NO_2 and NO_2/NO ratios were much higher at site 1 compared to site 2, close to the traffic road, due to the consequent oxidation of NO to NO_2. Maximum hourly concentrations of NO_2 during days before the arrival of pilgrims as well as during their residence in the Valley were highly dependent on traffic activities, time of the day and meteorological conditions with reference to wind speeds and directions (Tables 1 and 2 and Fig. 1).

Typical daily pattern of NO_2 variations in Mina atmosphere show concentrations of less than 60 $\mu g/m^3$ after midnight, peaking to high levels reaching sometimes more than 200 $\mu g/m^3$ at site 2. These concentrations may be compared with NO_2 of 32-59 $\mu g/m^3$ as 1h average values recorded in urban site and street kerbside stations in Stockholm (Eerens, *et al.* 2005) and 77 $\mu g/m^3$ NO_2 the highest level recorded in Mexico city (WHO, 2005). This diurnal variation was highly dependent on time of auto-exhaust peak emissions and oxidation of NO to NO_2. The recorded maximum hourly concentration of NO_2, although less than the WHO (2000) air quality maximum allowable level of 200 $\mu g/m^3$, can pose health problems for pilgrims. This is confirmed by observing a significant increase in complaints of shortness of breath, persistent wheeze and chronic cough among children exposed to NO_2 concentrations between 38 and 147 $\mu g/m^3$ (WHO, 2000). It should be noted that asthmatics and patients with obstructive pulmonary diseases are more susceptible to acute changes in lung function, airway responsiveness and respiratory symptoms. The problem with the presence of NO_2 in air in Mina and Makkah is not only because of the adverse health effects, but also because it plays a major role in the formation of ozone, since the photolysis of NO_2 is one of the key initiator of atmospheric reactions leading to the production of ozone (WHO, 2000; Nasralla, 2001).

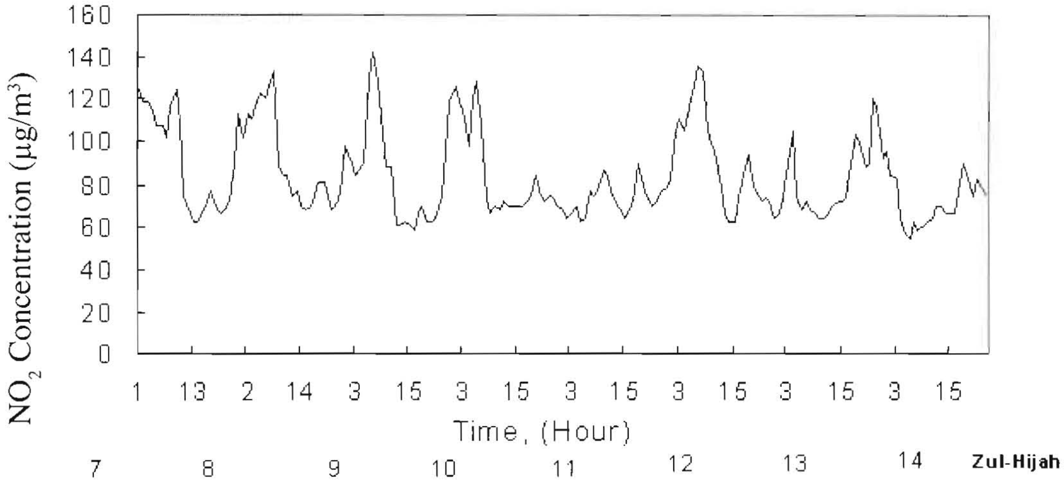

Fig. 1. Diurnal variation of NO_2, 29th Jan -5th Feb 2004 (7th - 14th Zul-Hijah) at Mina valley, site 1

Table 1. Summary of recorded concentrations at site 1, Mina valley, during 1424 H (2004).

Date (H)	Date (G)	NO µg/m³		NO_2 µg/m³		NOx µg/m³		Wind speed ms^{-1}	
		Mean	Max 1h	Mean	Max 1h	Mean	Max 1h	Mean	Max 1h
3/12/24	25/01	81.0	243.5	124.1	208.7	215.8	539.6	1.7	4.5
4/12	26/01	38.3	98.4	81.5	105.3	120.6	235.0	4.2	9.8
5/12	27/01	18.7	33.2	63.9	84.6	72.4	109.0	4.9	11.6
6/12	28/01	36.6	97.2	96.4	148.5	133.2	246.3	1.9	5.9
7/12	29/01	58.0	179.6	91.2	124.1	160.6	376.0	1.6	4.6
8/12	30/01	77.9	250.9	91.1	133.5	191.0	485.0	2.5	8.5
9/12	31/01	51.2	247.2	83.7	142.9	142.7	503.8	3.9	8.2
10/12	01/02	31.1	71.3	86.9	127.8	114.7	212.4	4.2	9.3
11/12	02/02	16.6	23.37	72.8	90.24	78.6	103.4	3.8	6.9
12/12	03/02	23.9	78.72	91.2	135.4	108.0	235.0	3.3	7.3
13/12	04/02	26.7	108.2	80.6	120.3	101.7	267.0	3.8	7.3
14/12	05/02	19.3	29.5	72.5	95.9	81.7	116.6	3.2	6.6
15/12	06/02	16.4	24.6	59.4	65.8	64.3	80.8	6.0	10.7
16/12	07/02	15.1	18.5	56.7	65.8	59.6	69.6	2.1	8.3
17/12	08/02	19.0	84.9	65.9	116.6	74.9	218.1	2.2	5.4
18/12	09/02	57.8	210.3	94.5	142.9	163.5	421.1	0.9	3.7
19/12	10/02	41.2	221.4	86.2	118.4	129.3	430.5	1.0	3.2
20/12	11/02	24.3	94.7	77.5	97.8	94.9	218.1	2.1	5.3
21/12	12/02	27.0	62.7	78.3	105.3	99.8	174.8	1.3	4.5
22/12	13/02	14.4	16.0	68.5	92.1	70.4	95.9	1.9	4.7
23/12	14/02	15.2	18.5	67.7	82.7	70.9	90.2	4.5	11.0
24/12	15/02	14.7	16.0	56.3	67.7	58.5	69.6	6.9	12.2
25/12	16/02	23.7	103.3	78.5	112.8	95.0	225.6	1.7	3.8
26/12	17/02	21.6	62.7	71.6	110.9	84.6	144.8	2.3	6.0
27/12	18/02	15.1	19.7	65.3	79.0	68.4	88.4	3.3	6.8
28/12	19/02	17.2	49.2	69.0	95.9	75.2	152.3	3.2	9.9
29/12/24	20/02	14.6	16.0	63.5	88.4	65.6	92.1	3.5	7.7
01/01/25	21/02	22.0	86.1	72.3	110.9	86.0	216.2	2.4	5.7
02/02/25	22/02	16.8	24.6	74.7	94.0	80.7	110.9	1.9	3.5

Table 2. Summary of recorded concentrations at site 2, Mina valley, during 1424 H (2004).

Date (H)	Date (G)	NO μg/m³		NO_2 μg/m³		NOx μg/m³		NMHC ppm	
		Mean	Max 1h	Mean	Max 1h	Mean	Max 1h	Mean	Max 1h
3/12	25/01	62.6	177.3	63.1	170.2	156.2	320.5	0.80	1.33
4/12	26/01	42.6	159.7	42.9	64.9	105.6	286.4	0.68	1.40
5/12	27/01	19.2	98.7	50.1	266.3	77.32	277.7	0.43	1.92
6/12	28/01	62.7	185.4	69.4	182.3	163.1	357	0.32	2.28
7/12	29/01	91.4	358.4	48.8	102.3	186.5	586.4	0.23	1.04
8/12	30/01	107.1	385.9	32.6	72.6	188.7	550.1	0.51	1.45
9/12	31/01	138.3	615.7	33.8	71.9	240.4	926	0.62	2.81
10/12	01/02	112.1	578.8	32.8	58.2	197.2	830.2	0.58	1.93
11/12	02/02	14.4	26.9	48.7	75.8	68.85	111.6	0.25	0.53
12/12	03/02	33.9	102.7	59.2	94.1	109.2	237.8	0.43	1.17
13/12	04/02	42.1	233.5	41.8	65.0	103.9	365.3	0.39	1.92
14/12	05/02	14.5	45.7	24.8	58.2	44.71	125.5	0.06	0.27

Ozone

Ozone is a very reactive and toxic gas and the most irritant of the common air pollutants. Ozone concentrations in the atmosphere in Mina Valley during the 4th day of Zul-Hijah, before the occupancy of the area, as well as days of relatively active winds in Makkah (> 6 ms^{-1}) during February 2004 ranged between 45 and 60 $\mu g/m^3$. It should be noted that the incoming ultraviolet radiation over the area during February was in the range between 35-40 W/m^2 (Seroji, 2007). This was also the case in Mina before the arrivals of pilgrims to the valley (Fig. 2 and 3).

Therefore, the 60 $\mu g/m^3$ ozone can be considered the maximum natural background concentration reaching the air of the city during winter. This level is very similar to that found in Cairo, Delhi and UK Cities (Nasralla and Shakour, 1985; WHO, 1992). Consequently, the recorded high levels of ozone reaching a maximum value of 140 $\mu g/m^3$ (Fig. 4 - 6) in Mina Valley during the pilgrimage period confirms the local production of ozone through the photolysis of nitrogen dioxide, as indicated by the reaction equations 1-3 (Fowler, *et al.* 1997; Seinfeld and Pandis, 1998; Carslaw, *et al.* 1999).

$$NO_2 \longleftrightarrow NO + O \quad (1)$$
$$O + O_2 \longleftrightarrow O_3 \quad (2)$$
$$O_3 + NO \longleftrightarrow NO_2 + O_2 \quad (3)$$

It should be noted that the presence of hydroxyl radicals and hydrocarbons in polluted air shift the reaction equilibrium to produce much higher O_3 concentrations (Carslaw, *et al.* 1999; Carslaw, *et al.* 2002; Seroji, 2007), as shown in equation 4-6.

$$RH + O_3 \longleftrightarrow RO_2 + OH \quad (4)$$
$$H_2O + O(^1D) \longleftrightarrow 2\,OH \quad (5)$$
$$OH + NO \longleftrightarrow NO_2 + H \quad (6)$$

Non-methane hydrocarbons (NMHCs) in Mina Valley reached a value of 2.8 $\mu l/L$ (ppm) during the peak time of the movement of pilgrims to Arafat on the morning of 9th Zul-Hijah. The 3 hour maximum value of NMHCs during peak times in Mina and Makkah air reached about 10 times the limit of 0.23 $\mu l/L$ (ppm), set for 3 hours in USA to control the production of ozone in polluted air. Moreover, it has been reported that conditions of intermediate values of 4:1 to 10:1 volatile organic compounds to NOx are favorable for the formation of appreciable concentrations of ozone in polluted atmospheres (WHO, 2000). These conditions were found to prevail in Mina and Makkah during the pilgrimage period (Table 2). Thorough investigation of recorded data and diurnal cycles of NO, NO_2 and ozone clearly indicates the local production of ozone in Mina and Makkah atmospheres (Fig. 4-7).

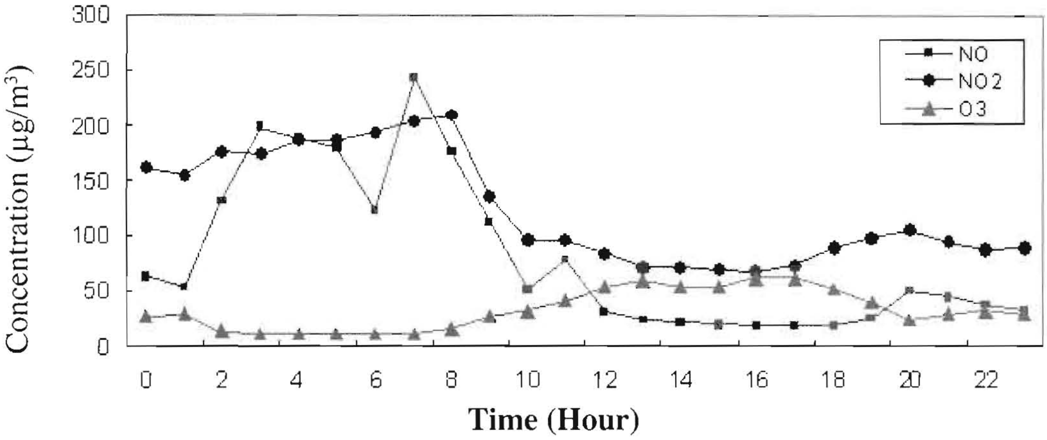

Fig. 2. Diurnal variation of NO, NO_2 and NO_3 in Mina valley 25th Jan 2004 (3rd Zul-hijah).

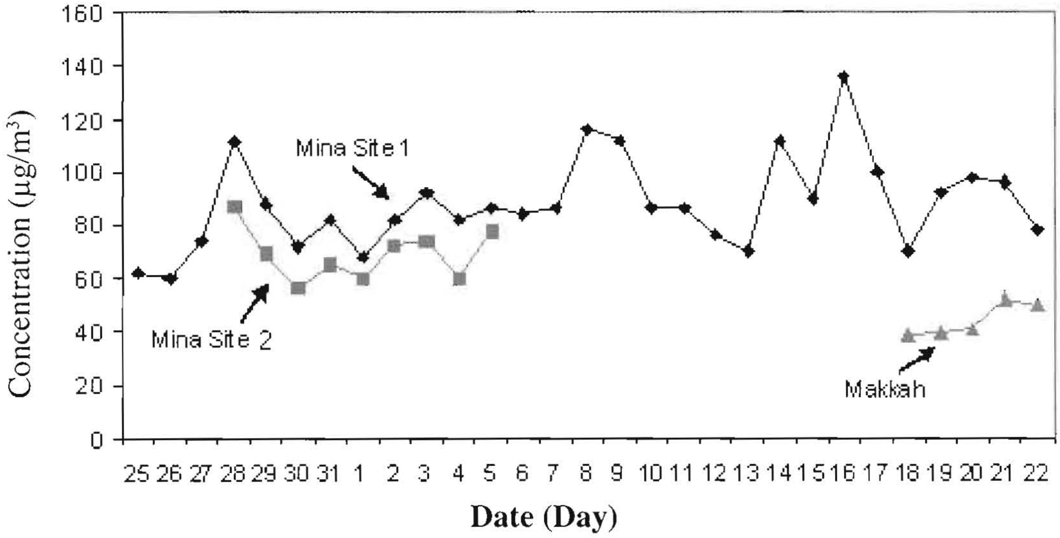

Fig. 3. Maximum hourly Ozone Concentration 25th Jan - 22th Feb 2004 (3rd Zul-hijah 1424 H - 2nd Muharram 1425 H).

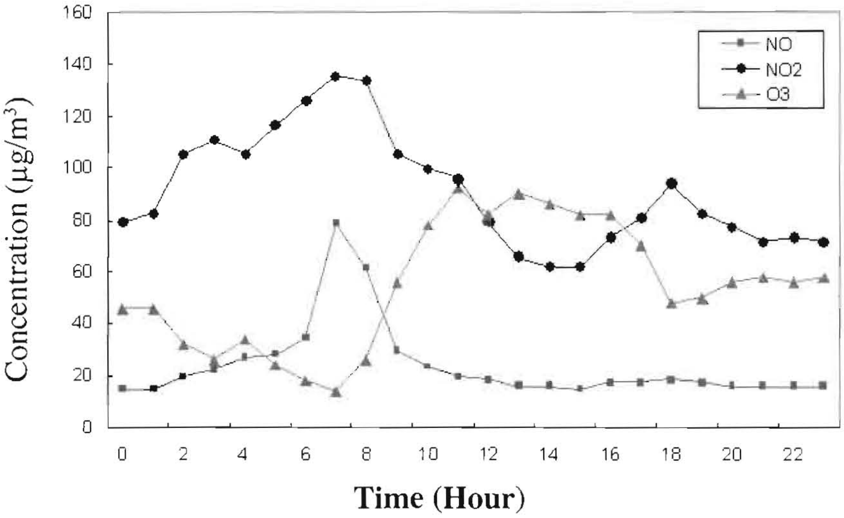

Fig. 4. Diurnal variation of NO, NO_2 and NO_3, Mina valley 3rd Feb 2004 (12th Zul-hijah).

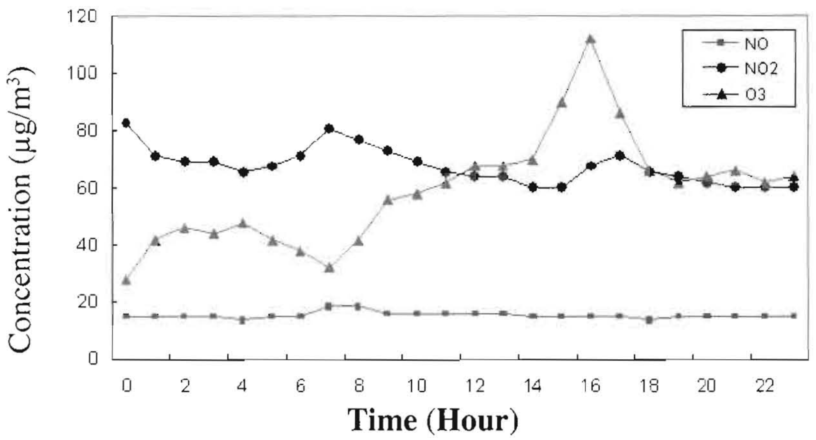

Fig. 5. Diurnal variation of NO, NO_2 and NO_3 in Mina valley 14th Feb 2004 (23rd Zul-hijah).

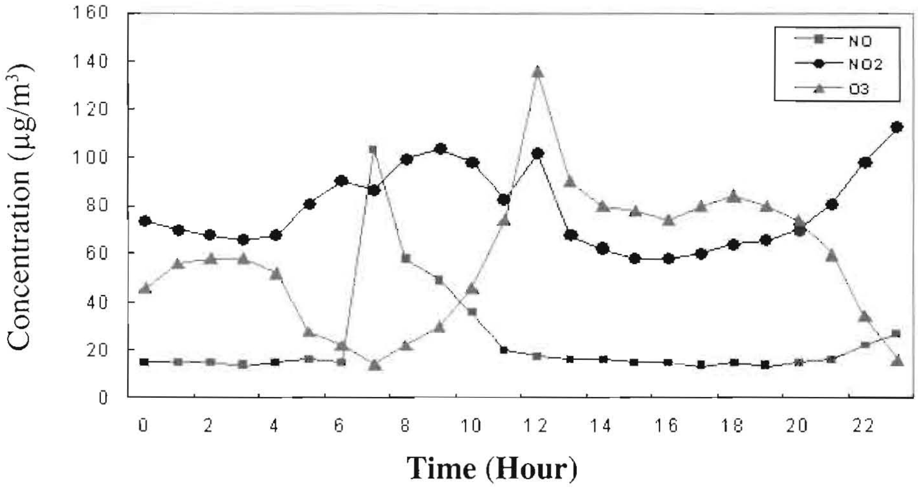

Fig. 6. Diurnal variation of NO, NO_2 and NO_3 Mina valley 16th Feb 2004 (25th Zul-hijah).

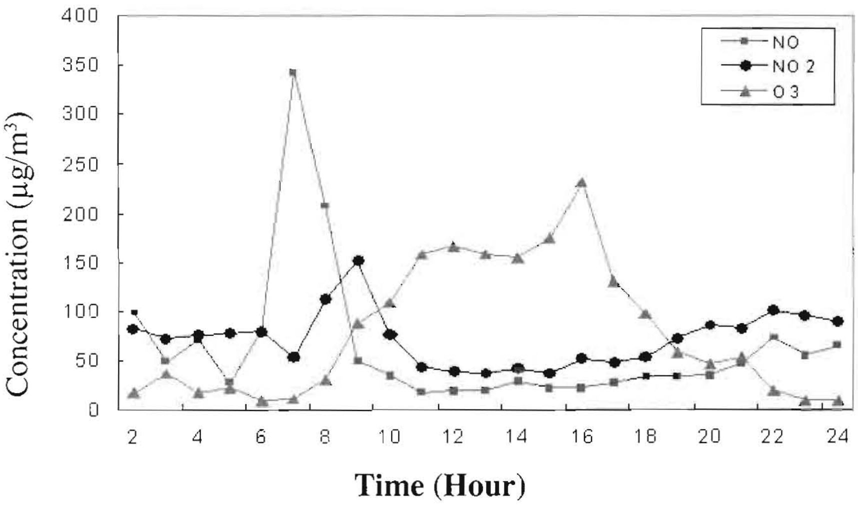

Fig. 7. Diurnal variation of NO, NO_2 and NO_3 (2nd May 2004) Makkah.

One interesting finding of this work is the increase of ozone concentrations in Mina valley after the evacuation of the valley on 13th Zul-Hijah (4th Feb, 2004). This proves the occurrence of atmospheric reactions producing ozone during the shift of pollutants with the predominant W and NW winds of low speeds from other districts of Makkah to Mina. These days were characterized by high emissions of air pollutants in the city center around the Holy Mosque when pilgrims moved to this area of Makkah. Although the maximum hourly values of ozone in Mina Valley did not exceed the guideline of 150-200 $\mu g/m^3$ hourly value set by WHO, recoded ozone concentrations can pose deleterious health impacts on pilgrims.

According to the data compiled by WHO Regional Office for Europe (2000), ozone levels found during pilgrimage period of 1424 H (2004G) can cause an increase in hospital admissions for respiratory conditions by more than 20%. These levels may also cause exacerbation of symptoms among individuals with obstructive airway disease or asthma. Associations between an increase in ozone concentration of 20 $\mu g/m^3$ and the development of asthma in exposed men has been reported (USEPA, 1994; Greer, 1993). Moreover, recorded ozone levels can contribute to the formation of significant amounts of other oxidants, as well as sulphuric acid, nitrates, sulphates. This was confirmed by recording high levels of these compounds during pilgrimage period in Makkah and Mina air (Nasralla, 2005).

The pilgrimage periods come annually during Zul-Hijah the last month of the Muslim calendar year, which is 350-355 days (12 lunar months). Consequently, the month of pilgrimage is rounded through the different seasons of the 365-day solar year. It should be noted that ozone concentrations increases significantly in the summer months due to the increase of incoming solar radiation with reference to ultraviolet reaching a level of more than 55 W/m^2 during summer months as compared to only 35-40 W/m^2 during winter (Seroji, 2007). This helped with other favorable weather conditions to produce and accumulate high ozone levels reaching more than the air quality guideline of 200 $\mu g/m^3$ in Makkah central district during May 2004, only 2 months after the pilgrimage period of year 2004 (Fig. 7).

CONCLUSION AND RECOMMENDATIONS

In conclusion, pilgrims may suffer adverse health effects during the few coming years due to the potential increase in ozone formation in Makkah and Mina and the transport of pollutants by the predominant westerly and north-westerly winds of low speeds as the month of pilgrimage slowly moves towards the summer season. Alternative means of transportation, management of the massive movements of pilgrims during this period and control of hydrocarbon emissions from the poorly maintained diesel engines (buses) and other measures of air quality management are urgently needed to protect pilgrims from potential air pollution episodes.

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