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Assessing the impact of petrol stations on their immediate surroundings

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ABSTRACT

This paper describes a novel methodology for evaluating the extent to which petrol stations affect their surroundings. The method is based on the fact that the ratio of the concentrations of aliphatic and aromatic hydrocarbon pollutants in the air of the petrol stations and their surroundings (basically determined by vapor emissions from unburned gasoline) differs from the ratio found in urban air, which is mainly influenced by traffic emissions. Bearing this in mind, the spatial limit of influence of petrol stations in any direction would be the first point, moving away from the station, where the ratio becomes equal to the urban background ratio. Application of the methodology involves multipoint measuring campaigns of the air at the studied petrol station and built-up area in general and processing the data with software capable of providing isoconcentration contours. The procedure should help local authorities in terms of land management, so that a "belt" can be established around petrol stations where housing or vulnerable populations and activities such as those in schools, hospitals and community centers should be restricted.

1. Introduction

Volatile organic compounds (VOCs) belong to a very heterogeneous group of chemicals characterized by their relatively high vapor pressures. Exposure to these compounds can bring about a variety of adverse health effects, including asthma, headaches, mucosal symptoms (Steinemann, 2008) and, in some cases (e.g. benzene), an increased risk of cancer (Ott et al., 1978; Lynge et al., 1997). In Europe, the only legally regulated VOC as regards air quality is benzene (Directives 2000/69/EC and 2008/50/EC). The indirect effects of VOCs on health are related to their role as precursors of ozone and other photochemical pollutants.

In urban areas, the main source of VOCs is usually traffic. Additional sources are petrol stations and small-scale industries (paint, adhesives, etc.) which use organic compounds as solvents. Petrol stations as emission sources of VOCs have been the subject of considerable study, a particular interest being those related to the design and evaluation of control systems in an attempt to diminish emissions (Uren, 1997; Ohlrogge et al., 2000), those related to their effects on workers (Brugnone et al., 1997; Periago and Prado, 2005) and environmental studies to evaluate associated air quality (Gonzalez-Flesca et al., 2002; Palmgren et al., 2001; Srivastava et al., 2005; Fernández-Villarrenaga et al., 2005).

Although Spanish petrol stations were traditionally located in largely uninhabited areas, the continuous urban growth experienced by Spain in recent years has resulted in many petrol stations being built within urban areas surrounded by buildings. This situation has led to controversy between the citizens whose houses are close to the petrol stations and the authorities responsible for land management. However, few methods permit us to establish the impact of these petrol stations on the surrounding environment, the work carried out by Karakitsios et al. (2007) in this respect being a notable exception. These authors concluded that petrol stations make a significant contribution to ambient benzene concentration in their vicinity. They calculated this contribution by comparing the background concentration, measured by passive samplers, to the contribution of the roads, estimated with both COPERT and CALINE4 models. However, the use of dispersion models is always affected by considerable uncertainty.

In this paper, an alternative methodology is described for use as a land management tool by responsible authorities. The methodology is based on the fact, which we demonstrate in this study, that the ratio of the concentrations of aliphatic and aromatic hydrocarbon pollutants in the air in the vicinity of petrol stations differs from that found in "normal" urban air. The point of using concentration ratios is that they eliminate from the analysis

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a number of local factors (e.g. wind speed) which affect absolute concentrations and which are impossible to take into account (Gelecsér et al., 1997).

2. Experimental section

2.1. Location

The study was carried out in the city of Murcia, located in the south-east of the Iberian Peninsula, which is characterized by very low annual rainfall (around 375 l/m^2), annual average temperature of 18 °C and extreme temperatures of 40–45 °C in summer and −2–0 °C in winter.

The air of the urban surface of the city, around 465 hm^2, was the subject of the study with regard to VOC concentrations together with that of a petrol station known as “La Fica”. This petrol station is located on the east side of the city and is Stage I implemented, that is, it uses vapor recovery devices to return the VOC-saturated volume of air displaced from the storage tank being filled to the tank being emptied (Gonzalez-Flesca et al., 2002). It has four pumps for gasoline and diesel and one for motorbikes. The N and S approaches to the petrol station are unhindered due to the presence of a 30 m-wide road. A single low building is situated east of the station and has no more construction behind it. On the other side of the road (west of the station) stand several 3-storey blocks. This explains why this particular petrol station was chosen: the west area is inside the built-up area of the city, whereas the east is practically undeveloped (Fig. 1).

2.2. Sampling

As the traffic conditions in the city are not the same throughout the week, working days (from Monday 08.00 h to Friday 22.00 h) were distinguished from weekends (Friday 22.00 h to Monday 08.00 h). This study was carried out in two different Monday-to-Friday campaigns (from 26/05/08 to 30/05/08, and from 16/06/08 to 20/06/08) with a total exposure time in each campaign of 110 h.

One-hundred-and-five Radiello® passive samplers (RAD130 Cartridge Adsorbent and RAD120 Diffusive Bodie, Sigma Aldrich, Inc., St. Louis, Missouri (US)) were used to measure VOCs in the urban area. Information about sampler deployment throughout the city and quality control can be found in González Ferradás et al., 2010. The present study forms part of a larger one aimed at evaluating different aspects related to the influence of VOCs in Murcia (e.g., the best locations for siting air monitoring stations to measure benzene (González Ferradás et al., 2010)), which explains the great number of measuring points chosen. Therefore, fewer samplers can be used to reduce cost and time in future studies.

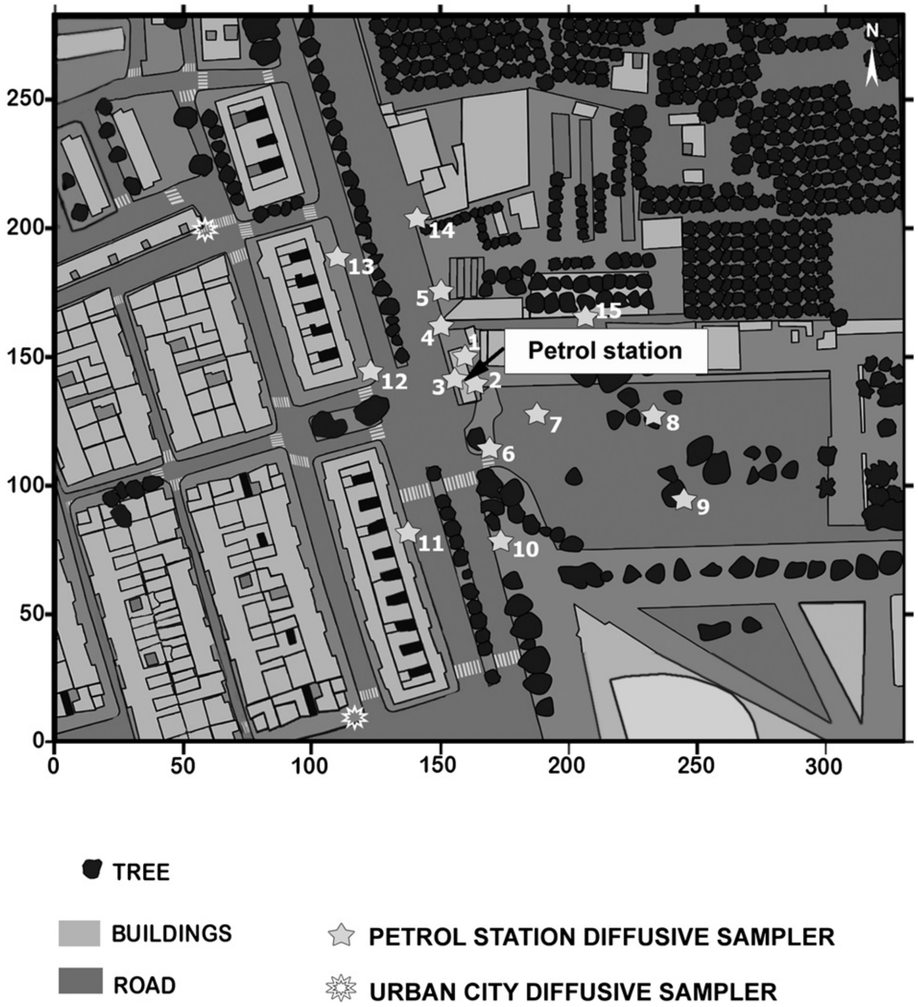

Fig. 1. Location of the passive samplers inside and around the petrol station “La Fica”. (2 passive samplers from the 105 passive samplers used in the urban sampling are also shown). (Distances in meters).

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With regard to the measurements in the petrol station, fifteen Radiello® passive samplers (apart from the 105 passive samplers used in the city area) were set up in the vicinity, three of them within the petrol station itself, close to the pumps, and the rest were set up to achieve a maximum radial layout of 100 m. The final location was determined by the availability of fastening places. The distribution of the passive samplers around the petrol station can be seen in Fig. 1.

Temperature, wind speed and wind direction were measured hourly during the two sampling campaigns at an air quality monitoring station located near the petrol station. Temperature was measured with a PT100 Hygroclip S3 sensor (Rotronic Instruments Ltd, Crawley, West Sussex (UK)) and wind speed and direction with a Young 03002 anemometer (R.M. Young Company, Traverse City, Michigan (US)).

2.3. Analysis and concentration calculations

Benzene, n-hexane and cyclohexane were the VOCs chosen for analysis during the two different Monday-to-Friday campaigns as these compounds are representative of aromatics, paraffins and cyclic paraffin hydrocarbons, respectively. Hydrocarbon desorption from the active charcoal in the passive samplers was carried out using carbon disulfide. Further details regarding solvent desorption can be found in González Ferradás et al., 2010. A gas chromatograph (HP 6890, Agilent Technologies, Inc., Santa Clara, California (US)) equipped with a flame ionization detector and a semi-capillary DB-5 column (J&W Scientific; 50 m long and 0.32 mm i.d.; film thickness 1 μm; Agilent Technologies, Inc., Santa Clara, California (US)) was used for quantitation. The oven temperature was programmed from 50 °C (0 min) at 10 °C/min to 80 °C (10 min) and at 10 °C/min to 200 °C (10 min). The injector and detector temperatures were 250 and 350 °C, respectively, and the flow rate of the carrier gas (N_2) was 40 ml/min. Operational procedures (sampler set up and recovery, desorption and analysis of VOCs) were developed in order to assure comparable results.

Once the mass of each hydrocarbon was known, m_i, the concentration, C_i, was calculated using the following equation: $C_i = m_i/(SR_i \cdot t)$, where SR_i is the sampling rate of the Radiello diffusive sampler for the i-component (8.0×10^{-5} m^3/min for benzene, 6.6×10^{-5} m^3/min for n-hexane and 5.4×10^{-5} m^3/min for cyclohexane), obtained from Fondazione Salvatore Maugeri (2008), and t is the exposure time.

3. Data treatment

First of all, linear correlations between the different measured hydrocarbons in each campaign were noted. Unusual observations were detected with the help of Minitab 13.0 (Minitab Inc., State College, Pennsylvania (US)). Minitab is a statistical software package which includes, among others, modules for general univariate statistics, multivariate statistics, time series analysis and non-parametric statistics. We used the regression option to obtain regression equations, the Pearson coefficients, and unusual observations. We could distinguish two types of atypical points; points that were atypical for all of the pollutants (a behaviour that was observed in only one campaign) were omitted because they were probably due to analytical mistakes, whereas points that were atypical for just one or some of the pollutants or which showed atypical behaviour in more than one campaign were not removed because a source other than traffic could have existed. The percentage of points removed was around 5% in the urban sampling whereas no data from the vicinity of the petrol station was eliminated. After that, a statistical analysis with SPSS 15.0 (SPSS Inc., Chicago, Illinois (US)) was carried out to determine the frequency distributions of pollutant concentrations. These concentrations showed a lognormal behaviour whereas pollutant ratios followed normal distributions for a 95% confidence level.

From the comparison of the measurements in the city and in the petrol station, a methodology for evaluating the influence of petrol stations on their vicinity was determined. The methodology was based on the fact that the ratio of certain VOCs differs in the general ambient air of the city from that in the air of petrol stations. For each pair of hydrocarbon concentration ratios, the following parameters were calculated: URB(N), defined as the average value in the urban area of the N ratios (∼105) obtained for each pair of contaminants, and PS(3), defined as the average value of the 3 ratios obtained for each pair of contaminants within the petrol station. For instance, URB(N) and PS(3) for the quotient of benzene and n-hexane were calculated with equations (1) and (2), respectively.

$$(Benz/nHex)_{URB(N)} = \frac{\sum_{i=1}^{i=N}(Benz/nHex)_i}{N} \tag{1}$$

$$(Benz/nHex)_{PS(3)} = \frac{\sum_{i=1}^{i=3}(Benz/nHex)_i}{3} \tag{2}$$

For each average ratio of pollutants, the relative difference between that found in the urban area and that found in the petrol station, D, was calculated, taking as the reference the lower value of each pair which coincided with that found in the urban area, equation (3).

$$D = \frac{\left[\left((Benz/nHex)_{PS(3)} - (Benz/nHex)_{URB(N)}\right)\right]}{(Benz/nHex)_{URB(N)}} \times 100 \tag{3}$$

The next step was to draw the isoconcentration curves of the two hydrocarbons with the highest relative difference in ratios, radiating from the petrol station by means of suitable software, in this case SURFER 8.0 (Golden Software Inc., Golden, Colorado (US)), and using as inputs the concentration data of the passive samplers located inside (3 points) and around the petrol station (12 points). It was also necessary to include some background concentrations taken from the urban monitors. These graphs permit us to obtain the concentration values of each selected pollutant at different distances from the petrol station in each primary direction (N, S, E and W). The extent of the influence of the petrol station is taken as the first point in each direction moving away from the station where the ratio becomes equal to the urban background ratio.

4. Results and discussion

4.1. Meteorological data

The average temperature during each campaign was 19.7 and 23 °C, respectively. With regard to wind speed, the average value for the first campaign was 1.8 m/s, whereas in the second campaign an average value of 1.5 m/s was found. In both campaigns, similar wind behaviour was observed. The main wind direction was NE-ENE during 30% and 40% of the time of the first and second campaign, respectively. When wind speed was <3 m/s no prevalent direction was shown in any case.

4.2. Concentration data in the city

Statistical analysis of the distribution of the concentration data by Kolmogorov–Smirnov and Shapiro–Wilks tests showed that spatial concentrations in the area of the petrol station and the

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urban city could be fitted to lognormal distributions for a significance level of 0.05. Ratios of concentrations also passed the normality tests with the same level of significance. The average values of the concentration logarithms for n-hexane, benzene and cyclohexane (and their standard deviations in parenthesis) were 0.581 (0.179), 0.519 (0.195) and 0.491 (0.193) for the first sampling campaign and 0.547 (0.184), 0.493 (0.203) and 0.554 (0.210) for the second one, respectively. Analysis of variance showed that there were no significant statistical differences in the results obtained in the urban area from the first to the second campaign at a 95% confidence level.

A significant result found was the very good linear correlation ($R^2 > 0.9$) between the different pairs of pollutants in the city. Taking benzene as a reference, the Pearson correlation coefficients obtained for n-hexane and cyclohexane versus benzene were 0.946 and 0.954, respectively, for the first campaign, and 0.970 and 0.928, for the second campaign, after eliminating unusual observations (around 5%) by means of Minitab 13.0 as explained in Section 3. In Fig. 2, the relationship between n-hexane and benzene in the urban area (solid lines) can be observed for the two campaigns. Similar graphs were obtained for cyclohexane and benzene. From the examination of Fig. 2, the following aspects can be concluded:

1. The concentrations of benzene and n-hexane in Murcia (far away from other sources such as petrol stations) are mainly from traffic emissions as there is a clear linear trend, shown in Fig. 2 by means of solid lines. Any point located outside this general trend is indicative of an emission source other than traffic, which is the case of the concentrations found inside the petrol station, linked by dotted lines.
2. Uncertainty calculation of the slopes of the solid lines in Fig. 2 showed that the slight differences found in them are inside the variability of the slopes for a confidence interval of 95%. Nevertheless, it could happen that there were significant differences from one campaign to another as petrol and diesel composition is not constant over time. Meteorology, traffic density and architectural elements do not influence the ratio but they can proportionally affect the concentration changes of both compounds in air. In this sense, the more traffic there is, the higher the concentrations of n-hexane and benzene will be, however, the ratio, as seen in Fig. 2, remains constant.
3. Taking into account the two previous points, the air in the city of Murcia and in other similar cities in relation to emission patterns, i.e. with no important emission sources other than traffic, can be defined by means of straight lines that link the concentrations of two organic compounds emitted by traffic.

4.3. *Calculating the impact of petrol stations*

N-hexane, benzene and cyclohexane concentrations measured in and around the petrol stations are shown in Table 1. F1–F15 are the codes of the diffusive samplers. F1–F3 were located inside the petrol station and the rest of them outside as explained in Section 2.2. The higher concentrations obtained in F1–F3 in the second campaign are probably due to the higher ambient temperature registered in the latter campaign. They could also be due to a greater activity in the petrol station during the second campaign but, unfortunately, we have no information on the petrol and diesel dispensed during the sampling periods.

As can be seen in Table 1, the lowest concentrations are found in E direction. There are several reasons for this. The first is that no other anthropogenic emissions are present in this direction whereas in all other directions the influence of the road is clearer. The presence of buildings also hinders pollutant dispersion favouring higher concentrations between them and the petrol station. Finally, the wind blowing from NE-ENE favours a higher impact of pollutants from the petrol station in WSW-SW directions.

It can be checked from Fig. 2 that the concentrations of n-hexane and benzene measured at the petrol station (3 sites) do not follow the same behavior (dashed lines) as the points in the city located a distance away. In Fig. 2b, point F3 is not shown because of its high concentrations, but it was taken into account in the linear regression. These high concentrations were probably due to a greater proximity of the corresponding sampler to an emission source (tanks, pumps) together with a possibly higher amount of petrol/diesel sold in that campaign and higher temperatures.

From the above, it can be concluded that the air at the petrol station does not have the same proportion of VOCs as the general city air. Presumably, there is an area close to the petrol station where the VOC concentrations are influenced by both the petrol station and traffic. As we go further from this area, the VOC concentrations are closer to that found in the general air. The closest places to the petrol station, in each direction, where the ratio becomes equal to the background one delimit the area which is influenced by the petrol station.

Following the methodology described in Section 3, the best pair of pollutants was n-hexane and benzene in the two campaigns, as their ratio had the highest relative difference between the petrol station and the rest of the city. The differences for each campaign are shown in Table 2.

In both cases, the highest deviations were obtained when comparing n-hexane (non-aromatic hydrocarbon) with benzene (aromatic), while aliphatic–aliphatic comparisons, and presumably, aromatic–aromatic comparisons, showed lower deviations.

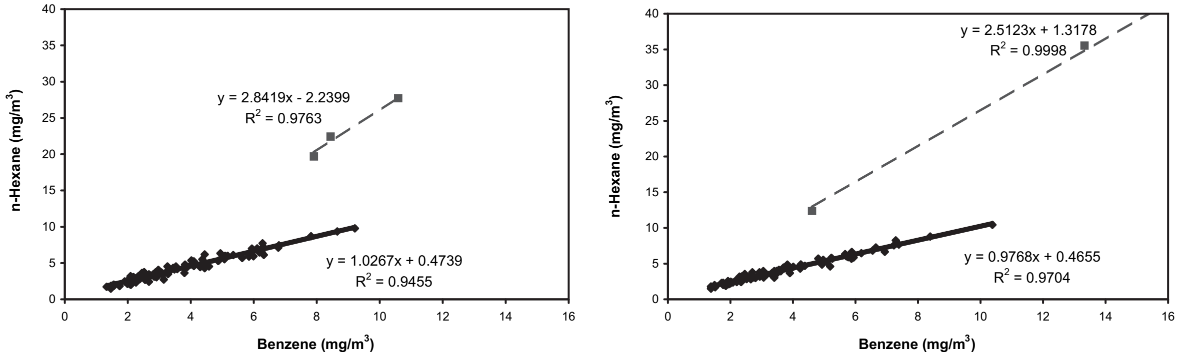

Fig. 2. N-hexane vs benzene concentrations in the city (solid lines) and in the petrol station (dashed lines). (a) First sampling campaign; (b) second sampling campaign.

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Table 1
N-hexane, benzene and cyclohexane concentrations (μg/m^3) measured with the 15 passive samplers in and around the petrol station. F1–F3 were located inside the petrol station. F4–F15 were deployed to achieve a maximum radial layout of 100 m. Their orientation respect to the petrol station is also indicated.

Passive sampler code	Orientation	1st Campaign			2nd Campaign		
		n-hexane	benzene	cyclohexane	n-hexane	benzene	cyclohexane
1	Centre	19.7	7.91	14.9	12.4	4.61	11.1
2	Centre	22.4	8.44	15.9	35.5	13.3	31.9
3	Centre	27.7	10.6	21.2	94.8	37.3	100.2
4	N	9.63	4.69	6.51	19.6	8.60	21.9
5	N	7.92	3.84	5.76	8.69	4.14	8.56
6	S	2.03	1.72	1.66	2.24	1.81	2.19
7	SE	2.21	1.88	1.88	2.21	1.64	2.83
8	ESE	1.77	1.46	1.40	2.08	1.27	1.86
9	SE	1.53	1.38	0.98	1.69	1.36	1.82
10	S	2.29	2.18	2.22	2.26	1.99	3.08
11	SSW	5.45	4.82	4.49	5.84	5.33	6.48
12	W	4.96	4.84	4.44	6.91	5.59	6.43
13	NNW	3.71	3.65	3.02	6.00	4.61	6.29
14	N	5.32	4.19	3.55	6.05	4.22	5.99
15	ENE	1.88	1.36	1.39	1.36	1.16	2.34

This can be attributed to the thermal stability of these compounds during the combustion process. Aliphatic compounds are more easily burned than aromatic compounds, which meant that aliphatic/aromatic concentration ratios were significantly lower in the city air than in the petrol station, where the air was mainly characterized by unburned gasoline vapours. These findings were corroborated by their autoignition temperatures, being 498, 260 and 240 °C for benzene, cyclohexane and n-hexane, respectively (INSHT, 2003).

Selected pollutant (n-hexane and benzene) isoconcentration curves for each campaign were obtained by means of SURFER 8.0, selecting a multiquadric interpolation (Figs. 3 and 4, respectively), using as inputs the concentration data of the 15 passive samplers located inside and in the vicinity of the petrol station (Fig. 1). Only one point representative of the petrol station was used, obtained from the average concentration of the three passive samplers located inside the station. It was also important to use as input several concentration values typical of the urban air to set the background concentrations of the city relatively close to the petrol station. Finally, using the grid node editor tool of the above-mentioned software, concentrations of the selected compounds were obtained at different distances from the centre of the petrol station, in each geographical direction, so that the ratio of n-hexane/benzene could be calculated at different distances from the petrol station.

The relationship between the n-hexane/benzene ratio and the distance from the petrol station for each campaign is represented in Fig. 5. The horizontal solid line represents the average n-hexane/benzene ratio obtained in the city during each campaign. The 95% confidence interval of this average is shown as dashed lines. The distances in each direction at which the ratios start having similar values to that found in ambient air, that is, when the ratios are inside the 95% confidence interval of the typical urban air ratios, delimit the area influenced by the petrol station.

As shown in Fig. 5, the further we go from the petrol station the concentration ratio gets closer to that found in city air, which is mainly characterized by traffic emissions. From Fig. 5, the impact of the petrol station in each direction can be calculated as the point where the n-hexane/benzene ratio falls within the region of the 95% confidence interval of the urban average ratio. Table 3, summarizes the distances obtained in both campaigns.

A comparison of the distances obtained in both campaigns shows a higher influence of the petrol station in the second campaign probably due to the higher temperatures registered in the latter campaign. Although emissions from vehicle traffic have an evaporative loss contribution (around 10%), motor vehicle exhaust represents the main source of the total VOC on-road emission rate (around 90%) (Doskey et al., 1999; Olson et al., 2009). High temperatures favour evaporation losses in both locations – urban city and petrol station – but they are likely to be more noticeable in the air around the petrol station as they represent the main emission source. Anyway, the distances were similar in both campaigns and, in all cases, below or equal to 75 m.

It is also worth noting that, in both sampling campaigns, the most affected directions were N and E, whereas the less affected direction was W. This can be explained by the following: The petrol station is located in the limit of the urban perimeter. An NNW-SSE heavy traffic road splits the area into two: one with a considerably high level of traffic (West half) and one largely uninhabited (East half). The higher the traffic near the petrol station, the higher the concentrations of pollutants coming from it and, consequently, the ratio of any pair of aromatic and paraffinic hydrocarbons will be more representative of traffic than of the petrol station, that is, the emissions coming from the petrol station will be more and more insignificant as other emission sources become more important. Also, the presence of buildings in the W direction acts as a barrier against the pollution coming from the petrol station and the main road. This implies that the distance

Table 2
Average concentration ratios in the urban area, URB (N) and inside the petrol station “La Fica”, PS(3), and relative differences, *D*, for the two sampling campaigns.

	1st Campaign			2nd Campaign		
	n-Hex/Benz	n-Hex/Cyclohex	Cyclohex/Benz	n-Hex/Benz	n-Hex/Cyclohex	Cyclohex/Benz
URB(N)	1.16	1.23	0.95	1.10	0.96	1.14
PS(3)	2.59	1.34	1.93	2.63	1.06	2.49
D, %	123	8.94	103	139	10.4	118

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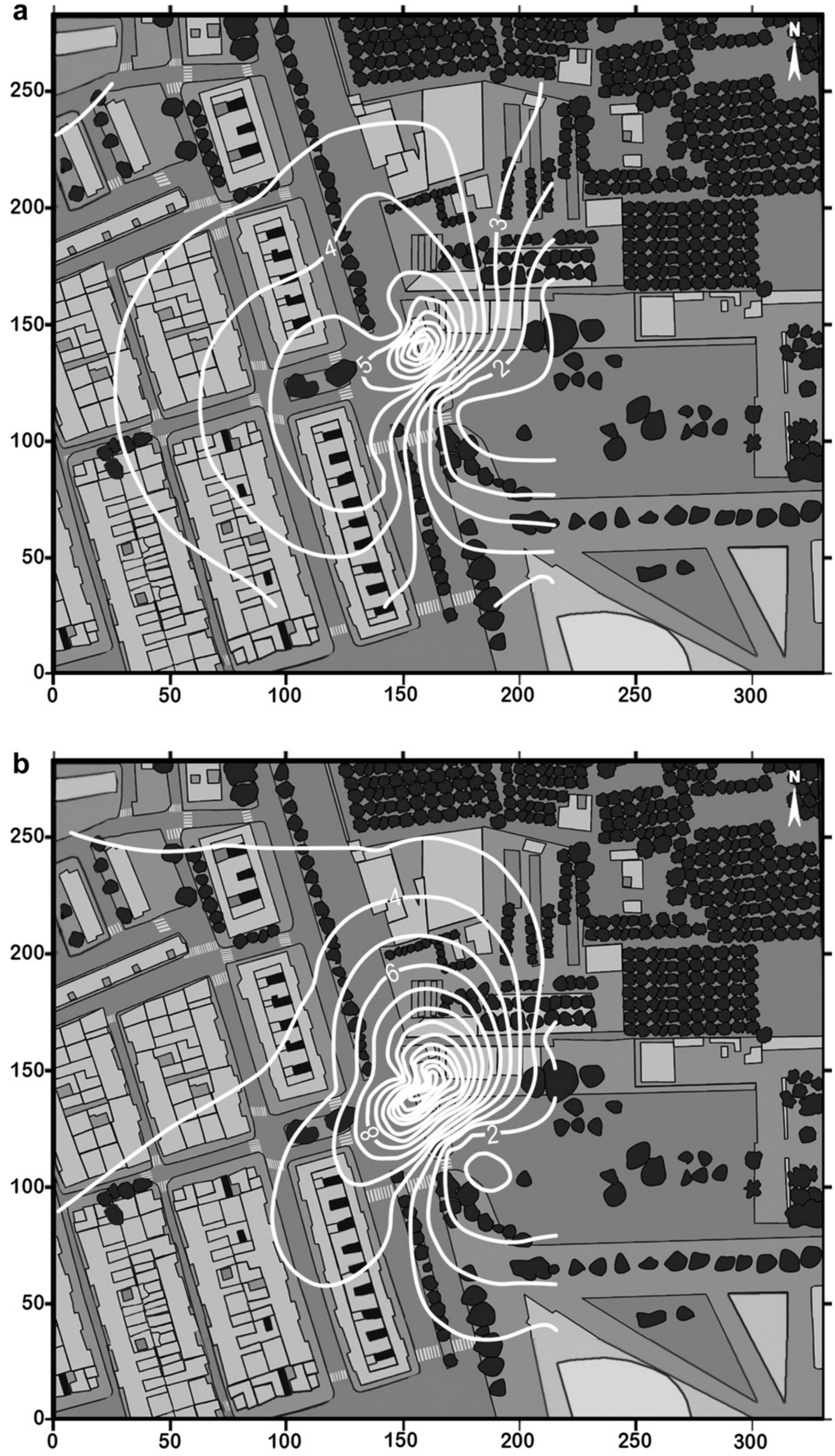

Fig. 3. Isoconcentration curves (μg/m^3) of (a) benzene and (b) n-hexane in the first sampling campaign. (Distances of the axis in meters).

between the buildings and the petrol station is likely to be the maximum influence distance in this direction as, behind them, concentrations are very close to the city background ones. These results highlight the fact that the influence of petrol stations depends on the characteristics of the surrounding area (mainly traffic density), thus we can establish that their spatial influences are relative. However, the special location of the studied petrol station, the time of year the samples were taken and the results

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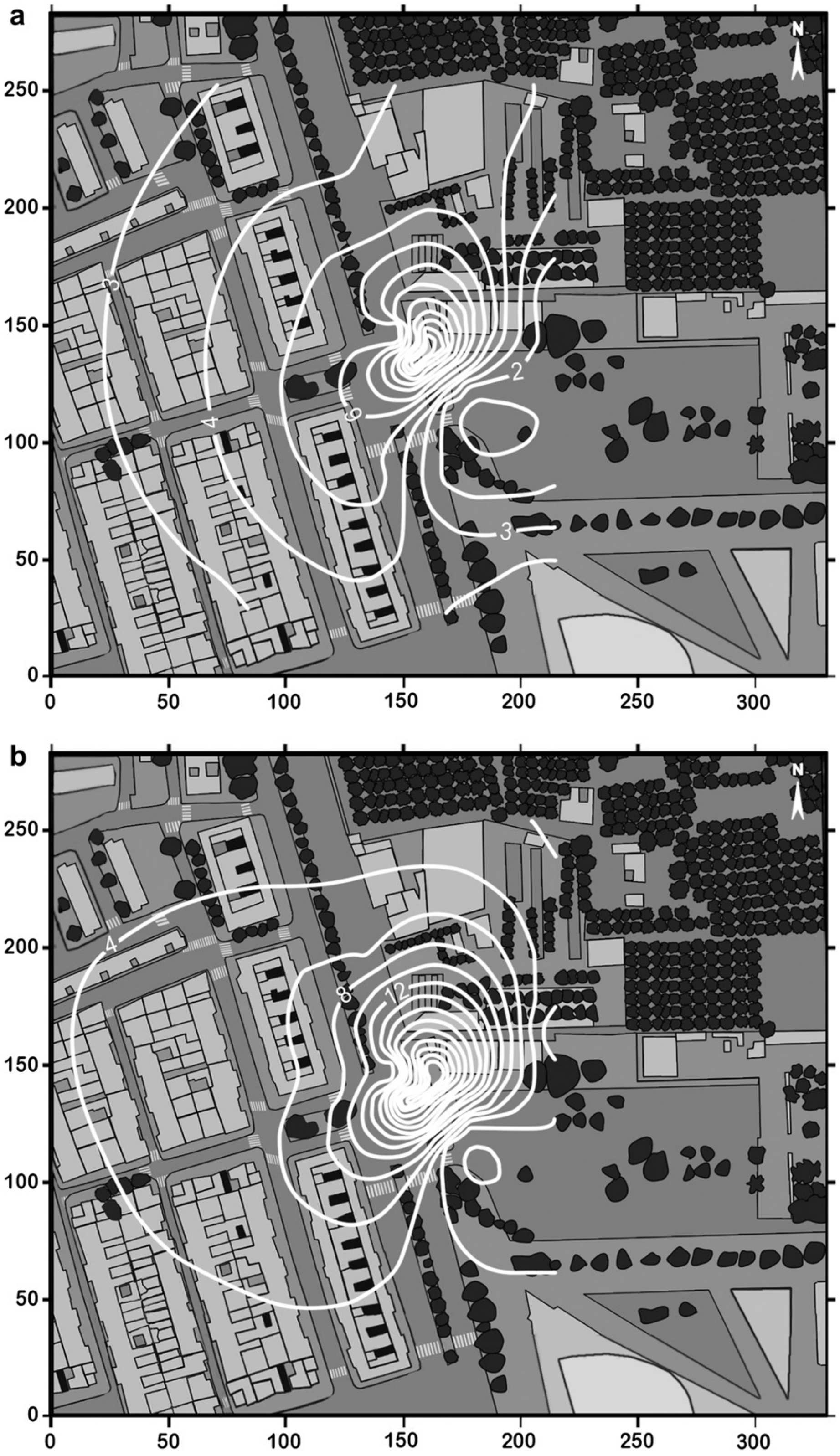

Fig. 4. Isoconcentration curves ($\mu g/m^3$) of (a) benzene and (b) n-hexane in the second sampling campaign. (Distances of the axis in meters).

obtained suggest that the impact of small and medium sized petrol stations will be, approximately, within a 100 m radius. Nevertheless, it would be advisable to apply this methodology in any case in order to establish with more precision the impact of petrol stations on their environment in order to carry out more efficient land management. It would be also interesting to carry out indoor samplings in the buildings close to the petrol station to estimate resident exposure.

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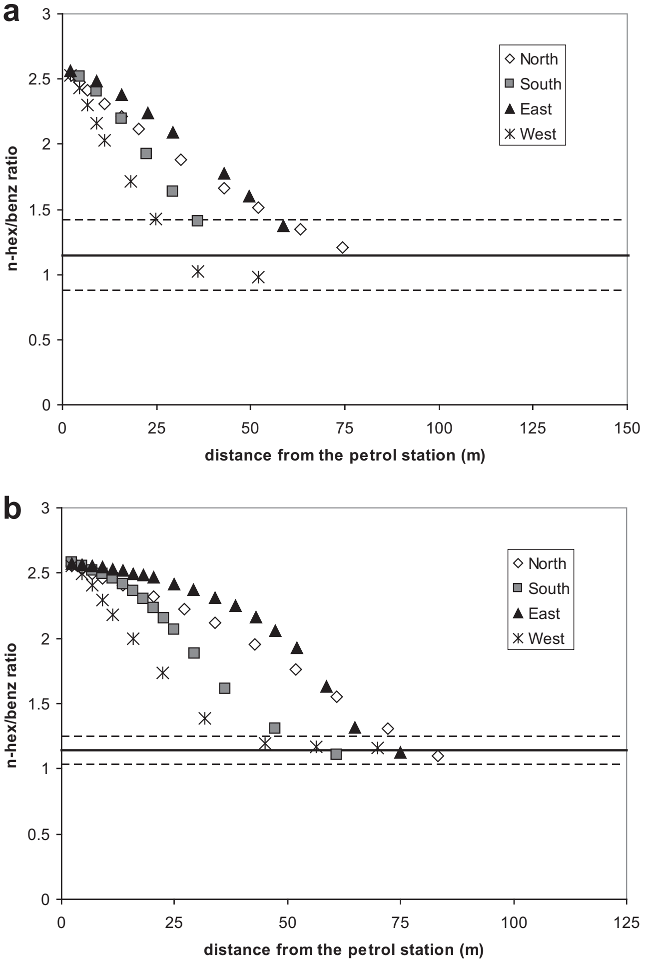

Fig. 5. Relationship of the n-hexane/benzene ratio vs distance from the petrol station. (a) First sampling campaign; (b) second sampling campaign.

Table 3
Maximum spatial influence distances (in meters) of the petrol station “La Fica” in the two sampling campaigns.

Direction	1st Campaign	2nd Campaign
North	60	75
South	36	49
East	60	67
West	25	39
Average	45	58

Finally, we should remember that it is necessary to aim at avoiding the impact of any type of major pollution source on the population. As has been shown before, in the W direction, the influence of the petrol station was lower because a different pollution source was even more important than the one coming from the petrol station with the subsequent harmful effects of that source on the population. Minimization of concentrations of chemicals of concern, regardless of their sources, is indispensable for effective health protection.

5. Conclusions

In this work, we present a methodology for estimating the spatial influence of petrol stations on their surroundings based on the fact that the concentration ratio of n-hexane and benzene found in the air of the petrol stations is different from that found in city air (mainly determined by motor vehicle exhaust). The first point in each geographical direction moving away from the petrol station where the ratio becomes equal to the urban background ratio delimit its area of influence.

The reach distances obtained in the second measuring campaign (June) are higher than those of the first one, probably due to the higher ambient temperature in June which favours evaporation losses in the petrol station during filling up. Anyway, the distances were similar and, in all cases, below or equal to 75 m.

In both campaigns, lower impacts were obtained in those directions with heavy traffic density (W and S), whereas directions N and E, without buildings or roads, showed higher impacts. This can be explained as follows: when the petrol station is surrounded by roads with intense traffic, the emissions from the petrol station are not as important as those coming from vehicle combustion. If the petrol station emissions are mixing with relatively clean air the typical n-hexane/benzene ratio from these stations changes more slowly. From these results, it can be concluded that the influence of petrol stations is a relative question although, in no case, was the influence greater than 75 m.

Presumably, small and medium sized petrol stations will have a similar impact on their surroundings as the one in this work; however, it is recommended that this methodology be applied to specify their reach with more precision. This methodology will help establish a “belt” around petrol stations where vulnerable populations and activities such as those in schools and hospitals should be restricted. Finally, we should remember that minimization of concentrations of chemicals of concern, regardless of their sources, is indispensable for effective health protection. For this purpose, similar or different methodologies should be applied or developed to assess the spatial limits of influence of these other sources.

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