Evaluation of Nitrogen dioxide,Benzene, Toluene, Ethylbenzene and Xylene concentrations in the urban environment of Meknes city, Morocco

Abstract

The present study focuses on the monitoring and evaluation of air quality in the city of Meknes, based on the tracers of pollution of proximity cars (NO_2 (nitrogen dioxide) and BTEX (Benzene, Toluene, Ethylbenzene and Xylene). In the absence of a telemetric station and a mobile laboratory within the city, the use of passive diffusion tubes, which are analyzed offline in the laboratory after exposure, was the most plausible solution. The low cost, ease of implementation of the technique, and the possibility of covering a large geographic area with a large number of samplers all argue for the adoption of this approach.

The coupling of the results of the measurement campaigns and the counting sessions under geographical information system allowed to determine the zones most affected by the automobile pollution and to carry out a cartography at high scale of spatial resolution of the prospected pollutants.

The deployment of passive sensors at 14 measurement sites revealed the existence of a spatio-temporal variability of the studied pollutants. This variability is due to the climatic conditions that prevailed during the measurement campaigns, notably wind and precipitation, to the photochemical nature of the said markers, to the proximity to the emission sources, to the type of measurement site and to the topography and road infrastructure.

The results of our study show that air concentrations of NO_2 and BTEX reach maximum values in the city center and decrease towards its periphery. Their concentrations at the car proximity sites exceed those at the background level. The concentrations recorded in summer are much lower than in winter.

A slight increase in levels of xylene and ethylbenzene isomers was observed at background sites near industrial areas.

The approach developed could be used as a decision-making tool for the competent authorities in this field and adapted for the monitoring of other types of pollutants (pesticides, tracers generated by industrial units, furan dioxins, etc.).

Key words: Passive diffusion tubes, air quality, nitrogen dioxide, BTEX, Meknes

I. Introduction

Air is essential for the survival of any living being. The human being consumes on average every day 14 kg of air for 1.5 kg of food and 2 kg of water (Ait Bouh, 2012). Man can subsist 5 weeks without eating, 5 days without drinking and less than 5 minutes without breathing (Ait Bouh, 2012). The deterioration of air quality, impacts human health by increasing the incidence of cardio-respiratory pathologies (Shah et al., 2013; Lee et al., 2014; Song et al., 2014; Almagro et al., 2015), premature deaths (Atkinson et al., 2014; Cesaroni et al., 2014; Thurston et al., 2016), and a number of cancer (Liu et al., 2009; Turner et al., 2014; Turner et al., 2016). Environmental acidification (Smith et al., 2001), ecosystem disruption (Brakke, 1994), photochemical pollution (Hallquist et al., 2017), eutrophication (Smith et al., 1998; Hautier et al., 2009), altered plant growth (Proietti, 2016; Sicard et al., 2017), and degradation of the built environment (Vallet et al., 2006) are the harms caused by air quality degradation.While the problems of air pollution are not new they only gained momentum with the industrial revolution of the 19th century (Caruana, 2017). In the 14th century, being aware of the negative impacts of air pollution, King Edward II of England (1307-1327) had taken corrective measures against polluters with coal (Aubier and Lambrozo, 2000). Among the dramatic events linked to the degradation of air quality that marked the 20th century and increased interest in the various aspects of air pollution, we can cite the disaster in the Meuse Valley near the city of Liege in Belgium where sixty people died in December 1930 as a result of the increase in sulfur dioxide (SO_2) levels (Aubier and Lambrozo, 2000; Nemery et al., 2001; Nemmar et al., 2003). Exposure to levels of the same tracer ranging from 1400 to 5500 $\mu g/m^3$ in 1948 resulted in 20 deaths in Donora, Pennsylvania (Schrenk et al., 1949; Aubier and Lambrozo, 2000; Nemmar et al., 2003). In 1952, nearly 4,000 people succumbed to London smog resulting from increased SO_2 levels (1260 $\mu g/m^3$) combined with particulate matter concentration (2650 $\mu g/m^3$) within 24 hours (Aubier and Lambrozo, 2000; Bell and Davis, 2001; Nemmar et al., 2003; Roussel, 2015). The number of recorded deaths exceeded 5 times the seasonal average, and two-thirds of the victims were over 65 years of age, 80% of whom had pre-existing cardiorespiratory problems. In 1984, the city of Bhopal in India suffered 3828 deaths following the explosion of a factory specialized in the production of phytosanitary products by releasing more than 40 tons of methyl isocyanate into the atmosphere (Kurzman, 1987). To this can be added the explosion of the two nuclear reactors, Chernobyl and Fukushima (Brumfie, 2013; Yajima et al., 2014) whose damage is very difficult to determine so far.To avoid the reproduction of this kind of problems that cost the lives of hundreds or thousands of people (Aubier and Lambrozo, 2000; Nemery et al., 2001; Nemmar et al, 2003; Roussel, 2015) and mitigate the dramatic impacts of air pollution, several actions have been implemented by the international community: The adoption of renewable energies (hydro, wind, photovoltaic and geothermal); The replacement of fossil fuel vehicles with electric and hybrid cars; The banning of the use of certain chemical compounds (chlorofluorocarbons (CFCs), persistent pesticides,...); The creation of low emission zones, eco-neighborhoods and the implementation of alternating traffic and urban tolls; And the adoption of environmental protocols such as the Kyoto Protocol which set the thresholds of atmospheric emissions of each country and which gave birth to CDM projects (clean production mode). Despite all the actions taken, air pollution still represents a major public health issue and continues to claim millions of victims. According to the latest WHO report, 7 million deaths are caused each year by ambient and indoor air pollution (WHO, 2018). Among the serious disorders identified: respiratory (asthma, COPD, bronchiolitis, bronchitis, rhinitis, pneumonia,...), cardiovascular (stroke, myocardial infarction, ischemic heart disease, arrhythmia, heart failure, high blood pressure,...), reproductive system (decreased male fertility, increased intrauterine mortality and premature births) and cancers. The scientific literature on this subject is very rich but concerns, essentially, Europe, North

America and Asia. In Morocco, air pollution is not sufficiently evaluated and the work done so far is often ad hoc and not very representative (Ministry of Health, 1998; Casa-Airpol, 2000; Mohammedia-Airpol, 2002; Croitoru and Sarraf, 2017).Research studies aimed at characterizing air quality have been carried out in several Moroccan cities in order to fill the existing gaps in air quality (Bounakhla, 1998; Bounakhla et al., 1999; Bounakhla et al., 2003; El Abidi et al., 2000; Ouarzazi, 2003; El Khoukhi et al, 2004; Zaghaid et al., 2009; Zouir, 2009; Ouhakki, 2009; Ait Bouh, 2012; El Ghazi, 2012; Monna et al., 2012; Belhassan, 2013; Tahri et al., 2013; Ouali Alami et al, 2014a; Ouali Alami et al., 2014b; Boularab et al., 2015; El Rhzaoui et al., 2015; Khouddane et al., 2015; Khouddane et al., 2016; Inchaouh et al., 2017; Boularab, 2018). At the national scale, air quality monitoring is carried out by a telemetric network consisting of 29 fixed stations distributed in 15 cities belonging to 7 regions. To this are added two mobile laboratories.In the absence of a telemetric station in the city of Meknes, the use of passive diffusion tubes for air quality monitoring and assessment is justified. The city of Meknes is characterized by a fragmented industrial sector, weakly developed and little income generator. Despite its modest economic weight and its fairly limited social impact, industry is one of the main sources of environmental pollution. In addition to this, the transport sector whose emissions contribute significantly to air pollution and the degradation of the urban environment (MRE, 2002; Abdouh et al., 2004; AUM, 2010; ORE, 2012; AUM, 2013; Boularab, 2018). It is in this context that the present work, which adopts an interdisciplinary methodological approach that aims at the surveillance and monitoring of ambient air quality at the level of the city of Meknes. The second added value of this study is the elaboration of a database that could be used by the competent authorities to take the necessary decisions in the fight against air pollution. To achieve these objectives, we adopted the passive sampling technique for the collection of tracers of vehicular proximity pollution in the absence of an air quality measurement and monitoring station. This easy-to-implement method allows for coverage of a large geographic area with a large number of samplers at low cost which allows for high spatial resolution (neighborhood scale) mapping (Brown, 2000; Garcia-Fouqué et al., 2000; Adon et al., 2010; Pienaar et al., 2015). We were interested in several types of tracers in this work, including volatile organic compounds including BTEX (Benzene, Toluene, Ethylbenzene and Xylene) and nitrogen dioxide (NO_2). Two types of axially symmetric passive diffusion tubes were used at each proximity (transport) and background site. Perkin Elmer tubes for BTEX trapping and Palmes for nitrogen dioxide sampling (Atkins, 1990; Garcia Fouqué et al., 2000; Meybeck et al., 2000; Plaisance, 2004; Laurinavičienė and Dėdelė, 2013). After exposure the tubes are analyzed off-line in the laboratory. In parallel with the measurement campaigns, motor vehicle counting sessions were carried out at the main roads of the city of Meknes. The results obtained were introduced into a geographical information system (GIS) which could be used as a database in the future. We have adopted the following approach: Identify and quantify the major sources of air pollution; To determine the share of the transport sector in the air pollution at the level of the city of Meknes through the surveillance and the monitoring of the tracers of the automobile pollution (NO_2 and COV) and the realization of sessions of counting of the motor vehicles at the level of the principal road axes; To carry out a cartography with high spatial resolution of the contents of the prospected pollutants; To compare the results obtained with the standards of national and international quality of the air.

II.Materials and methods

II. 1. Pollutants prospected

II. 1.1. BTEX

BTEX are monocyclic aromatic hydrocarbons of the family of volatile organic compounds. They are colorless liquids, slightly soluble in water, flammable and highly volatile. They are present in many household products (solvents, detergents, paints, glue, inks, pharmaceutical products, cosmetics, cigarettes and pesticides,...). They are emitted by gasoline-powered vehicles as well as in exhaust fumes. As they can also be generated by industrial activities such as oil refining, paint production, glue, plastic, nylon, detergents, cosmetics and drugs (INERIS, 2004). In addition, there are storage and distribution units for petroleum products.

II. 1.2. NO_2

NO_2 comes mainly from the combustion of fuels (gasoline, petrol, coal, fuel oils). High temperatures catalyze the NO_2 formation reaction by combining nitrogen (atmospheric and contained in fuels) and atmospheric oxygen. The transport sector remains the main contributor to these emissions.

II.2 Effects on health and the environment

II.2.1. Toxicological aspect

By inhalation, BETX can cause various disorders in humans, which depend on the nature, concentration and duration of exposure, but also on the sensitivity of the individuals (INRS, 1992; SFSP, 1996).

The short- and medium-term effects, depending on the doses of BETX inhaled, are reflected in various symptoms: irritation of the respiratory tract and eyes, headaches, vision, hearing and memory problems, abdominal pain, convulsions, etc. These symptoms can lead, in case of high absorbed doses, to a coma or even to death.

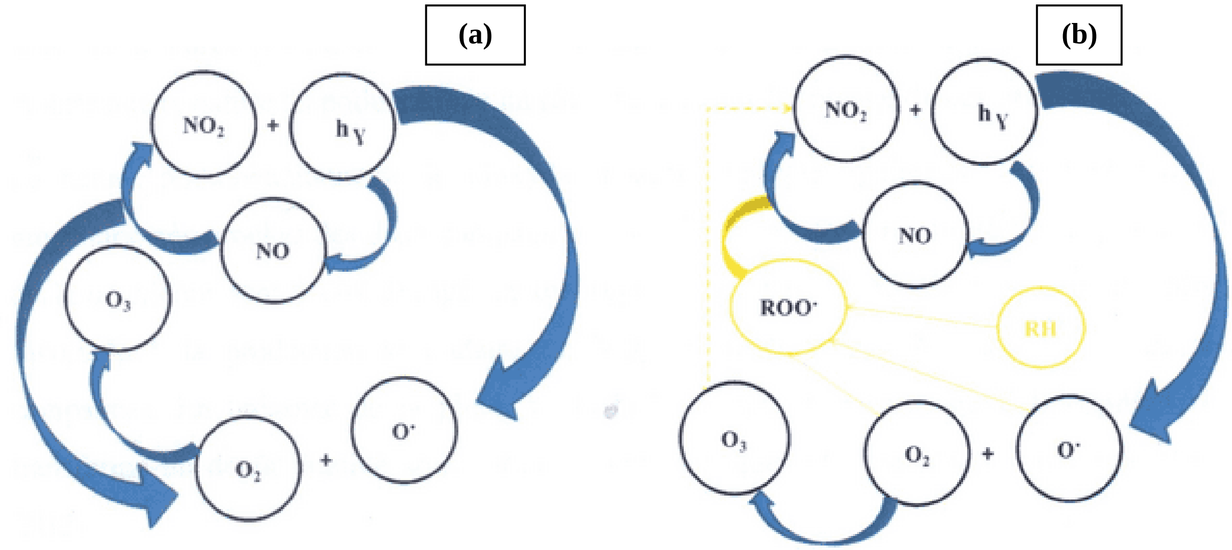
Numerous studies have been carried out and are underway to identify the complex mechanisms of hematological toxicity and carcinogenicity of benzene (Snyder and Kalf, 1996). The WHO has established risk threshold values for benzene and toluene (WHO, 1996). The mutagenic character of benzene is due to its lipophilic properties which allow it to penetrate cells where it is oxidized (Zhang et al., 1996; Lovern et al., 1997). The metabolites formed can then insert themselves into DNA molecules (Andreoli et al., 1997) and cause errors in gene transcription which, in the few cases where they are not corrected, generate undesirable proteins that can lead to a tumor (INRS, 1992; SFSP, 1996).

Benzene can also cause benzolism, which is a reduction in the number of white and red blood cells and platelets. This causes headaches, fatigue, dizziness, anorexia, and, in extreme cases, can lead to haemorrhage and even death. This disease is quite exceptional in developed countries due to the reduction of benzene exposure rates. Toluene and xylenes have a lesser impact on health. Their oxidation takes place preferentially on the methyl groups with the formation of an acid function which confers a hydrophilic character to the molecule which will be easily eliminated by the body. The risk of penetration of the compound into the cell is thus strongly attenuated (SFSP, 1996).

Nitrogen dioxide has an impact on respiratory functions. It penetrates the fine ramifications of the respiratory system and can cause bronchial hyperreactivity in asthmatics (SFSP, 1996).

II.2.1. Environmental impact

In addition to their direct action on humans, BTEX and NO_2 generate, by reaction in the troposphere with free radicals, other so-called secondary pollutants, such as ozone or aldehydes, which have harmful effects on humans and their environment (INRS, 1992; SFSP, 1996). In the troposphere, in the absence of volatile organic compounds, the O3 content is governed by the NO-NO_2-O_3 ozone formation-destruction reaction cycle shown in Figure 1a, p. 6. Ozone is formed by combining O_2 with an oxygen atom from the dissociation of NO_2 under the effect of radiation of wavelength less than 400 nm. This cycle is completed by the reaction of NO on the ozone previously produced. There is thus a photostationary equilibrium between NO, NO_2 and O_3. On the other hand, in the presence of VOCs (identified as RH in figure 1b, p. 6), this equilibrium is disturbed by the privileged conversion of NO into NO_2. The 'OH' radicals react with RH and give rise to alkyl radicals which, by a series of rapid reactions with O_2, lead to the formation of peroxide radicals R02' which, in turn, rapidly oxidize NO to NO_2 with regeneration of the 'OH' radical; O_3 is thus no longer consumed by NO: the system can become productive in O_3, It thus follows an increase in tropospheric ozone, and an indirect production of aldehydes or nitrated compounds, whose impacts on human health and on the environment are notable (Dégober, 1992).



a. Chapman cycle in the absence of VOCs
b. Chapman cycle in the presence of VOCs

Figure 1. Chapman's cycles

I.3. Sampling protocol

II. 3.1. NO_2 sampling

For NO_2 monitoring, passive diffusion tubes with three stainless steel grids impregnated with triethanolamine (Atkins, 1990; NBN EN 16339) were deployed at 14 sampling sites. The choice of passive sampling is based on the ease of implementation, the operation without electrical connection or use of a pump, and the possibility of covering a large geographical area with a large number of samplers at low cost. The samplers used consist of 74 mm long, 9.5 mm diameter acrylic tubes.

The samplers were fixed vertically to straight supports, 3-4 m above ground level (NBN EN 16339). A specific shelter was used to protect the sampling equipment from weather and wind influence (Plaisance et al., 2004). After two weeks of exposure, the Palmestubes were lifted and the NO_2 trapped in the grids was extracted via the addition of 3 ml of Saltzman's reagent (Atkins, 1990; NBN EN 16339) and its amount in the extracts was then determined by UV for a wavelength of 542 nm.

II.3.2 Sampling of BTEX

For BTEX, the tubes used are of the Perkin Elmer type. They have a length of 90 mm, an external diameter of 6.35 mm and an internal diameter of 5 mm. Each tube has a unique identification number engraved. They are hermetically sealed, before and after sampling, with Swagelok® type brass caps, including Teflon ferrules. The exposure time of these tubes is 30 days. Sampling and analysis are performed according to ISO 16017-2: 2003. After sampling, the cartridges are analyzed by thermal desorption followed by gas chromatography with a mass spectrometer as detector (**Figure 2**).

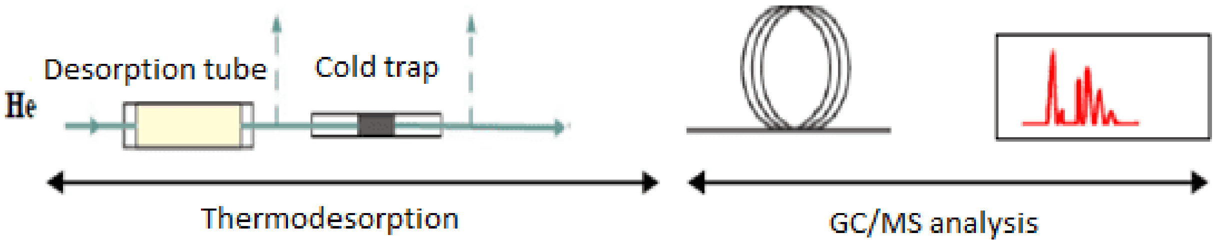


Figure 2. Schematic representation of the analysis steps of a Perkin Elmer tube

II. 4. Location of the sampling sites

Passive sensors were deployed at 14 measurement sites divided into automotive proximity (P) and background (F) sites (Figure 3 and Table **1).**

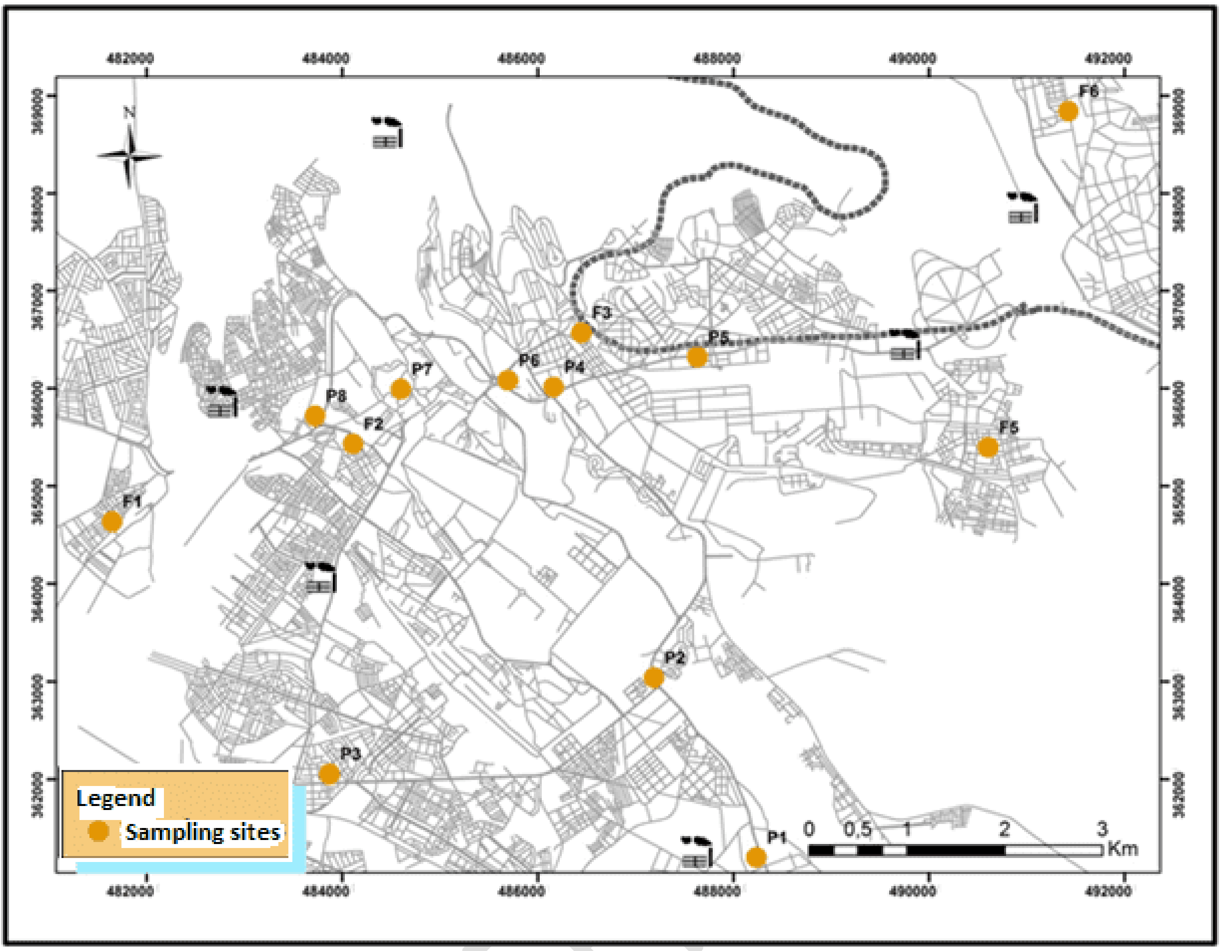


Figure 3. Location of sampling sites

Table 1. Distribution of sampling sites by type and location

points	Lieu	Typologie
P1	Point of intersection between the national road n°13 and the national road n°6	Traffic
P2	Point of intersection of Bair Anzarane Avenue and Zitoune Boulevard	Traffic
P3	Zitoune Avenue (Marjane district)	Traffic
P4	Point of intersection of Bairanzarane Avenue and the Avenue of the Royal Armed Forces	Traffic
P5	Avenue of the Royal Armed Forces near the main railway station	Traffic
P6	Point of intersection of Mohammed VI Avenue and the Avenue of the Royal Armed Forces	Traffic
P7	Daar smane Street, the point where the old Medina meets the new city	Traffic
P8	The bus station of the city of Meknes	Traffic
F1	Toulal	Background
F2	Riad	Background
F3	New city near the station of el Amire Abdelkader	Background
F4	Hacienda	Background
F5	El Bassatine	Background
F6	Wisslane	Background

II. 4.Road traffic counts

In parallel with the winter measurement campaign of the tracers of the pollution of proximity cars (NO_2 and BTEX), sessions of counting of the road traffic were carried out in the principal roads of the city. The counting was carried out by operators equipped with manual clickers (Figure 4).

Figure 4. Manual clickers used for traffic counting in the city of Meknes

For the study of intraday variations, we focused our census mainly on the peak hours: morning (7:30/9:30), noon (11:30/14:30) and evening (17:30/19:30). The counts were carried out over 15-minute periods.

In order to study inter-day variations and fluctuations in road traffic during working and non-working days, one week of counts per site was required. Twelve traffic sites were selected and their location was dictated by the nature of the road section and the location of the passive samplers (**Figure 5**).

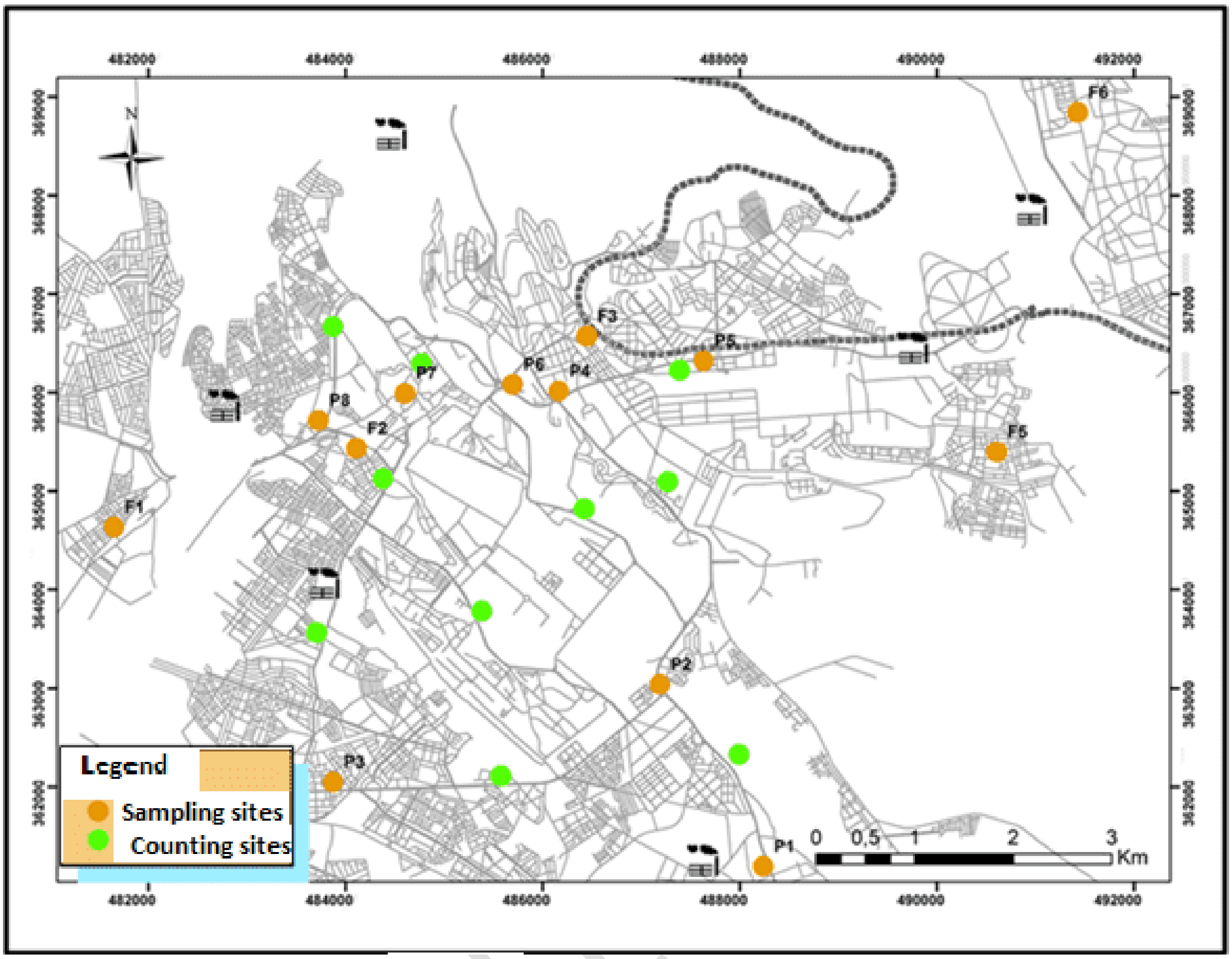


Figure 5. Location of the counting point

II.5. Mapping of NO_2 and BTEX levels

Mapping of NO_2 and BTEX concentrations was performed by spatial interpolation using the inverse distance weighting (IDW) method. Interpolation is a technique that allows estimation of spatially continuous variables at unknown locations from values measured at specified locations.
IDW is one of the most widely used deterministic interpolation methods. The IDW function generates the interpolated surface by estimating NO2 and BTEX concentrations at unsampled points, which is based on linear combinations of values at sampled points weighted by an inverse function of distance (Shepard, 1968).The assumption of this technique is based on the fact that the difference between the value of the measurement point and the values of the unmeasured points is relative to the distance. The weights (wi) are expressed as follows:

$$w_i = 1/d_i^p \Big/ \sum_{i=1}^{n} 1/d_i^p$$

Where di is the distance between the point of interest x_0 and the sampled point x_i, p is a power parameter, and n represents the number of sampled points used for estimation.
The main factor affecting the accuracy of the IDW is the value of the power parameter (p), which expresses the relative importance of nearby points compared to distant points (Li et al., 2008). Indeed, the weight decreases as the distance increases, especially when the value of the power parameter is high, so that the neighboring samples have a heavier weight and more influence on the estimation. The selection of the power parameter is arbitrary, but the most common choice is equal to 2 and the resulting method is often called the Inverse Distance-Squared/IDS.

II.6. Data processing and statistical analysis

The characterization of the pollution due to NO_2 and BTEX in the city of Meknes requires a spatio-temporal analysis, by determining its sources as well as the possible factors which can influence its concentration and its dispersion. To achieve this, the data from the field forms and the laboratory results were recorded and processed using the Excel spreadsheet.
In addition to the data on NO_2 concentrations, data on the main climatic factors (Temperature, Precipitation and Wind) were imported from the platform www.wunderground, for both study periods.
These different data were summarized and represented by adopting the basic statistical indicators (Mean, Minimum and Maximum). The data on wind directions and speeds were used to develop wind roses using the openair package of the R statistical software (www.r-project.org).
Finally, a mapping of the dispersion of NO_2 and BTEX at the level of the city of Meknes was carried out by spatial interpolation according to the method of the weighting by the IDW. The IDW function generates the interpolated surface by estimating the concentration of NO_2 and BTEX at unsampled points, which is based on linear combinations of values at sampled points weighted by an inverse function of distance (Shepard, 1968).

III. Results and discussions

III.1. Meteorological conditions

Wind directions and speed

Wind distribution affects the spatial spread and distribution of pollutants. In our study, the average wind speed was 7.97 km/h during the summer period, with the dominant directions being northwest and west with moderate intensities ensuring good pollutant dispersion (**Figure 6a**).

However, during the winter campaign the average wind speed is 9.6 km/h and almost all directions are recorded, nevertheless the most frequent directions are South-East and East, and more violent come from East and North-East with a maximum speed of 37 km/h (**Figure 6b**). Such a multidirectional wind can produce localized pollution and plume deposition, which can be seen very clearly in the maps.

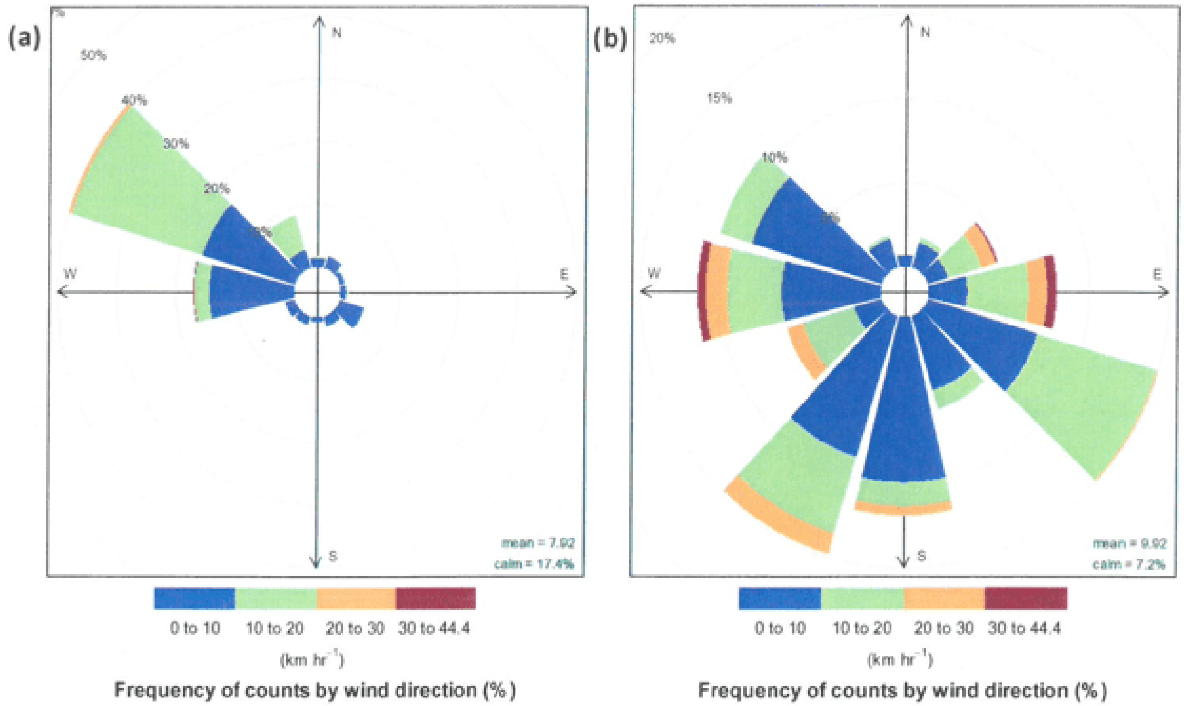


Figure 6. Frequency distribution of wind direction and speed during (a) The summer period and (b) the winter period

Temperature

During the summer period, the recorded temperatures show a maximum of 39 C° and a minimum of 15 C° with an average of 24.31 C°. However, the winter period recorded a maximum temperature of 20 C°, a minimum temperature of 1C° and an average of 9.31 C°. These findings are consistent with the city's weather pattern.

Precipitation

Rainfall is a major contributor to the atmospheric leaching process and the removal of pollutants. When interacting with H_2O, NO_2 reacts to form a mixture of nitrous and nitric acids that can be leached by rain, unlike NO. These effects explain the difference in concentrations between campaigns conducted in different seasons (Stern, 1968; Hill, 1974).

During both campaigns, no precipitation was recorded except for a few showers on 12/29/2014.

III.2 Study of NO_2 concentrations

The highest concentrations of NO_2 detected, were measured at the level of the sites of car proximity (41.68 µg/m^3). On the other hand, the lowest levels were recorded at the background sites (20.47 µg/m^3) (**Figure 7**).

The average NO_2 concentration measured during the summer campaign (31.96 µg/m^3) is very close to that reported in winter (31.71 µg/m^3) (**Figure 7**).

In winter, the average NO_2 concentration noted at the car proximity sites is equal to 39.71 µg/m^3 while that of the background sites is 23.72 µg/m^3.

In summer, the average NO_2 concentration was 46.71 µg/m^3 at the car proximity sites and 17.21 µg/m^3 at the background sites.

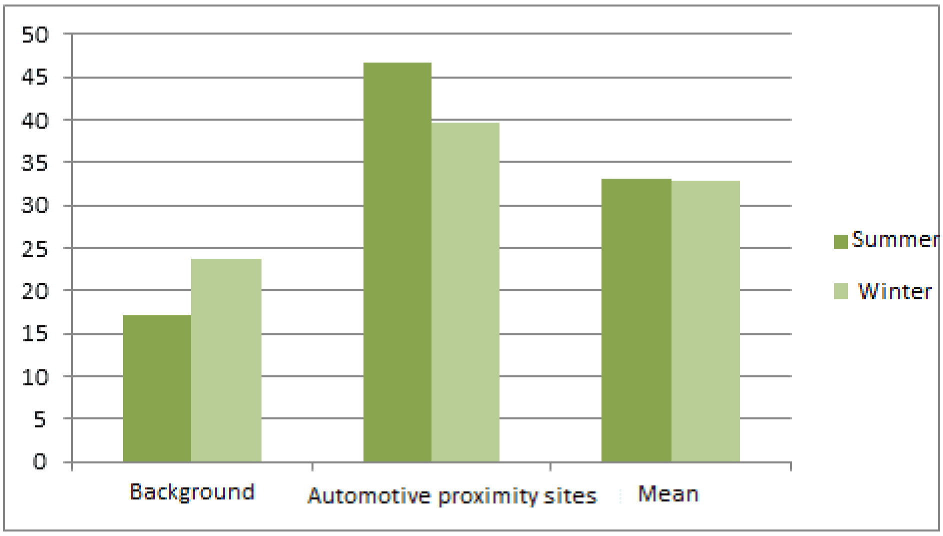


Figure 7. Average NO_2 concentrations

All background sites have NO_2 levels below 40 µg/m^3, the European limit value (Directive 2008/50/EC). Five out of 8 sites in the vicinity of the car have levels exceeding 40 µg/m^3. Among these sites, two exceed the Moroccan limit value of 50 µg/m^3.

Atmospheric concentrations of NO_2 reach maximum values in the city center and tend to decrease towards its periphery (**Figure 8**). The highest concentrations recorded correspond to sites near the city center: P7 located at the level of Dar Smane Street (58.37µg/m^3), P4 which represents the intersection between the Avenue of the Royal Armed Forces and Bir Anzarane Avenue (57.96 µg/m^3), P2 point of intersection of Bir Anzarane Avenue and Zitoune Boulevard (48.23 µg/m^3) and P6 intersection of Mohammed VI Avenue and Royal Armed Forces (45.12 µg/m^3). Indeed, these roads are characterized by heavy daily traffic, which is the source of a large part of NO_2 emissions (Meybeck et al., 2000; Laurinavičienė et al., 2013). The high levels reported at site P7, located at Dar Smane Street, used by as many as 12000 vehicles per day exceed those measured at site P4, an intersection point crossed by more than 24000 vehicles per day. This can be explained by the structure of the street which is not conducive to a better dispersion of NO_2 and favors its accumulation. The height/width ratio at this street is greater than 1, which allows it to be described as a canyon street (Meybeck et al., 2000). The sites with concentrations below 15 µg/m^3 are Wisslane (F6) and Toulal (F1) given their geolocations. For sites F6 and P1, despite their proximity to the Lafarge cement plant and the industrial district of Sidi Bouzekri, we found that the measured levels are below the permissible limit value. These results are in agreement with those of the surveys carried out by the Moroccan Ministry of the Environment, which showed that road traffic is responsible for 75% of NO_2 emissions and that the industrial sector does not exceed 25% (MRE, 2001). This also rhymes with the study carried out in the city of Toulouse, which showed that the highest NO_2 levels were reported at the car proximity sites in the city center (Meybeck et al., 2000). Another study conducted at the level of the city of Kaunas, the second largest city in Lithuania, showed that NO_2 emissions are strongly correlated with road traffic density (Laurinavičienė et al, 2013).

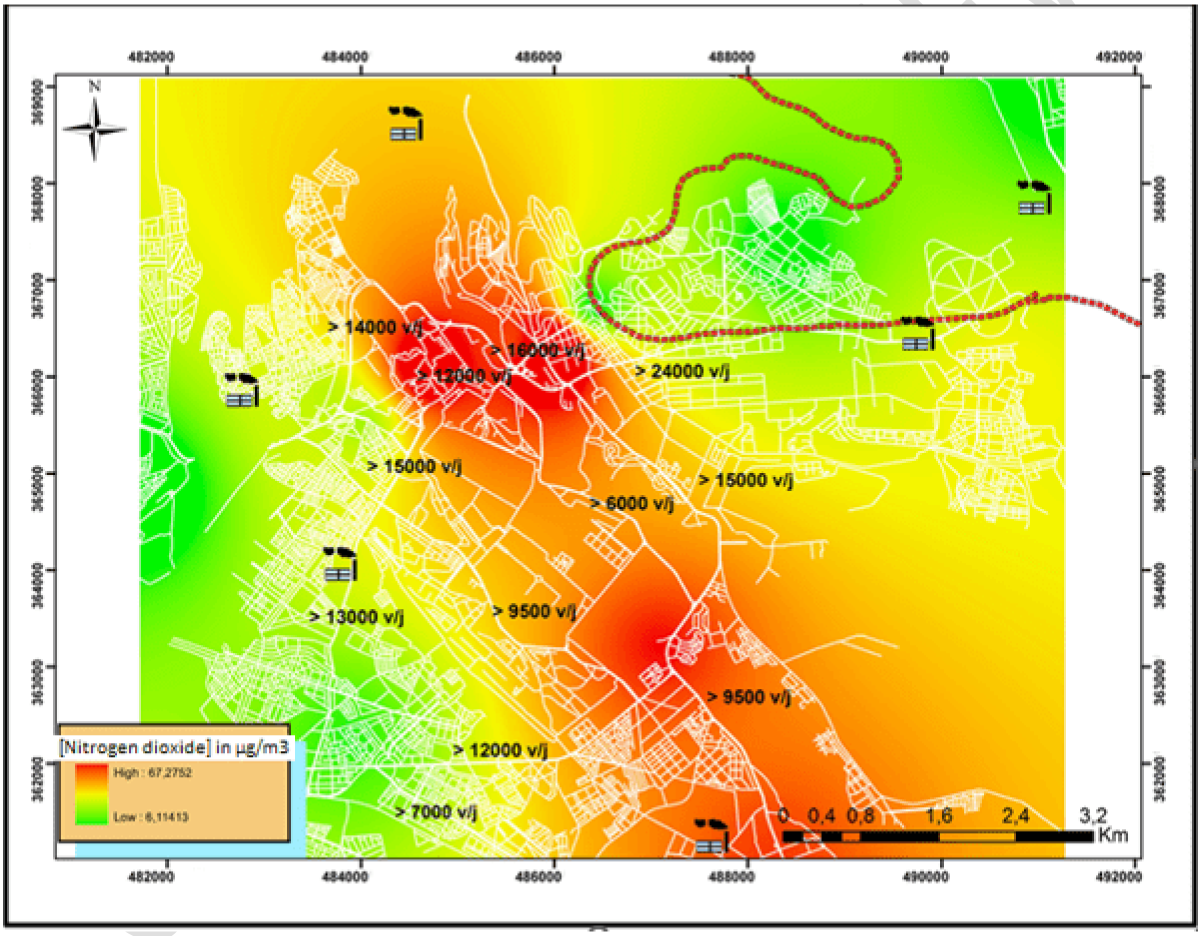


Figure 8. Spatial variations in NO_2 concentrations

According to this study, the city of Meknes appears as a moderately polluted city compared to other urban sites. The average NO_2 concentrations obtained are very similar to those of Elche (Spain), Edinburgh (United Kingdom) and Granada (Spain) with almost the same population (Table 2). These concentrations are in the range of large agglomerations with populations over one million such as Kanpur (India) and Bamako (Mali). On a national scale, a similar passive tube study was conducted in other Moroccan cities such as Casablanca, Rabat, Marrakech, where the level of nitrogen dioxide is higher than in Meknes (**Table 2**). However, this average concentration is comparable to that of Fez (DSPCT, 2010).

These observed disparities between cities could be attributed to differences in urban structure, traffic flows, pollutant emitters, and climatic conditions (Lewné et al., 2004).

The following table shows nitrogen dioxide concentrations measured worldwide by the passive sampling technique, as well as by automatic monitoring networks in some neighboring countries.

Table 2. NO_2 results were obtained in the city of Meknes compared with other urban agglomerations worldwide.

Study Area	$[NO_2]$ $\mu g/m^3$	Study Period	Country	References
Kocaeli	14	July 2006	Turkey	(Pekey et al., 2013)
Bouni Region	14.8 *	Average of 7 months of measurement in 2011	Algeria	(Fadel et al., 2011).
Windsor	23.31	Average of four 14-day campaigns in February, May, August and October 2004	Canada	(Wheeler et al., 2008)
Malaga	22.8	September 2001 and from December 2001 to February 2002	Spain	(Lozano et al., 2011)
Pampelune	23	From June 2006 to 2007	Spain	(Parra et al., 2009)
Gothenburg and Mölndal	23.5	7–20 May 2011	Sweden	(Habermann et al., 2015)
Asturies	23.6	Average of two 7-day campaigns in June and November 2005	Spain	Fernandez-Somoano et al., 2014)
Northern Ireland	24.3	Annual average for 1997	UK	(Stevenson et al., 2001)
Kampala	24.9	From 30 June to 13 July, 2014	Uganda	(Kirenga et al., 2015)
Kocaeli	25	January 2007	Turkey	(Pekey et al., 2013
Wales	27.26	Annual average for 1997	UK	(Stevenson et al., 2001)
Scotland	27.26	Annual average for 1997	UK	(Stevenson et al., 2001)
Bamako	30.45	From June 2008 to 2009	Mali	(Adon et al., 2010)
Meknes	31.83	From 14 July to 28 July 2014 and from 25 December 2014 to 12 January 2015	Morocco	This study
Elche	32	Average for 2007–2008	Spain	(Caballero et al., 2012)
East Anglia	34.78	Annual average for 1997	UK	Stevenson et al., 2001)
South East England	34.78	Annual average for 1997	UK	Stevenson et al., 2001)
Edimbourg	34	From 2 December 2013 to 13 January 2014	UK	(Lin et al., 2016)
West Midlands	35.72	Annual average for 1997	UK	(Stevenson et al., 2001)
Granada	36.5	Average of two campaigns: from July to September 1999 and from December 1999 to February 2000	Spain	(Lozano et al., 2011)
Kanpur	36.9	February and March 2004	India	(Behera et al., 2015)
Edimbourg	37	From 2 August to 13 September 2013	UK	(Lin et al., 2016)
East Midlands	40.42	Annual average for 1997	UK	(Stevenson et al., 2001)
Yourkshire-and-Humber	42.3	Annual average for 1997	UK	(Stevenson et al., 2001)
London	42.3	Annual average for 1997	UK	(Stevenson et al., 2001)
Agadir	44	From 20 April to 27 April, 2006	Morocco	(Chirmata et al., 2017)
Durban	45.12	Average of one week in summer 2001	South Africa	(Moode et al., 2011)

Rawalpindi	55.74	Annual average for 2008	Pakistan	(Ahmad et al., 2011)
Dakar	59.9	From January 2008 to December 2009	Senegal	(Adon et al., 2010)
Al-ain	59.3	From 21 February 2005 to 20 February 2006	United Arab Emirates	(Salem et al., 2009)
Delhi	68.6	February and March 2004	India	(Behera et al., 2015)
Sfax	Between 37.6 and 112.8 *.	Fall 1996, Winter 1997, Spring and Summer 1998	Tunis	(Azri et al., 2008)
Durban	110.92	Average of one week in winter of 2001	South Africa	(Moode et al., 2011)

* Measurements made with an automatic monitoring network

Nitrogen dioxide is a photo-reactive product, whose content is controlled by the NO-NO_2-O_3 formation-destruction reaction cycle, under the effect of a radiation of wavelength lower than 400 nm schematized on figure 1, p.6. This cycle ensures a photostationary equilibrium between NO, NO_2 and O_3, which is disturbed in the presence of other pollutants such as the VOCs identified RH on figure 1, p. 6, by bringing benefit to the conversion of NO into NO_2. The OH radicals react with RH and give rise to alkyls that lead to the formation of peroxide radicals through a series of rapid reactions with O_2. These peroxides promote the rapid oxidation of NO to NO_2, resulting in an increase in nitrogen dioxide near the emission source and ozone at more distant locations (Azri et al., 2008).

These reactions explain the low concentrations detected in the peri-urban site, and the concentrations that exceed the limit values in the sites of road proximity and in the sites of industrial proximity juxtaposed to the roads, revealing the presence of a spatial variability, due to the impact of type of sites on the concentrations obtained of nitrogen dioxide. The revealed results were proved by a statistical analysis of variance ($p< 0.05$).

This significant spatial variability is represented on the map as a pollution gradient. It shows higher concentrations in the city center, near roads, and at locations and intersections with high traffic loads, gradually deteriorating towards the outskirts of the city.

Net spatial gradients of NO_2 are the common element in the different studies of nitrogen dioxide as a pollutant in urban environments (Lozano et al., 2011; Matte et al., 2013). These gradients are attributed to the location of pollution sources, type of measurement site, topography, and road infrastructure (Jerrett et al., 2005). For example, high NO_2 concentrations at high traffic sites can be attributed to traffic congestion and high NO emissions that rapidly oxidize to NO_2 near emission sources, nevertheless, some NO is oxidized before reaching the tailpipe (Short et al., 2006; Behera et al., 2015). In urban areas, some air pollutants can exhibit high spatial variability more than others (Parra et al., 2009; Smith et al., 2006), showing that nature plays a crucial role in tracing these gradients.

The photoreactive nature of nitrogen dioxide also explains the very similar average concentrations of the two campaigns, 75% of the days in the first campaign and 58% in the second are clear sky, representing a similar meteorological profile favorable to NO2 production in the absence of rain in both campaigns. In the presence of rain, nitrogen dioxide leaches from the atmosphere and is transformed into wet deposition as nitric acid according to the following reaction (Defra, 2012):

$$NO_2 + H_2O \longrightarrow HNO_3$$

The effects of meteorology on nitrogen dioxide concentration and dispersion have been revealed in many studies. Like our case study, some confirm the absence of significant difference in average NO_2 concentration between different study periods (Caballero et al., 2012; Lin et al., 2016). In contrast, other studies have revealed periodic variability attributed primarily to differences in the meteorological profiles of these periods (Wheeler et al., 2008; Moodle et al., 2011; Jang et al., 2016).

Nevertheless, the difference between the two periods of the present study is clearer on the dispersion of the pollutant in the urban fabric. During the summer campaign, the dispersion of NO_2 is characterized by a plume shape spread towards the South-East of the city of Meknes and influenced by a dominant wind of moderate intensity coming from the North-West and West, which ensures a good dispersion of the pollutant.

During the winter campaign, the dispersion of the pollutant is characterized by a localized pollution near the sources due to the multidirectional winds, and/or to a layer of inversion of the temperature at a few hundred meters of the ground. This phenomenon is present during this time of the year which slows down the diffusion of pollutants (Ait Bouh, 2012).

Chemical reactions and dilution processes are not the only ones responsible for the spatial and temporal variations of the studied concentrations, but also the short lifetime of NO_2. Therefore, the highest concentrations are found in areas close to the emission source, which decreases with distance, contributing to a heterogeneous pattern of its dispersion in space (Pekey et al., 2013).

III.3. Study of BTEX concentrations

The average concentrations of BTEX noted at the automotive proximity sites exceed those of the background stations (**Table 3**). This result is consistent with the one obtained in the city of Toulouse, where the calculation of toluene/benzene, m, p xylene/xylene and o-xylene/benzene ratios, which are markers of vehicular pollution, revealed that BTEX are, to a large extent, derived from road traffic.

The average concentrations of BTEX measured during the winter campaign are higher than those in the summer (**Table 3**).

In winter, benzene, toluene, p+m-xylene, o-xylene and ethylbenzene are characterized by average concentrations of 2.59, 23.415, 27.705, 15.3, 5.69 and 3.95 µg/m^3 respectively.

The average concentrations recorded, during the summer campaign, are respectively 1.01 for benzene, 14.21 for toluene, 6.5 for p+m-xylene, and 4.58 for O-xylene and 1.65 µg/m^3 for ethylbenzene.

Table 3. Variations saisonnières des teneurs des BTEX

Saison	Type de site	Benzène	Toluène	o-Xylène	p+m Xylène	Ethylbenzène
Hiver	**Fond**	1,01	15,07	3,09	9,61	2,2
	Proximité Automobile	3,01	31,76	8,29	20,99	5,7
	Moyenne	2,66	23,45	5,69	15,3	3,95
été	**Fond**	0,62	9.94	2,05	6.03	1,86
	Proximité Automobile	1,01	18.48	7,11	6,97	1,45
	Moyenne	0,83	14,21	4,58	6,5	1,65
Moyenne	**Fond**	1,40	12,50	2,57	7,82	2,03
	Proximité	2,04	25,12	7.70	13,98	3,57

	Automobile					

The increase in BTEX levels measured in winter can be explained by:
- The degradation and transformation processes of BTEX which are less important in winter;
- A greater stability of the lower layers of the atmosphere which limits the phenomena of dispersion (winter anticyclone).

The increase in temperature catalyzes the reaction of ozone formation from VOCs and the decrease in BTEX emissions from automobiles resulting from the drop in the number of vehicles circulating in the city during the summer period (vacations) (Meybeck et al. 2000).

For all exposed tubes toluene remains the main pollutant, followed by m, p- xylene, o-xylenes, ethylbenzene and benzene (Figure 9). A similar result was observed in the city of Tangier (Zouir, 2009).

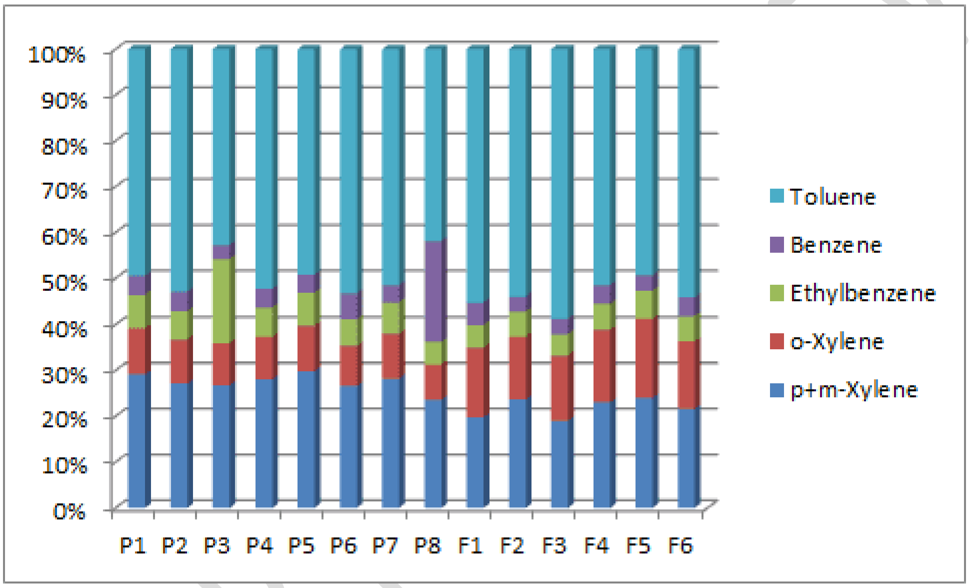


Figure 9. Comparison of the average concentration of each compound versus the sum of BTEX at each sampling site

Atmospheric concentrations of BTEX reach maximum values in the city center and tend to decrease towards its periphery (**Figure 10**). The highest levels of BTEX were recorded at site P7 corresponding to Daar Smane street with a peak of 90.76 µg/m3 of BTEX. The P5 (site located at the level of the avenue of the FAR) takes the relay with 82,445 µg/m^3. These results are quite logical because the structure of Daar Smane street is of canyon type (structure not favourable for a better dispersion of BTEX and favours their accumulation). Similar results for NO_2 and C_6H_6 were obtained at this street. The density of road traffic at P5 is at the origin of this increase. A slight increase in the levels of xylene isomers and ethylbenzene was noted at the background sites near the industrial areas (F6, F2 and F5). These results are in agreement with those observed in the city of Toulouse where the lowest ratios of toluene/benzene concentrations were noted at the background sites most isolated from the Toulouse agglomeration while the maximum values occurred at the sites most frequented by the means of transportation of the city. In addition, BTEX concentrations were highest in the center of Toulouse and tended to decrease towards the outskirts (Meybeck et al., 2000). In Tangier, the highest BTEX concentrations were recorded at the municipal landfill site with a total BTEX sum of 166.7 µg/m^3 followed by an industrial proximity site with a value of 111.7 µg/m^3. Toluene was the dominant pollutant, followed by m-p xylene, benzene and o-Xylene with a slight increase in the levels of xylene and ethylbenzene isomers at the automotive proximity sites (Zouir, 2009).

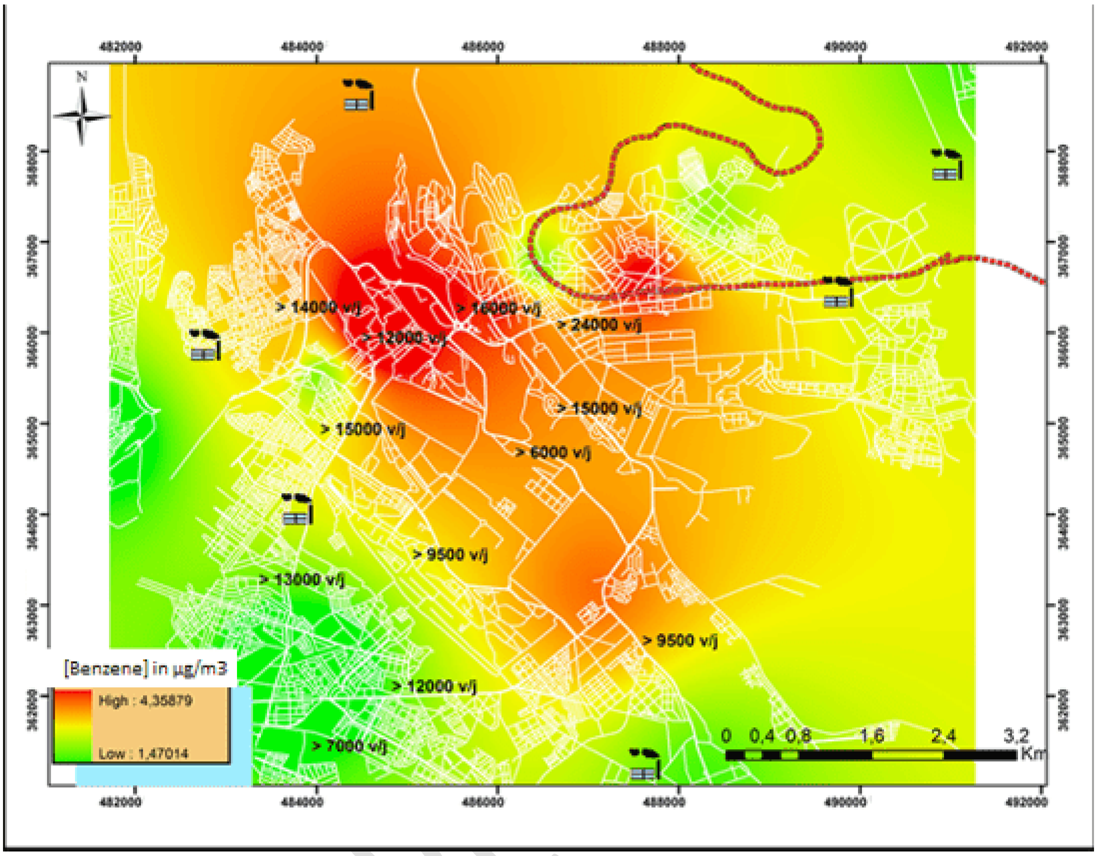


Figure 10(a). Spatial variations of C_6H_6 concentrations

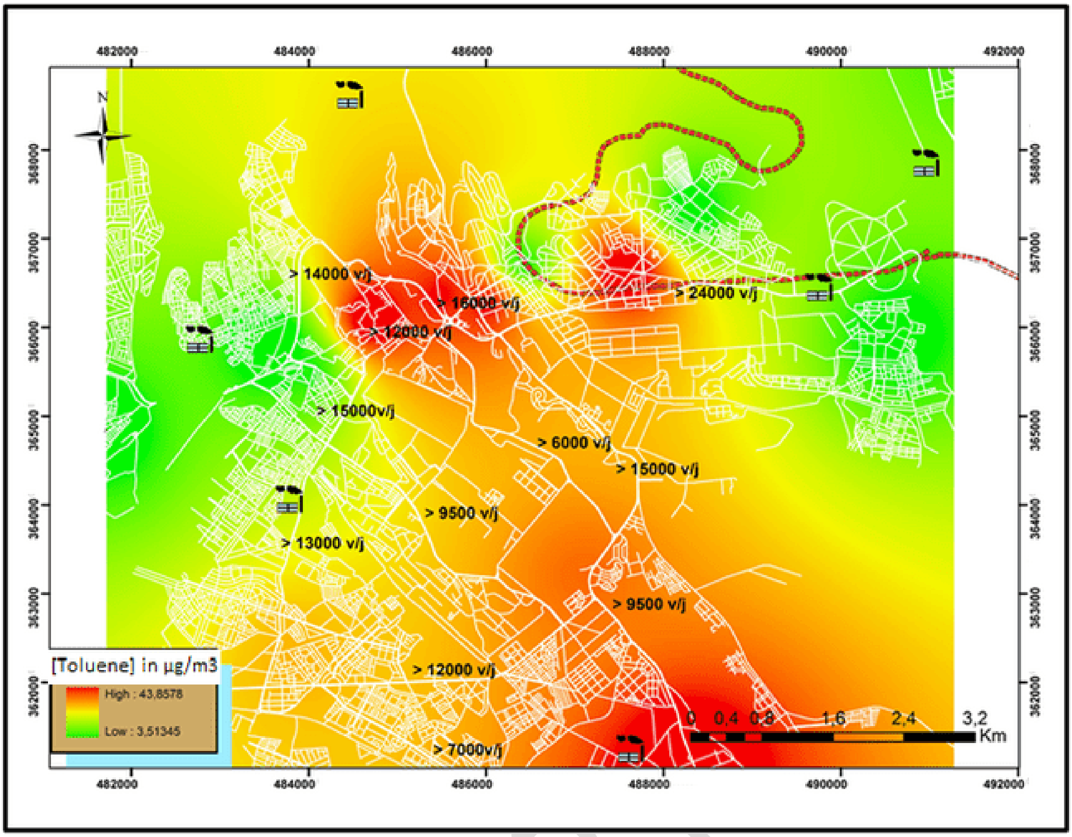


Figure 10$_{(b)}$. Spatial variations in toluene concentrations

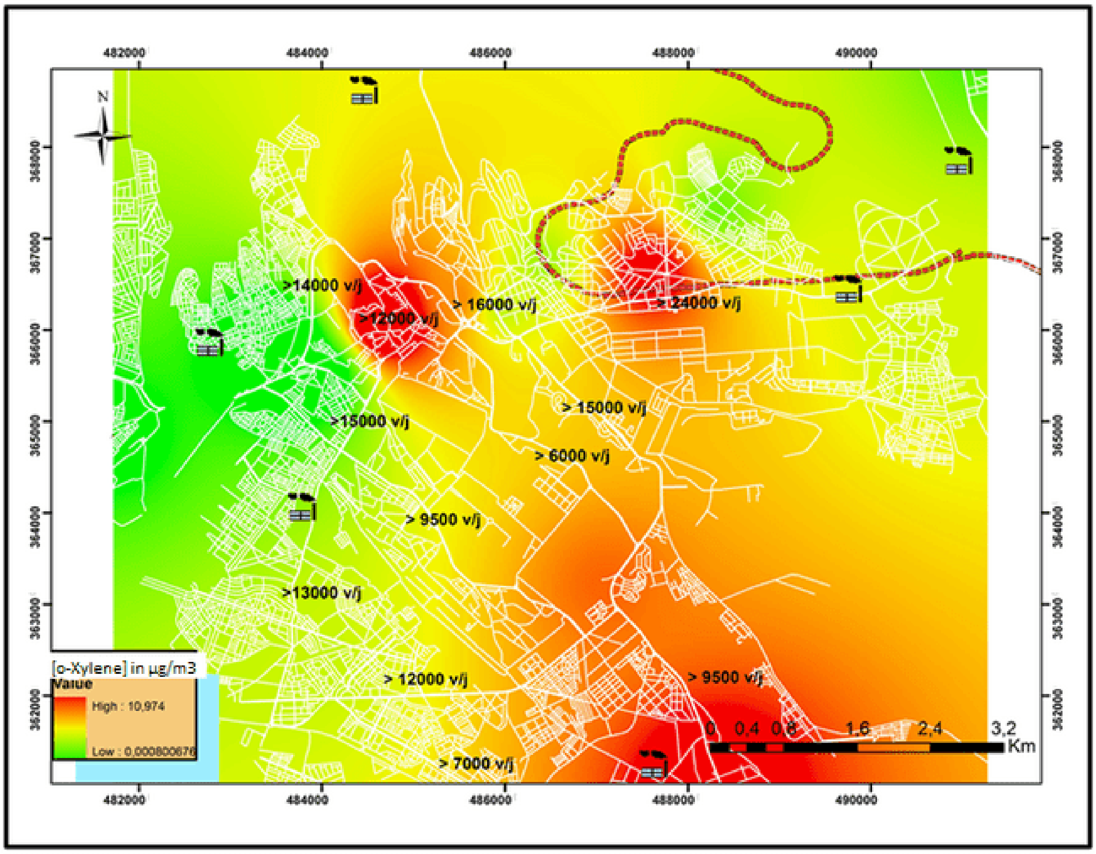


Figure 10$_{(c)}$. Spatial variations in o-Xylene concentrations

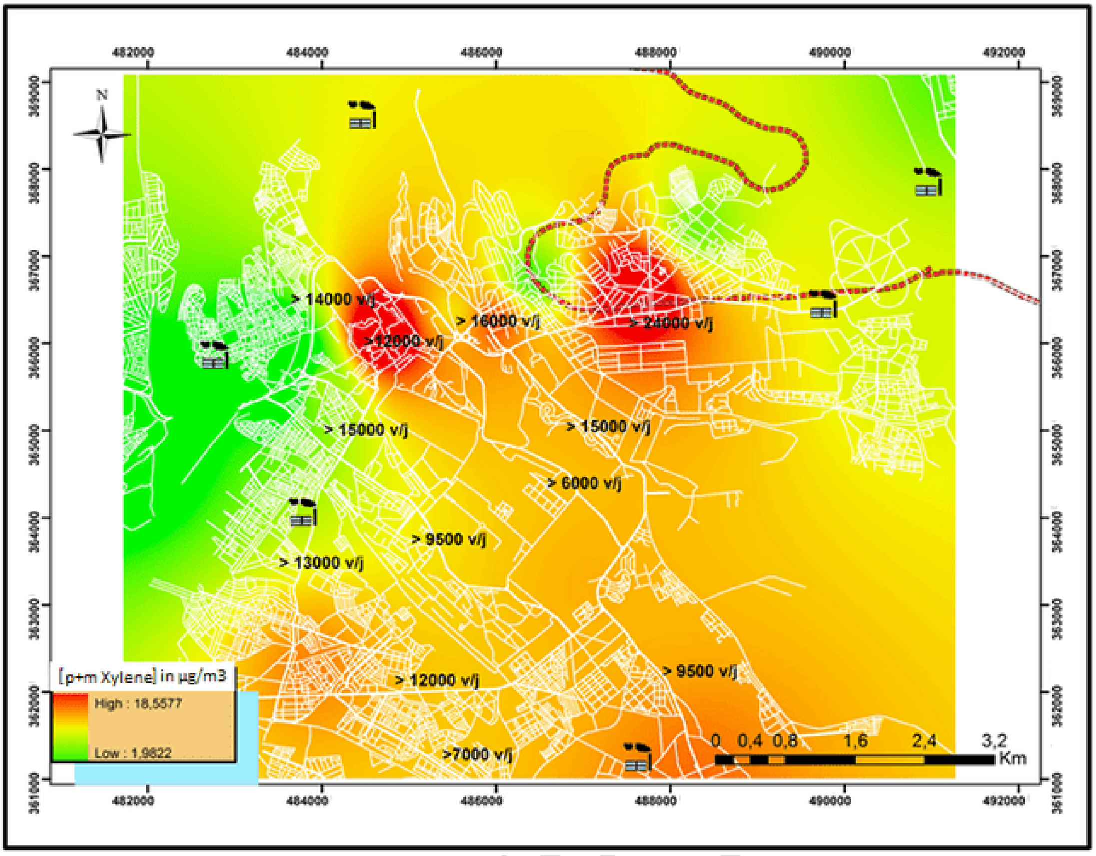


Figure 10$_{(d)}$. Spatial variations in p+m Xylene concentrations

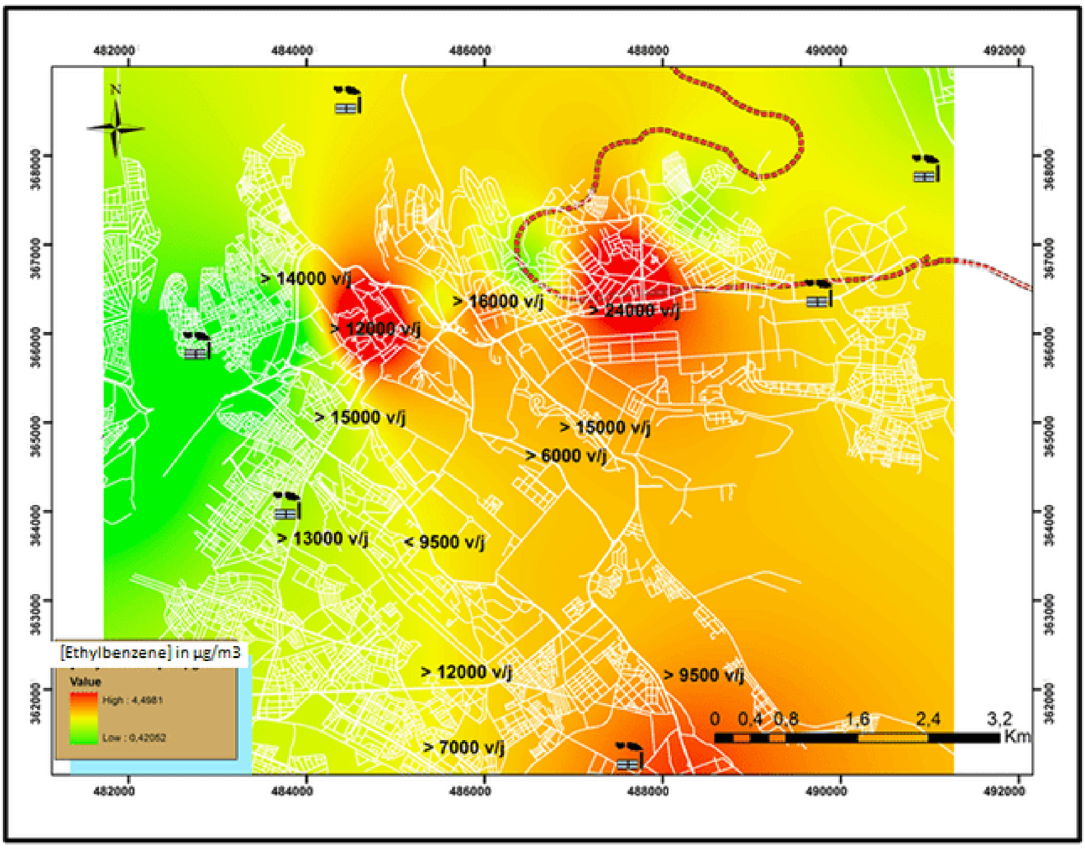


Figure 10$_{(e)}$. Spatial variations in ethylbenzene concentrations

For C_6H_6, 10 µg/m^3 is the Moroccan threshold, respected on all the sites. At the level of 4 sites among 14, there is an exceedance of the quality objective fixed by the European Union (2 µg/m^3). All background sites show C_6H_6 levels below 2 µg/m^3, the limit value tolerated in Europe (Directive 2008/50/EC).

Toluene is not subject to regulation. However, the World Health Organization (WHO) recommends not exceeding 260 µg/m^3 on average over 7 days. The values of toluene measured in the city of Meknes are very largely lower than this threshold. Even though toluene reached 18.81 µg/m^3, this is only 1/15 of the recommended value for health.

The WHO recommends 4800 µg/m^3 over 24 h of xylene in ambient air (WHO, 2000). The results of our monthly measurement campaigns do not allow a real comparison with this standard. For o-xylene and p+m xylene none of the measured values exceeded this value.

For ethylbenzene, the WHO recommends an annual value not to be exceeded of 22 000 µg/m^3 of ethylbenzene in ambient air (WHO, 2000). The average ethylbenzene level recorded during the study period is 2.80 µg/m^3, which is well below this threshold.

IV. Conclusion

The present study focuses on the monitoring and evaluation of air quality in the city of Meknes, based on two tracers of vehicular proximity pollution (VOC and NO_2) emanating from road traffic. In the absence of a telemetric station and a mobile laboratory within the city, the use of passive diffusion tubes, which are analyzed offline in the laboratory after exposure, was the most effective solution. The low cost, ease of implementation of the technique, and the possibility of covering a large geographic area with a large number of samplers were all arguments in favor of adopting this approach.
The coupling of the results of the measurement campaigns and the counting sessions under GIS made it possible to determine the zones most affected by the automobile pollution and to carry out a cartography with a high spatial resolution of the prospected pollutants.

The deployment of passive sensors at 14 measurement sites revealed the existence of a spatio-temporal variability of the pollutants surveyed. This variability is due to the climatic conditions that prevailed during the measurement campaigns, in particular wind and precipitation, to the photochemical nature of the said markers, to the proximity to the emission sources, to the type of measurement site and to the topography and road infrastructure.

The results of our study show that the air concentrations of NO_2 and BTEX reach maximum values in the city center and decrease towards its periphery. NO_2 and C_6H_6 levels in the winter campaign exceed those in the summer.

A slight increase in the levels of xylene isomers and ethylbenzene was noted at the background sites near the industrial areas.

The exceedances of the admissible thresholds of the pollutants noted at the level of some sites of sampling, pushes us to study the relation between the exposure to these tracers and the development and the exacerbation of the respiratory and cardiovascular pathologies in the city of Meknès.

The developed approach could be used as a decision-making tool for the competent authorities in this field and adapted for the monitoring of other types of pollutants (pesticides, tracers generated by industrial units, dioxins furans...).

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