J. Mater. Environ. Sci. 6 (6) (2015) 1787-1895
ISSN : 2028-2508
CODEN: JMESCN

Atmospheric BTX measurements in the urbano-industrial site of Anza, northwest of Agadir city, Morocco

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Abstract
In Morocco, air pollution is mainly due to road traffic and industrial sources. Until today, only standard gases such as NO, NO_2 CO, SO_2 and $O_{3,}$ as well as PM_{10}, are continuously monitored in the Moroccan large cities. VOC measurements are very rare. This paper presents for the first time the results of measurement campaigns of BTX in a small town with industrial character, Anza locality, located 7 km northwest of the city of Agadir (southwestern Morocco). The BTX measurements were conducted from February 2012 to November 2012. Benzene, toluene, and the three isomers of xylene were quantified in ambient air in four places carefully selected to reflect the main activities in the city. The outdoor average concentrations of benzene, toluene and xylenes were 12.9 $\mu g.m^{-3}$, 36.9 $\mu g.m^{-3}$ and 16.2 $\mu g.m^{-3}$ respectively. No seasonal variation was observed. The fluctuation concentrations are attributed to starting up of the activities and variations flow of cars. The wind is the meteorological factor which drives the observed results. An important decrease of the BTX concentrations was observed in 2012 compared to the values obtained in 2007.

Keywords: Atmospheric pollution; Benzene; Toluene; Xylenes; Active samplers; Urban air quality; Anza; Morocco

1. INTRODUCTION

Air pollution associated with traffic emission constitutes a major problem in developing countries. The air quality problems result from increased emissions of NOx and VOCs [1]. Rapid industrialization, urbanization and motorization induce serious air quality problems, especially in large cities. Fast increase in the transportation demand is a direct result of the fast urbanization rate and leads to a rapid increase of the number of private vehicles. The capacity of the road system is greatly exceeded and slow traffic is often observed, especially at rush hours, affecting overall quality of life of urban residents.

In the ambient air, VOCs are emitted from anthropogenic and biospheric sources [2]. Ozone (O_3) and secondary organic aerosols (SOA) are formed by photochemical reactions of NOx in the presence of VOCs compounds [3-5]. The BTEX (benzene, toluene, ethylbenzene and xylenes) constitute the main Volatile Organic Compounds (VOCs) commonly related to the vehicle exhaust [6], especially in metropolitan areas [7-9]. Due to their negative impacts on health, including anemic leukaemia and a carcinogenic effect [10-12] and their key role in tropospheric chemistry [5], extensive investigations of BTEX have been carried out in the troposphere [13-14]. Benzene (C_6H_6) constitutes the base of aromatic hydrocarbon group and is classified as a Class 1 carcinogen [11]. Toluene (C_6H_8) is the most abundant in air, and xylenes (C_8H_{10}) are regarded as the most reactive compounds in the BTX family. Benzene and toluene usually have common sources, but different atmospheric lifetimes. Their concentration ratio (B/T) is often taken as an indicator of recent emissions [15].

To our best knowledge, with the exception of the studies by the Ait Taleb et al. [16-17] and by Zouir et al. [18], no VOCs measurements have been performed in Moroccan areas. The present work aims to study the pollution by BTX in the air of the Anza locality, an urbano-industrial site located northwest of Agadir city in Morocco. This study is motivated by several reasons:

- In Morocco, measurements of air pollution are up to now limited to classic gaseous pollutants such as sulfur dioxide SO_2, carbon monoxide CO, nitrogen oxides NOx, ozone O_3 and particulate matter PM_{10},
- There is a great lack of information on the VOC levels, and particularly on BTEX in urban air areas,
- Several studies have shown that many large Moroccan cities present pollution peaks in summer [19],
- The concentrations of NOx and CO, obtained in the Anza site by the Mohammed VI Foundation for the Protection of the environment are below the Moroccan limit values (200 µg.m^{-3} for NO_2, 10 mg.m^{-3} for CO, and 110 µg.m^{-3} over 8 hours for O_3). Only ozone exceeds the limit value. Aldehydes concentrations measured between 2008 and 2010 by Ait Taleb et al. [16-17] in this site were relatively higher than those usually found in outdoor air, with average concentration values in the range of 31.8 to 89.1 µg/m^3 for formaldehyde, 24.0 to 67.6 µg/m^3 for acetaldehyde and 4.9 to 9.2 µg.m^{-3} for benzaldehyde.
- Finally, the Anza site was always subject to episodes of olfactive and respiratory nuisances because of emanations from cement factory (now closed since March 2011), of the industrial units, in particular food-processing and manufacturing of plastics, and a very dense traffic.

This study, which is the first of its kind in Morocco, presents the results of benzene, toluene and xylenes (BTX) measurement campaigns realized in outdoor air of Anza, where the atmospheric pollution is clearly visible, with, in addition, odours probably coming from atmospheric decomposition of VOCs. This work aimed also to compare the 2012 values with the 2007 values [20-21] obtained before relocating several industrial units from the studied area.

2. Materials and methods

2.1. Description of study zone

The area of Anza is located at the feet of the mountains of High Atlas on the Atlantic coast of Morocco, 7 km North-West of Agadir city, with more than 45.000 inhabitants. Anza is located at about of 30.25°N of latitude and 9°38 W of longitude, which confers it a long sunshine period during all year. The climate tends to be semi-arid, hot and relatively humid, with local variations due to the altitude (about 75 meters over the sea level), mountains barrier and the proximity of Atlantic Ocean. The yearly average maximum and minimum temperatures are 32 °C (August) and 14 °C (January) respectively and relative humidity ranging between 31% and 56%. Wind is mainly in the west-northwest direction, with an average wind speed varying between 0.2 and 3.5 m.s^{-1} [22]. These meteorological parameters create regularly in the year a system of temperature inversion which traps gaseous pollutants in the atmosphere and leads to an increase of their concentration in the air.

For a long time, Anza has constituted the main industrial zone in the south of Morocco. More than 71 industrial units were installed in the north of the port of Agadir, leading to a layer of dust in ambient air, irritating odors, and the deterioration of local vegetation. Several sources of pollution can be identified such as vehicular exhausts from a heavy traffic, and emissions from industrial activities (cement manufacturing, roasting of coffee, petrol product storage, production of fish meal and fish oil, production of plastics…), where the predominant processes are organic chemistry and combustion. Also, Anza is crossed by the coastal road (RN1) connecting Agadir to Essaouira city, which carries many thousands of diesel vehicles per day, including automobiles, trucks and buses.

2.2. Localization and classification of sampling points

BTX concentrations were measured at four places around the locality, carefully chosen to reflect the main activities. Figure 1 shows the geographical localization of these sites. Table 1 describes their immediate environments. Site 1 is in the garden of the administration building (first district), where the predominant traffic is light vehicles often fueled with regular diesel. The area can also receive emissions from the nearby industrial units as well as from the production of fish meal or fish oil. Site 2 is located in the middle of three lanes of heavy traffic all in each direction and where the traffic includes light and diesel fueled heavy vehicles. It is at proximity of the cement factory (now closed). Besides emissions coming from the chimney stacks of cement reactors, the area receives the emissions from the Oil Souss unit and from the fish processing plant, where many organic compounds are used. Site 3 is a roadside sampling point in a residential area which is located behind the former cement factory. Site 4 is situated less than 1 km away from the petrol products storage facility.

Figure 1 : Map of the Anza region with localization of the sampling points

Table 1: Placement and classification of BTX sampling points

Site	Placement	Immediate Environment
S1	Garden of administrative building of the district n°1 of Anza	Heavy traffic place, roadside of RN1 emissions, food processing units.
S2	Bordering the police station, proximity of cement factory	Place with high activities, roadside of RN1, unit of cement, Souss Oil-production
S3	New residential lot in Anza	Behind the cement unit and on the RN1 east roadside
S4	On a street in a moderately urbanized environment	Immediate periphery (about 1 km) of the regional deposit of petrol products storage and food-processing industries.

Table 2 gives the meteorological conditions during 2012. Figure 2 shows the local prevailing winds at the zone. Most of the samples were collected on sunny days with a clear sky, with peak solar irradiation occurring during the early afternoon (12:00-16:00 local time). In summer, warm conditions dominated the air during the sampling period, with daytime temperature ranging from 19°C to 39°C. In winter period, the temperature range was 11 to 26°C. The wind speed varied during all the sampling days between 0.1 and 3.9 m.s^{-1}.

Table 2: Temperature and relative humidity recorded during 2012

Month in 2012	Temperature (°C)		Relative humidity (%)	
	Range	Average	Range	Average
January	7-23	14.0	5-100	55.9
February	6-21	13.5	5-88	36.0
March	9-31	18.8	5-100	43.1
April	9-28	17.0	5-78	46.7
May	11-36	23.5	5-100	48.3
June	17-39	26.1	6-97	40.6
July	19-41	29.8	5-88	38.8
August	22-41	31.6	5-100	36.6
September	17-37	24.9	5-100	30.9
October	14-33	22.6	6-98	57.7
November	12-29	19.1	5-100	50.6
December	9-26	15.6	5-100	42.6

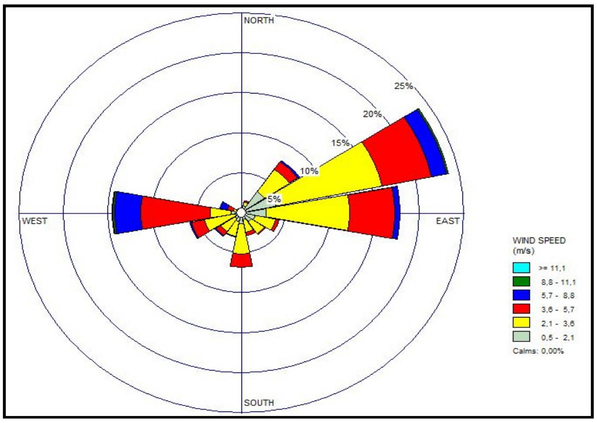

Figure 2 : Prevaling winds at the zone

2.3. BTX sampling and analysis

The measurement of BTX was carried out using active sampling, based on USA-EPA methodology [23]. The sampling system consists of a battery-operated pump (KNF NPK), a flow meter (Cole Parmer, 0.1-1 L.min^{-1}), SKC sampling tubes and Teflon tubing. BTX were sampled by drawing air through charcoal tubes (70 mm long, 4 mm id, particle size 20/40 mesh, reference 226-01 from SKC Inc.) containing two beds (a 100-mg section and a 50-mg section) separated by urethane foam. The aromatic compounds were sampled over 60 to 90 min sampling time with a flow of 0.5-0.6 L.min^{-1}. Protection of the exposed charcoal tubes is a very important part of the BTX measurement method. Therefore, as soon as the pump was turned off, the SKC sorbent tubes were disconnected from the sampling system, labelled and the two open sides were tightly closed with special caps to avoid any desorption and contamination. The samples were immediately placed into closed special plastic bags and kept in freezer (at 4°C) until analysis, within 2 to 8 days. In order to detect breakthrough, two tests were performed: analysis of the second section of the SKC tube, which indicated that less than 12% of the BTX compounds were trapped in this portion. The second test was based on a few experiments carried out with two SKC tubes connected in series. In this case, the results indicated no significant BTX levels (< 5%) were collected in the second tube. For that reason, a single sorbent tube was used most of the times to sample BTX in ambient air.

Before analysis, contents of both sections in the sorbent of SKC tubes were transferred to 4 mL vials, weighed and extracted by adding 2 or 3 mL of cold dichloromethane CH_2Cl_2 (below 5 °C to prevent losses by volatilization) as extraction solvent with agitation for 20 or 30 min [24,25]. Carbon disulfide (CS_2) was not used here because of its high toxicity, even though it is considered as the solvent of choice for charcoal tubes extraction. Analysis and quantification were performed by injection of 2 µL of the extracted solution in an Agilent 6890 gas chromatograph coupled to a FID detector. Samples were separated with a capillary column (Restek, Rtx®-5, Crossbond® 5% diphenyl/95% dimethyl polysiloxane, 30 m long, 0.32 mm internal diameter, 0.25 µm thickness film). The carrier gas was helium, delivered at a flow rate of 1.5 mL.min^{-1}. The injection temperature was 300 °C in splitless condition with 1 or 2 µL of injection volume. The oven temperature of the gas chromatograph was initially held at 60 °C for 4 min, then raised at a rate of 10 °C.min^{-1} to 180 °C and finally kept constant at this temperature for 6 min [26].

Examples of chromatograms are presented in Figure 3 ((a) calibration and (b) sample). With these experimental conditions, the retention times are RT(benzene) = 2.9 min, RT(toluene) = 3.5 min, RT(m-xylene) = 4,5 min, RT(p-xylene) = 4,58 min and RT(o-xylene) = 5.1 min respectively. The concentrations of the BTX compounds were read from external calibration lines obtained generally with 0.5 or 13.5 mg.L^{-1} of BTX prepared in refrigerated pure dichloromethane (benzene : Riedel de Haan, purity 99 % ; toluene : Prolabo, purity 99% ; m-p-o- xylenes : Sigma-Aldrich, purity 98 % ; dichloromethane : Sigma-Aldrich, purity > 99.9 %). Figure 4 presents an example of calibration curves with a regression coefficient (r^2) greater than 0.97 for all the compounds. In order to control the quality of samples and analysis, the reproducibility of the data was checked by analysis of duplicate samples, for which usually differences of less than 12% were observed. To determine the residual BTX concentrations, unexposed SKC tubes were analyzed before each analysis of samples in all campaigns series in this work. The amounts of some residues of benzene and toluene found in blanks were never exceeded 15% of any result in this work. Also the tests conducted regarding the storage time show very low increase in the blank values.

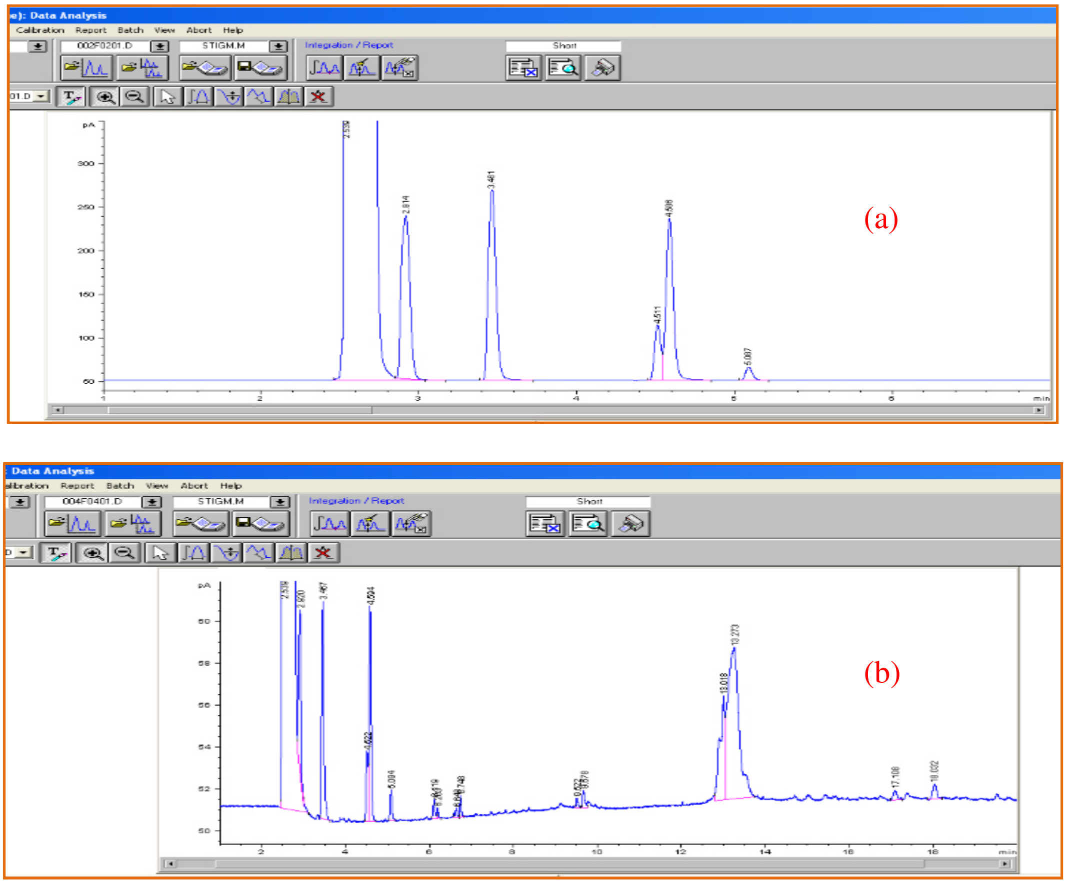

Figure 3: Examples of chromatogram of BTX (a) for calibration and (b) sample

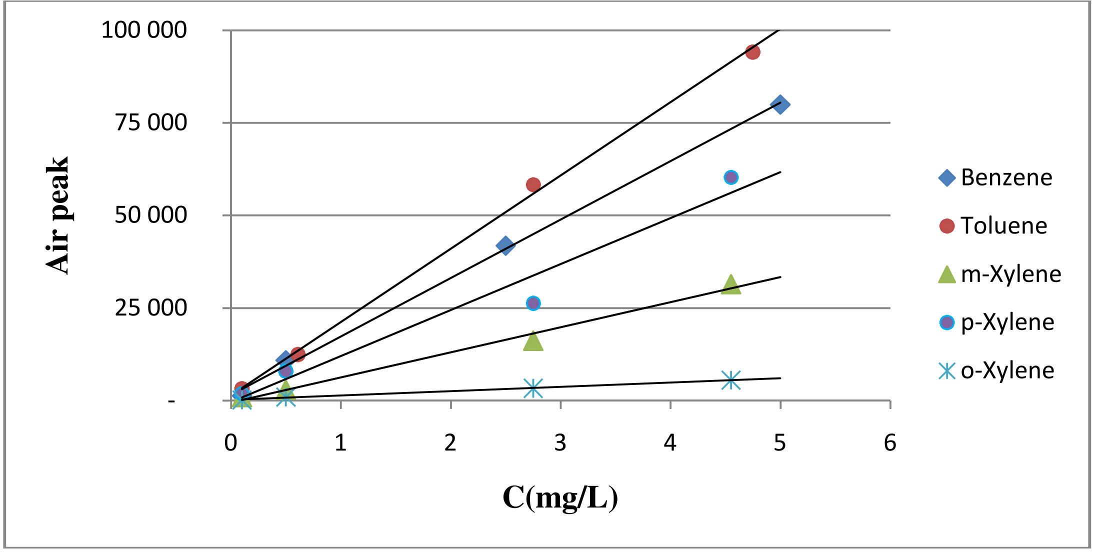

Figure 4: Example of calibration curves used for calculating the concentration of BTX

The concentration in µg.m^{-3} of each aromatic compound P in the sampled air is determined using the following equation:

$$[P] = (m/(Q\times t))\times 1\times 10^{6}$$

where m is the mass of analyte P (in µg) quantified in the extracted solution, Q is the sampling rate (in mL.min^{-1}) and t is the sampling time in min.

3. Results and discussion

The concentrations of the BTX monitored during the 2012 field campaign are summarized in Table 3. The BTX levels are generally related to the road traffic density since all selected sites are roadsides and directly exposed to road traffic emissions.

Table 3: Experimental BTX concentrations (µg.m^{-3}) obtained in the four sampling sites at Anza for the period February 2012-November 2012 (nd : not detected)

Sampling Sites	Benzene	Toluene	m-Xylene	p-Xylene	o-Xylene
S1	7.20	25.19	nd	8.73	1.9
	11.21	48.27	3.98	12.04	2.2
	8.52	21.19	4.4	7.73	0.94
	12.21	35.27	6.98	11.37	3.2
	16.20	39.31	4.4	4.73	4.34
	23.21	55.27	4.98	9.37	4.2
	12.70	29.19	nd	8.73	2.14
	6.56	23.16	nd	5.79	nd
	20.21	46.27	2.98	11.13	4.9
Mean ± SD	**13.1 ± 5.8**	**35.9 ± 12.2**	**4.6 ± 1.3**	**8.9 ± 2.5**	**2.9 ± 1.4**
S2	11.07	32.19	2.73	9.73	2.13
	16.21	59.27	6.98	13.37	5.2
	9.61	17.03	3.87	8.5	1.39
	13.32	41.02	7.0	11.81	3.02
	5.98	23.4	3.06	6.0	1.3
	25.03	78.9	3.7	14.8	2.1
	7.9	24.1	1.1	5.02	2.16
	17.84	35.2	3.4	11.2	3.8
Mean ± SD	**13.4 ± 6.2**	**38.9 ± 20.8**	**4.0 ± 2.0**	**10.1 ± 3.4**	**2.5 ± 1.3**
S3	5.20	25.19	2.73	6.73	2.03
	3.56	13.91	nd	3.79	1.9
	7.80	42.12	3.98	9.37	4.2
	3.79	14.12	1.4	3.91	1.56
	10.04	31.5	3.01	7.66	4.01
	8.37	25.7	nd	6.59	1.23
	11.34	49.9	2.19	8.71	4.95
	11	26.8	2.19	7.45	2.12
	6.31	15.7	nd	4.59	nd
	14.34	49.9	3.49	8.71	4.95
Mean ± SD	**8.2 ± 3.5**	**29.5 ± 13.8**	**2.7 ± 1.0**	**6.8 ± 2.0**	**3.0 ± 1.5**
S4	21.50	41.13	2	16.39	1.9
	27.21	65.27	3.4	20.97	5.2
	15.31	29.8	nd	12.4	3
	23.1	69.93	3.5	19.1	3.8
	12.39	36.03	2.046	7.79	3.6
	6.98	19.3	nd	3.88	0.78
	19.67	55.34	4.07	14.43	5.0
	8.18	20.04	nd	4.28	0.948
	16.16	55.34	4.07	11.43	3.26
Mean ± SD	**16.7 ± 6.8**	**43.6 ± 18.8**	**3.2 ± 1.8**	**12.3 ± 6.1**	**3.1 ± 1.6**

The BTX profiles exhibits similarity in the aromatic levels variation for all sites. This suggests that these emissions have the same origin, in this case the vehicle traffic. Toluene and xylenes were the most abundant aromatic compounds, followed by benzene, at all sampling points. The BTX measurements were carried out by active sampling method known to collect mainly fresh pollutants recently emitted and the results are affected by several factors and antagonistic effects. The level fluctuations recorded in this work may be due to spatial and temporary changes in local activities, traffic flow and meteorology. In fact the morning time was characterized by the lowest photochemical reactions and highest traffic density and beginning of industrial activities, the noon period is the time of high vertical dispersion and of highest photochemical process and the afternoon time is the late period of photochemical reactions with high traffic flow. The wind remains the key parameter for instant dispersion of VOCs.

The seasonal variations are generally dependent on emission types and its variation, meteorological factors, but also on OH/VOCs photochemistry that results in the loss of VOC species from the environment [27]. For the present study and taking into account of the experimental uncertainties, no net seasonal effect was observed.

The benzene average level (Table 4) was 12.9 ± 3.5 $\mu g.m^{-3}$, with a maximum of 25.1 $\mu g.m^{-3}$, toluene mean value was 36.9 ± 6.5 $\mu g.m^{-3}$ and a maximum of 78.9 $\mu g.m^{-3}$. The total xylenes concentration (Σ Xylenes) was 16.2 ± 2.6 $\mu g.m^{-3}$, with a maximum of 29.6 $\mu g.m^{-3}$. The site S4 appears to be the most polluted place, followed by site S2. All sites were impacted by traffic emissions associated with possible emanations coming from the petroleum products storage facility and the cement factory for S4 and S2 respectively. The site S3 is the least polluted place; it is a suburban site and located in windy zone.

The BTX concentration ratios are of great interest to identify their emission sources. Because benzene and toluene usually have the common sources but different atmospheric lifetimes, the B/T ratio is often used as an indicator of fresh emission mainly by vehicular exhausts. The study of Lee et al. shows that T/B ratio increases with increasing traffic volume, industrial emission and other urban sources. The ratios observed in the present work were found to be 0.2-0.6 for B/T and 1.2-4.1 for T/X ratios. The values of B/T were lower, because of high toluene concentration, which indicates that photochemical processes were negligible. The difference of ratio values during morning samples (8:00-10:00) and values during noon or early afternoon (12:00-16:00) samples were also very small. The values of B/T ratios were comparable with those found in Izmir of ~0.5 [27] and Rome 0.36 [29], much lower than those observed in Taiwan, between 2 and 5 [30]. The differences of BTX ratios found in these cities are probably due to differences in vehicle fleet, fuel composition and its quality, and industrial activities.

The BTX levels were higher than the usual concentrations measured in urban outdoor [7, 28-30] but they were in the range of values measured in similar urbano-industrial environment [26, 31-33]. BTX levels found in this study at Anza site are comparable for instance to the ones observed in Agra (benzene: 14.7 $\mu g.m^{-3}$; toluene: 8.1 $\mu g.m^{-3}$; o-m xylenes: 2.1 $\mu g.m^{-3}$; p-xylene: 5.1 $\mu g.m^{-3}$) [32], Mumbai (benzene: 13.4-38.6 $\mu g.m^{-3}$; toluene 10.9-33.5 $\mu g.m^{-3}$) [33], or Taiwan (benzene: 13.6 $\mu g.m^{-3}$; toluene 54.49 $\mu g.m^{-3}$) [34].

Table 4 presents the comparison of benzene and toluene concentrations observed at Anza in 2007 and 2012 campaigns. The 2012 values were relatively lower than 2007 values [20-21]. This might be considered as a direct consequence of the drastic measures adopted by the authorities in favor of the protection of the environment, such as the use of cleaner fuel, the replacement of old non-catalytic by catalytic vehicles. In addition, several production units left the Anza site since the 2007 measurement campaign.

Table 4: Average levels ($\mu g.m^{-3}$) of BTX in ambient air of Anza locality

Year	Benzene		Toluene		(*m*+*p*+*o*)-xylenes		Σ BTX	ref
	Max	Mean	Max	Mean	Max	Mean		
2007	34.4	20.5	87.2	43.7	-	-	-	[20-21]
2012	25.1	12.9 ± 3,5	78.9	36.9 ± 6.5	29.6	16.2 ± 2.6	65.9	this work

Ozone, NO and NO_2 ambient air levels in the area under study (sites S2, S3 and S4) were measured using gas analyzers (Environment S.A.) from 10:00 to 16:00 during 4 days in July 2012. The results are shown in table 5. The NO/NO_2 ratio is relatively low which suggests a high photochemical activity, leading to the production of NO_2. Ozone levels in S2 (51.6 $\mu g.m^{-3}$) and S3 (67.2 $\mu g.m^{-3}$) were relatively lower than that observed in S4 (82.1 $\mu g.m^{-3}$). Site S2 is an urban site, and subsequently ozone might react with other pollutants including NO. While S3 can be considered as a suburban site, due to its particular position in a windy zone, O_3 observed in site S4 can be assigned to local pollution because of its exposure to air mass coming from the Agadir harbor, not far from Anza.

As regards ozone production, VOC/NOx pollution have a high photochemical ozone formation potential [35]. With a short atmospheric lifetime (~8 h) and high reactivity, the xylenes are major contributors to O_3 formation among BTX, whereas benzene and toluene are considered to be relatively stable molecules with lifetimes of ~12 and 2 days respectively [36], and usually remain long in the ambient air after release. In addition, the benzene and toluene depletion in the atmosphere via their ozonolysis reactions ($<10^{-20}$ cm^3 $.molec^{-1}.s^{-1}$) [35] and their reactions with NO_3 radicals ($< 10^{-16}$ $cm^3.molec^{-1}.s^{-1}$) are negligible [35]. Thus, their atmospheric removal is due to their reaction with OH radicals (1.2×10^{-16} for benzene and 5.9×10^{-12} $cm^3.mol^{-1}.s^{-1}$ for toluene) [35].

Table 5: Average concentrations of NO, NO_2 and O_3 between July 4th and July 8th 2012, 10:00-16:00, at Anza

Sampling sites	Average concentration ($\mu g.m^{-3}$)					
	S2		S3		S4	
	Max	mean	max	mean	max	mean
NO	69.4	36.3	102.8	87.1	89.5	52.5
NO_2	86.4	62.5	137.2	79.7	97.4	67.9
O_3	102.0	51.6	114.9	67.2	135.7	82.1

Conclusion

Benzene, toluene and xylenes were measured in four places of the Anza locality, NW of Agadir, Morocco, carefully chosen to reflect the major activities close to the site. Active sampling method was used to collect air samples containing the most freshly and recently emitted pollutants. These air samples have not had the time to manifest their photochemical potential, especially when the sampling point is not very far from the edge of the roads. The results show high level of benzene, toluene and xylenes. These levels were sufficient to represent a degree of risk to the local residents. The site S4 and S2 were the most polluted places, due their proximity of pollution sources. No net seasonal variation was recorded and the value fluctuations were probably due to emissions variations and meteorology. The values measured during 2012 were lower than those already measured in 2007, which might be attributed to the drastic measures adopted by the Moroccan authorities in favor of the protection of the environment, especially to improve the air quality.

Acknowledgments - The authors acknowledge financial support from the Agadir Urban Community (Quantification of VOCs in Anza project (2008-2010)).The authors thank particularly the President of AUC (Agadir urban community) and Dr. F. El Baz. The authors thank also the local authorities for their collaboration (Wilaya of Agadir, the District of Anza), and the UATRS/CNRST service.

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