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The Influence of Meteorological Parameters on the Patterns Left by n-CH_4 and CO Gases on Concentration Roses Executed as an Environmental Source Determining Technique

S. Al-Salem

Petroleum Research and Studies Center, Kuwait Institute for Scientific Research
P.O. Box 24885, Safat 13109, State of Kuwait

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Abstract: Levels of normal methane (n-CH_4) and carbon monoxide (CO) have been increasing in the atmosphere for the past three decades. In this communication, two urban areas in the state of Kuwait (Fahaheel and Al-Riqa) were monitored for a two-year period (2004-2005). Concentration rose plotting was used to determine the major air pollution sources around both areas. A filtration procedure on the data secured was carried out. It was noticed that certain meteorological parameters, mainly wind speed had a significant influence on the shape and pattern of the concentration rose altering its shape and gas dispensing behaviour. A Chemical Mass Balance (CMB) model around the two receptor points of the areas under investigation was established to determine the major air pollution sources contributing to the ambient air load of both areas. It was determined based on chemical fingerprints of the sources that Kuwait's three refineries belt contributed 70% on Fahaheel, while 70% was associated with Fahaheel highway in the case of Al-Riqa.

Key words: n-CH_4, CO, Al-Riqa, CMB, Fahaheel, Kuwait.

Introduction

Kuwait produces two million barrels of crude oil per day (BPD), of which 40% is being processed locally in three oil refineries located on the southern part of the state (Al-Salem, 2007, Al-Salem et al., 2008). Primary air pollutants, such as Benzene, CO, CO_2, and H_2S are directly associated with oil related industries. However, other sources also contribute to the total gaseous pool of pollutants. These sources include automobile traffic (light and heavy vehicle emissions), transportation gas lines, gas dispensing networks and human activities. The contribution of these sources to the background concentrations of primary pollutants namely, methane (n-CH_4) and carbon monoxide (CO) in the outdoors could be as high as 60% depending on the area under investigation (Al-Salem and Al-Haddad, 2006, Al-Salem et al., 2007). CO has a long life span in the lower atmosphere reaching to a week's time, while n-CH_4 effect as a greenhouse gas with a serious global warming potential is twenty times greater than carbon dioxide (Kumar et al., 2004). Emissions of Green House Gases (GHGs) over the last two centuries led to a considerable increase of these gases in the atmosphere. Methane concentrations has more than doubled since pre-industrial times whereby it increased from about 0.7 to 1.7 ppmv in the ambient (El-Fadel et al., 1999). Dispersion modelling has evolved in the past three decades. That constituted a sophisticated tool, which basically reflected the current state of knowledge on turbulent transport in the atmosphere. A data set of 365 × 24 hourly samples collected at 15 different locations of five different categories was the basis of Anjali and Dutta (2005) study. These samples were used to source apportionment of the area under investigation in India using Chemical Mass Balance (CMB) modelling. The feature that is unique in this study was dividing the model source fitting equation into five categories. These categories include residential, commercial, traffic, industrial areas and petrol pumps in Delhi city. It was found that the major VOCs contributing source was the diesel combustion engines surrounding the downtown area. Al-Ajmi (1994) studied the exposure limits of a number of pollutants including methane and CO in the state of Kuwait. Point source graphing was the elementary tool used. Concentrations of certain toxic chemicals were cross-referenced using USEPA databases and suggested plans were viewed in the study. Kirchgessner et al. (1997) reported that global methane emissions from industries has been poorly quantified and not well known. As a direct result, the exact contribution of these emissions has a large error margin. Emissions were classified as fugitive, vented and combustion. The stress was on gas equipments that were related to the sources.

In this study by Fahaheel and Al-Riqa urban areas were monitored for a period of 2 years (2004-2005) in order to determine main normal-methane (n-CH_4) and carbon monoxide (CO) sources and the contribution of major gaseous air pollution sources on total ambient load of the areas. Another objective was to investigate the effect of meteorological parameters on the concentration roses plotted.

Study Areas, Data and Methods

Investigated Area Description

Al-Ahmadi governorate (where the two urban areas lie) is one of six in the state of Kuwait. It hosts over 250 hundred thousand residents. The main three urban areas of the governorate are: Fahaheel, Al-Riqa and Al-Ahmadi. Fahaheel is the largest and considered major area of the governorate. Al-Riqa could be classified as a residential area as well as an urban one due to the fact that it hosts a number of housing units that covers a large sector in case of other governorates. Figure 1 shows the governorate with both study areas (Al-Riqa and Fahaheel) indicated.

Fahaheel urban area is situated to the west of the Arabian Gulf. Its South end hosts the largest in capacity oil refinery in Kuwait (Mina Al-Ahmadi (MAA) refinery). However, Al-Riqa area is less populated than Fahaheel. The residential area of Al-Riqa host Kuwaiti residents of mid to low class living standards. Both areas downtowns are associated with human related activities summarized in restaurants, sporting establishments, shopping malls, car parking commercial complexes and automobile activities and they all contribute in terms of airborne pollutants to the ambient air load of gaseous pollutants. Fahaheel highway is one of the busiest roads in the state used by residents of Al-Ahmadi governorate and work commuters. The prevalent wind directions in Fahaheel and Al-Riqa (2004 and 2005) were north, northwest (71%), southeast (22%) and other directions (7%).

Equipment and Software

The data collected in this study included primary pollutants levels in the ambient; collected from the central location main health centre of Fahaheel and Al-Riqa from 15 and 7.5 m height, respectively. The ambient air samples were drawn from a fixed probe (Group Tek.

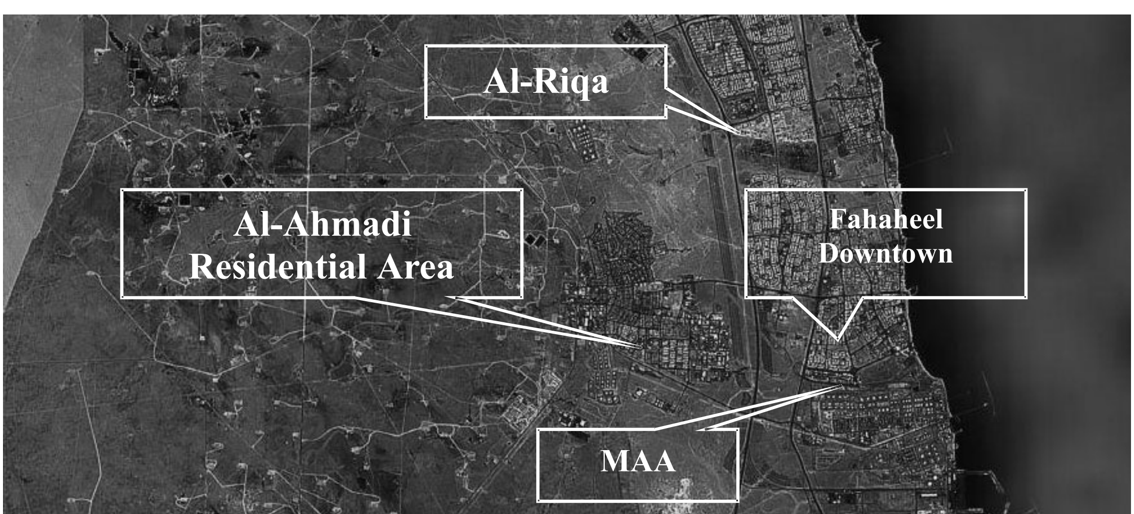

Figure 1: Satellite image showing Al-Ahmadi governorate with its main sectors: Al-Ahmadi residential area, MAA refinery, Fahaheel downtown and Al-Riqa residential area. Source: Google earth satellite archives.

Model, 3-5 m, The State of Kuwait; Fixed Photolytic converter, Group Tek. Model, The State of Kuwait) located on top of the building and were analyzed by different primary pollutants analyzers (Whatman 41, Air sample Grasbey-Anderson Ltd.) all connected with central online data acquisition system managed and controlled by *EnviDAS* software. The program transformed the data into Microsoft Office 2003, EXCEL program spreadsheets and stored for consecutive three months raw data. Data collection was conducted under the supervision of KUEPA (Fahaheel monitoring station), Air Pollution Division (APD).

The data were on two different spans, original five minute intervals and hourly averages (not used in this study). The pollutants collected included methane (CH_4) Benzene (C_6H_6), BTX, NO_x, NO_2, H_2S, NO, O_3, Total Reduced Sulfur (TRS) and total solid particulates with diameter less than or equal nominal 10 µm (PM_{10}). The data collected also included metrological parameters, which were: % relative humidity, ambient temperature (^{0}C), wind speed (ms^{-1}) and direction in degrees. The polyclinic of Fahaheel was chosen as a receptor point for being adjacent to the MAA refinery as well as the main highway. This is the central location in Fahaheel area which has all major pollution sources of airborne pollutants around in a 5 km circle. On the south side lie residential houses which could be considered close to the refineries and petrochemical industries. In the case of Al-Riqa, the main polyclinic of the area was the receptor point too. Figures 2 and 3 show a schematic drawing of the areas under investigation with receptor points indicated.

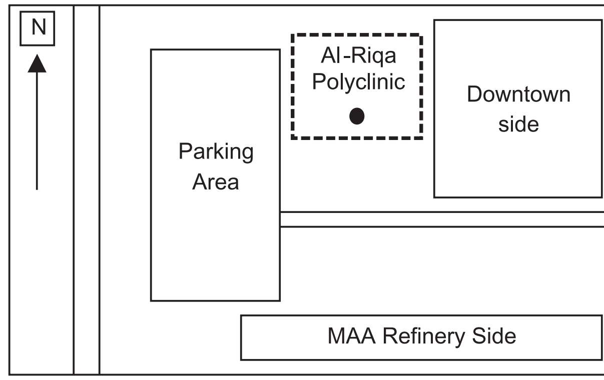

Figure 3: Schematic drawing of Al-Riqa urban area showing the location of receptor point (black circle) with respect to the main downtown area, MAA refinery and parking area adjacent to polyclinic.

As indicated by Figures 2 and 3, the main potential sources of air pollution were surrounding the receptor points chosen. This will aid in the analysis part of the work in terms of corridor distribution.

Data Filtration and Preliminary Analysis

Filtration procedure was performed by discarding zero and span check points, source hiccup values. Equation 1 shows the OX relation used to filter the raw data points based on the NO titration to ground level ozone (Klemp, 2002, Al-Salem and Al-Haddad, 2006, Al-Salem and Khan, 2006, Al-Salem et al., 2007). Any value contradicting NO, O_3, NO_x or OX was discarded.

$$OX = O_3 + NO_x - NO \ \{[=] = ppb\} \quad (1)$$

where OX = the concentration of total oxides in the

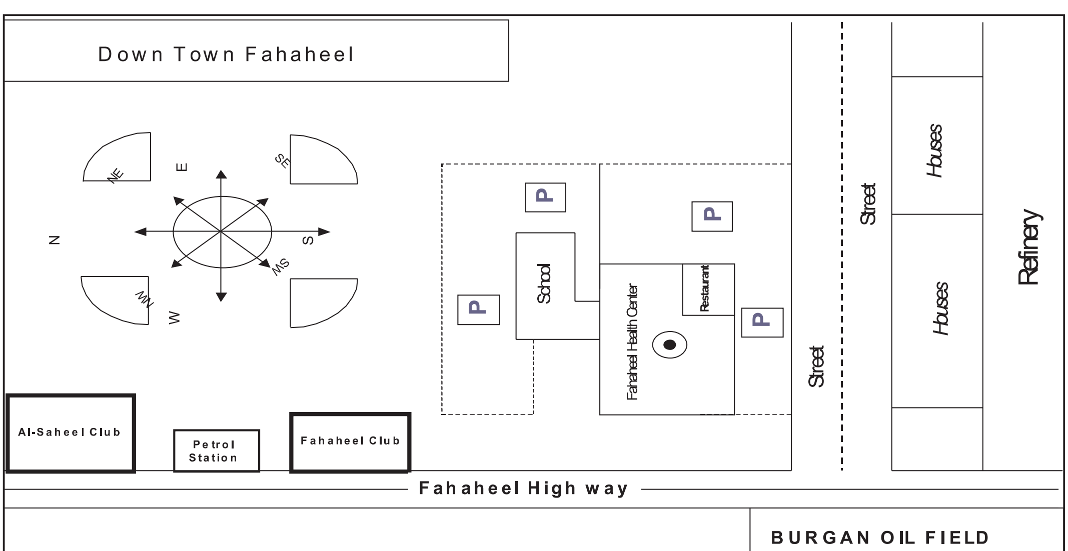

Figure 2: Schematic drawing of Fahaheel urban area showing the location of receptor point (black circle) with respect to the main downtown area, Fahaheel highway, MAA refinery and Burgan oil field.

ambient (ppb); O_3, NO_x and NO refer to ground level ozone, total nitric oxides and nitric oxide ambient levels (ppb), respectively. Figure 4 shows an example of the filtration curve executed for a day's duration.

OX levels clearly had a similar (but lagged) pattern compared with O_3 (Figure 4). Titration of NO and NOx to ground level ozone was clearly, taking its course at peak times, more specifically at cross-over time which was at 9:41 am for the day shown. No data points were discarded in the previous case. Figure 5 shows a case where the filtration was performed by discarding all values above the indicated horizontal line, i.e. exceeding 200 ppb, indicating source hiccup values.

Methane concentrations points below 1.3 ppm were discarded due to malfunctioning of the instruments from particulate accumulation, photo oxidation $r \times n$'s, choking or OH^- ion presence. In order to execute the source determining step, potential sources were referred to by identifying potential air pollution sectors around the receptor points (Fahaheel and Al-Riqa polyclinics). Table 1 and 2 show the sources with respect to corridor of wind direction. These sources stated are used in the modelling step in order to determine their weight contribution on the receptor point.

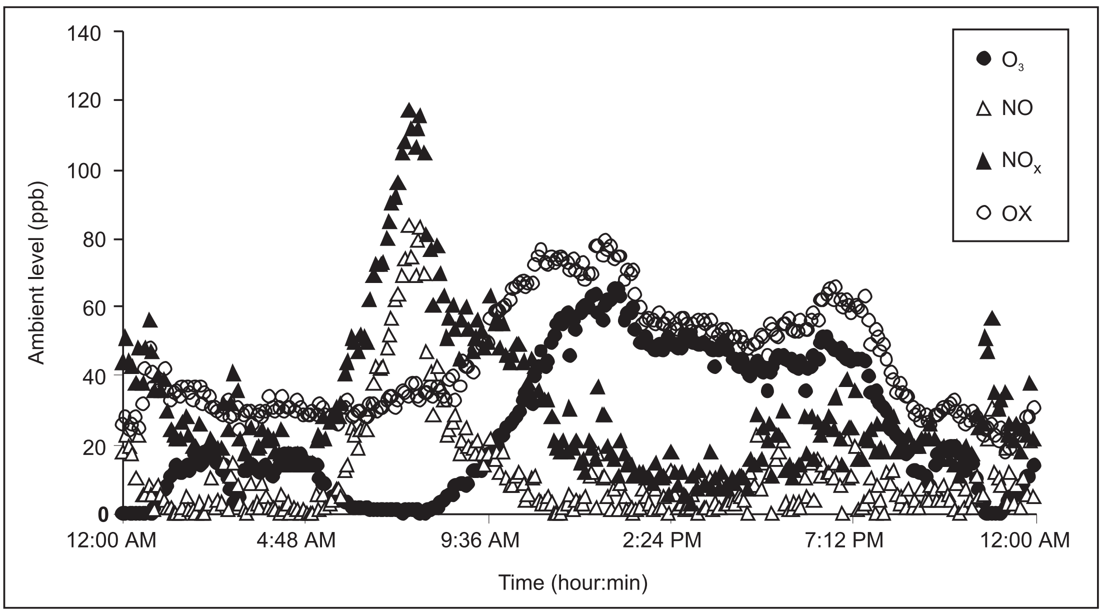

Figure 4: Filtration curve executed for a day's duration (11th July 2004) showing OX curve.

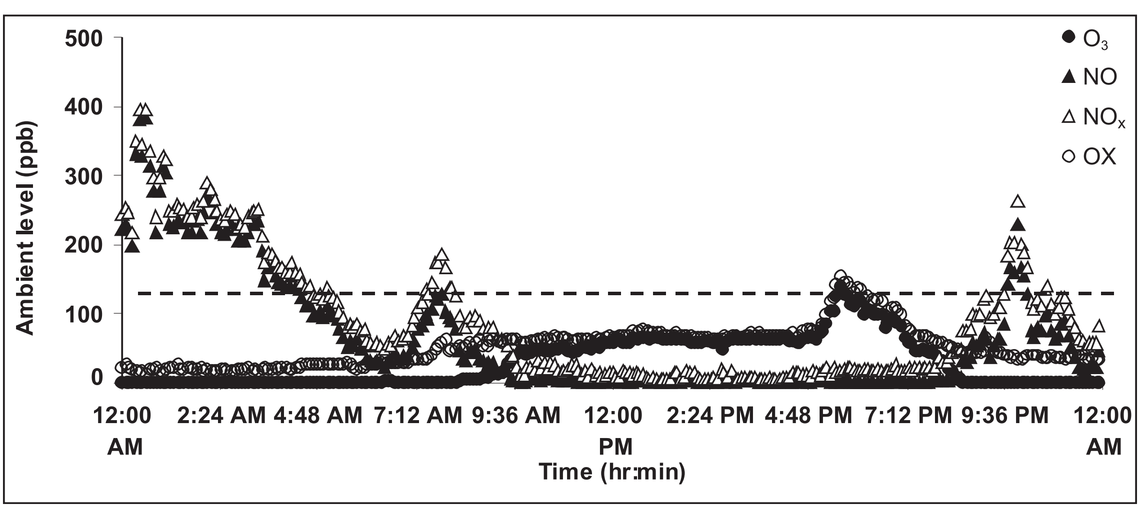

Figure 5: Filtration curve executed for a day's duration (13th July 2004) showing OX curve. Data points above line were discarded.

Table 1: Position distribution around outdoor data collection point in Fahaheel

Position in degrees	*Source*
0-135	Down town area
136-255	Refineries, petroleum and petrochemical industries
256-300	Oil production facilities (Burgan)
301-360	Traffic line sources (Highway), gas stations and sports clubs

Table 2: Position distribution around outdoor data collection point in Al-Riqa

Position in degrees	*Source*
001-130	Down town area
131-260	Refineries, petroleum and petrochemical industries
261-300	Traffic line sources (Highway), gas stations, and sports clubs

The raw data points (before filtration) were used to plot unfiltered concentration roses of methane and CO around both study areas. Concentration roses were executed for a month's average. All concentration roses in this study were done in a *blowing front* manner. It is common in unfiltered trends and graphs to have spike values shooting off the curvatures (Al-Salem et al., 2008). These spikes were noticed in the concentration roses of the raw data points. Filtering the data shapes shoot off values (spike like). Figure 6 shows unfiltered concentration roses for methane in Fahaheel urban area.

The main objective of constructing the unfiltered roses was to get an indication of the sources predominantly influencing the areas under investigation. In Fahaheel and Al- Riqa, methane and CO were blowing mainly from the southern and northwestern corridors corresponding to Kuwait's three refineries belt (downstream facilities) and Fahaheel highway (in case of Fahaheel).

Description of Executed *CMB* Model

The source allocation was ascertained by analyzing the data points collected and observing the wind directions of peak gaseous pollutants concentration values. The high concentrations of primary pollutants emitted from the sources initially thought of being of major concern. A Chemical Mass Balance (CMB) model was used to determine the exact weight contribution of each major source under investigation. Various major air-borne pollutants were present in the current pool of data. Fahaheel highway is a major line source adjacent to receptor points, resulting in high levels of certain pollutants (e.g. NO_x and CO, non-methane hydrocarbon), considering the heavy traffic influence. The downtown area had high concentrations of methane due to extensive use of bottled gas in fast-food, takeaway etc. and also had a great influence on all other types of pollutants. Based on the initial analysis and measured values of specified pollutants, the CMB model was setup in Microsoft office 2003 (Windows) in an Excel program. Non-methane Hydrocarbons (NMHC), methane, carbon monoxide (CO), total hydrocarbons (HCT) and ozone concentrations were also used in execution of CMB model as chemical fingerprints adapted from previous inventories (Al-Hajraf et al., 2005; Al-Bassam and Khan, 2004).

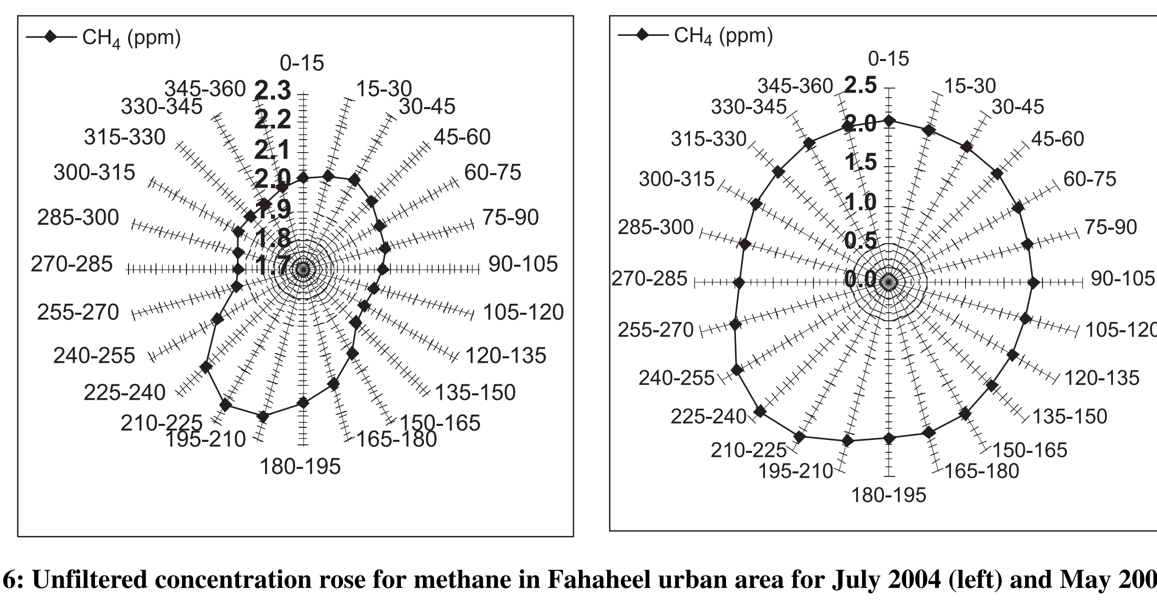

Figure 6: Unfiltered concentration rose for methane in Fahaheel urban area for July 2004 (left) and May 2004 (right).

The standard approach was applied for apportioning observed pollutant concentrations to their sources. The model implements a least square solution to a set of linear equations, expressing each source as a linear sum product of the source percent contribution with predominant wind sector.

The CMB equations were based on the assumption that the observed ambient quantity of a chemical species is the simple sum of the product of pollutants contributions affecting the airshed and fraction of the wind sector. CMB model uses the chemical and physical characteristics of gases and particulate at a given receptor point to identify the presence of and/or quantify source contributions. Equation (2) is the basic relation corresponding to the selected receptor point. This equation expresses the relation between the concentrations of the chemical species measured at the receptor point (Main health centre of Fahaheel) and the chemicals emitted from the source.

$$\Delta C_i = \Sigma F_{ij} - S_i \qquad (2)$$

where ΔC_i is the difference in concentration of a chemical compound *i* at the receptor point, F_{ij} is the fraction of concentration of the species *i* starting from the source *j*, S_i is the concentration of pollutant *i* at the receptor point.

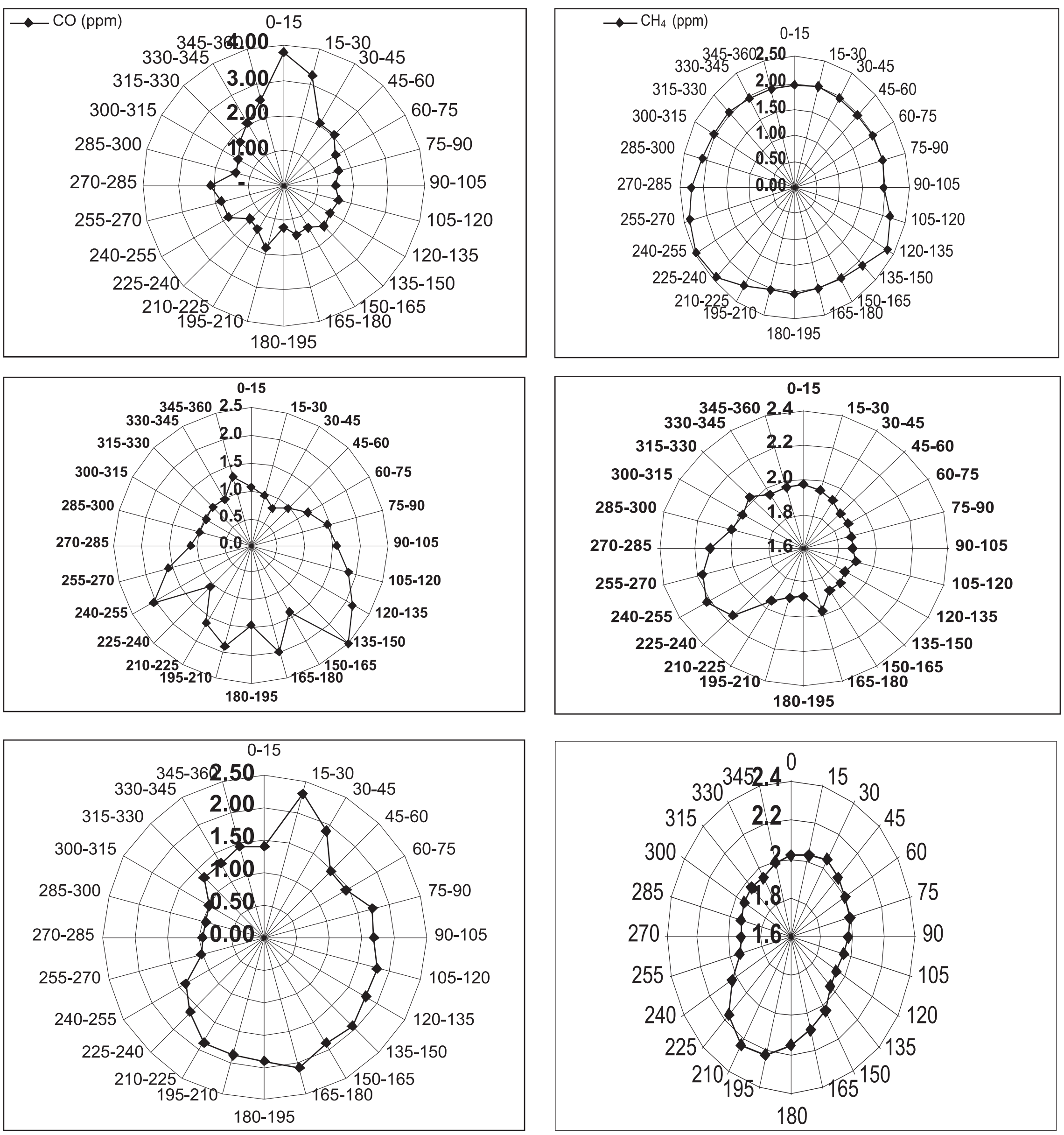

Figure 7: Sample of the concentration roses plotted for Fahaheel urban area for both CH_4 (left) and CO (right) for the period of April (top), June (mid) and July (bottom) 2004.

The total wind speed contribution must be calculated in order to get the percent wind speed contribution with respect to the desired range of wind directions i.e. the source. Equation (3) was used to calculate the wind speed contribution with respect to each source.

$$\%WS_i = (k_i/K) \times 100 \quad (3)$$

where $\%WS_i$ is the percent contribution of wind speed with respect to source i, k_i is the summation of wind speed points collected with respect to source i in ms^{-1}, K is the total summation of wind speed points in ms^{-1} excluding calm period.

In order to match the concentrations at the receptor point, predefined linear functions were solved with an objective function. The objective function which is defined as the sum of squares of difference between measured and the sum of fractional concentrations of different sources chemical fingerprints including the influence of wind sector, is minimized. The chemical fingerprints were average readings of concentrations reflecting the recorded inventories of the sources. Equation (4) is the one set to solve for the least linear square root. The linear function was introduced for the four major sources studied as well as, the receptor point, which represents the total cumulative concentration of a pollutant to be matched.

$$\text{LF} = \sum_{j=1}^{m}\sum_{i=1}^{n} C_i.WS_i.SC_i - \sum_{i=1}^{n} C_i.WS_i.SC_i \quad (4)$$

where L.F. is the linear function set to match the percent contribution of each source, C_i is the concentration of air-borne chemical i at a certain source or receptor point, $\%WS_i$ is the percent wind speed contribution at a certain wind direction range for source i, $\%SC_i$ is the percent source contribution for a source I and i represent pollutants and j sources.

Results and Discussion

Filtered data points were used to plot the concentration roses of both methane and CO. Figures 7 and 8 show sample of the concentration roses plotted for Fahaheel and Al-Riqa urban areas (2004-2005).

Fahaheel plots show clearly major contribution from refineries and highway side in the case of CO (Figure 7). It was clear in the month of April 2004; the influence of the highway side was greater (≈ 4 ppm) compared to July and August of the same year (1.25 and 1.2 ppm, respectively). In Al-Riqa urban area (Figure 8), CO mainly is blown from the refineries side reaching 1.4 ppm in the month of July 2004. In the case of methane

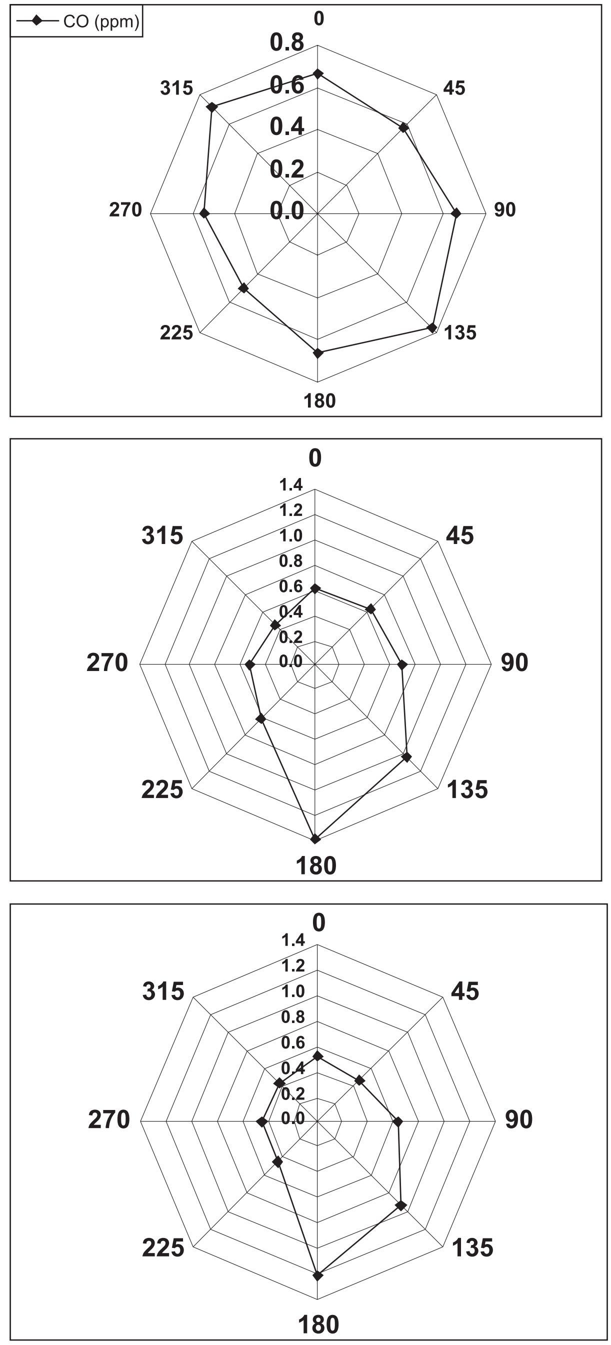

Figure 8: Sample of the concentration roses plotted for Al-Riqa urban area for CO in the period of May (top), July (mid) and August (bottom) 2004.

both in Fahaheel (Figure 7) or Al-Riqa (not shown), all corridors contribute to the ambient load.

Being a gas emitted from many human and non-human related air pollution sources; but mainly blown from the southern refineries side. Fugitive and vented emissions, as well as gas dispensing networks of southern industries area contribute to the methane load and could be easily detected from the corridor (Al-Salem and Khan 2006;

Table 3: Meteorological parameters in Al-Riqa and Fahaheel (2004)

Month	*Fahaheel* Mean ambient temp. (°C)	*R.H. %	**WS (ms^{-1})	*Al-Riqa* Mean ambient temp. (°C)	*R.H. %	**WS (ms^{-1})
January	16.23	67.90	2.10	16.10	66.00	1.99
February	16.90	55.63	2.92	17.83	54.63	2.91
March	22.45	56.33	3.30	20.50	54.11	3.10
April	26.40	34.70	3.72	27.00	47.90	3.20
May	33.80	18.10	3.41	17.40	42.33	3.20
June	36.90	22.98	3.79	28.6	39.40	3.30
July	37.11	39.00	3.33	29.20	38.40	3.34
August	39.34	38.22	3.23	38.40	35.70	3.00
September	36.10	32.34	3.89	35.50	25.53	3.61
October	27.50	44.51	2.67	30.30	46.60	2.52
November	20.80	56.80	3.64	21.40	55.50	2.71
December	15.50	66.12	3.03	15.97	74.36	2.60

*** R.H. is relative humidity of site (%), ** WS is wind speed measured in ms^{-1}.**

Al-Salem et al., 2007; Khan and Al-Salem, 2007). It could be seen clearly in Figure 7, the effect of dispensing winds on the shape of the concentration rose comparatively. In cooler durations in Kuwait, mainly in January to May, the shape of a dot appears on the plotted roses. While, in hot durations an ellipsoid shape appears on the roses. Hot periods in the state of Kuwait are usually associated with both high winds (exceeding 15 ms^{-1}) and dust storms. The reported ambient temperature, relative humidity and wind speed in both study areas are given in Table 3 for the year 2004.

High velocity winds help dispense the methane gas in the lower atmosphere decreasing its ambient levels. As can be seen in Table 3, Fahaheel being adjacent to the coastal line experiences sea breeze which affects the trend of ambient temperature compared to Al-Riqa. A phenomenon described clearly by Pasanen et al. (2004) states that dust in the ambient help adsorb methane particles in the lower parts of the atmosphere, also resulting in a decrease in its ambient levels. Figure 9 shows the methane levels measured in Al-Ahmadi governorate (mean Fahaheel and Al-Riqa level) with PM_{10} levels during the period of study. The patterns detected caused by meteorological parameters don't appear valid in the case of CO, since methane gas has more sources around the receptor points resulting in a clear changed manner in case of wind dispensation.

Figure 9 clearly shows the levels of methane decreasing in dusty seasons in the state (summer). Levels of minimum ambient methane coincide with peak PM_{10} values. Major load on the ambient was calculated to be from both Kuwait's three refineries side and Fahaheel highway. In the case of Fahaheel CMB results revealed that Kuwait's three refineries belt contributed 70% to

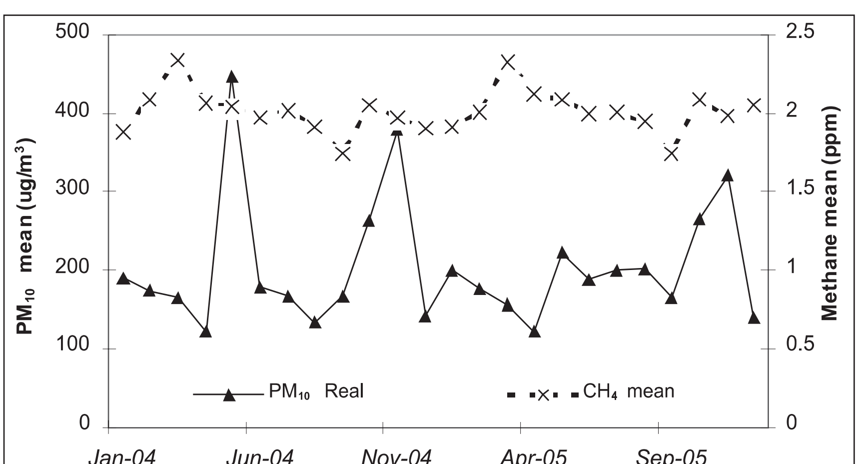

Figure 9. Mean methane and PM_{10} levels recorded in Al-Ahmadi governorate during the study period.

the ambient load, while in the case of Al-Riqa Fahaheel highway contributed 70% on its ambient load. Table 4 shows the results (mean) obtained by CMB for both urban areas.

Table 4: Percentage source contribution in Al-Riqa and Fahaheel urban areas based on CMB model results averaged over the period of the study

Source	*Fahaheel (%SC)*	*Al-Riqa (%SC)*
Petroleum downstream facilities	70	10
Petroleum upstream facilities	2	–
Fahaheel highway	18	70
Area's downtown	10	20

The location of upstream facilities referred to in the case of Fahaheel is relatively far, which helped decrease the contribution of associated activities in that area. A significant presence of both highway and refineries side was detected in both areas. In order to investigate in depth processes and emission sources (i.e. light vehicle emissions, combustion sources, stack gases from downstream area adjacent to investigated site) determining pollutants levels in the ambient air, statistical relationships between gaseous pollutants (CO, PM_{10}, NO, NO_2, O_3, and SO_2) were explored in Fahaheel urban area as a representative for the governorate. Principal component (PC) factor analysis was applied to the combined dataset in order to group and classify those pollutants displaying similar characteristics (Harrison et al., 1997; So and Wang, 2003). The eigenvector analysis was conducted on the correlation matrices of the dataset (daily mean values) so that the resulting factor loadings can be explained as correlation elements. For the determination of the number of uncorrelated factors to be retained the factor number/eigenvalue plots were examined (not shown). Factors, for which decrease in their eigenvalue became insignificant, were not retained (Wilks, 1995). Results of the cluster are shown in Table 5. Central location in the metropolitan area (ZOG) of Athens, Greece was also shown for comparative reasons (Grivas et al., 2008).

For Fahaheel urban area it was apparent that PM_{10} levels were highly related to primary pollutants emission sources as deduced by the very high factor loadings in PM_{10}, CO, NO, NO_2. It was characteristic that the first factor explains a rather high percentage of the dataset (56%). Previous studies had demonstrated that PM_{10} ambient levels were highly correlated to primary gaseous pollutants (CO, NO, NO_2) denoting the impact mainly of vehicle circulation (Chaloulakou et al., 2003). On the other hand, it can be clearly observed how O_3 was highly anti-correlated with primary pollutants in PC1 and especially with NO. This is due to the "titration effect" of NO to ground level ozone (Riga-Karandinos and Saitanis, 2005). The second factor in Fahaheel should be related to local conditions favouring ozone formation in the urban area. However, it can be seen that the relation of PM_{10} with this factor is loose. Regarding ZOG site in Athens (Greece), factors extracted explained a total of 71% of the variance (similar to Fahaheel total variance of 73%). The first factor in the site was also associated with primary emission sources; however the PM_{10} loading was very low (0.2). The second factor was characterized by high loadings in ozone and PM_{10} and should be associated with processes evolving on a regional scale.

Table 5: Results of the principal component factor analysis (correlation matrix) for the datasets including PM_{10} and other major gaseous pollutants

	Fahaheel		*ZOG*	
	PC1	*PC2*	*PC1*	*PC2*
PM_{10}	0.73	0.11	0.21	0.78
CO	0.9			
NO	0.81		0.89	
NO_2	0.72	0.46	0.85	0.22
O_3	–0.63	0.60	-0.62	0.57
SO_2	0.61	0.34	0.21	0.47
Eigenvalue	3.49	1.05	1.99	1.21
% of variance	56.0	11.0	42.5	28.0

Factors loadings lower than 0.2 were suppressed

ZOG in Athens Greece results source: Grivas et al., 2008.

Conclusion

CH_4 and CO ambient levels were monitored for a period of two years in Fahaheel and Al-Riqa urban areas. The concentration roses plotted confirmed the suspension of Kuwait's refineries belt and Fahaheel highway being the two major sources emitting both primary pollutants. Kuwait's three refineries belt had a 70% contribution on Fahaheel. However, Greater Burgan oil field contributed by only 2% due to distance. Fahaheel highway (associated with light and heavy vehicle emissions) had a 70% contribution in the case of Al-Riqa, while an 18% contribution was associated with it in the case of Fahaheel. Wind speed and PM_{10} levels were stated to be the major influencing parameters on methane ambient levels in both study areas. Dot shape concentration roses appeared in cool durations, while ellipsoid shaped ones appeared on hot periods. PM_{10} levels were highly related to primary pollutants emission sources as deduced by the very high factor loadings in PM_{10}, CO, NO, NO_2 obtained from the statistical analysis.

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Volume 4 Number 2 **July – December, 2007**

Contents