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INDOOR AND OUTDOOR VOLATILE ORGANIC COMPOUNDS MONITORING IN A MULTI-STOREY CAR PARK

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Abstract

A VOC monitoring was carried out inside and outside a multi-storey car park in order to characterize the emission profile of vehicular traffic source in an indoor environment. BTEX, and in particular toluene, were the most abundant compounds in all monitored sites, with different compositions between indoor parking areas and outdoor sites. The motor vehicle exhaust and gasoline vapor emission in these enclosed parking garages were characterized by diffusion or degradation phenomena different from those that occur in urban areas as the BTEX compositions, their ratios and their reactivity with OH and with NO_3 radicals demonstrate. In detail, xylenes/BTEX and ethylbenzene/BTEX ratios in indoor environments were twice than those in outdoor ones, while toluene/BTEX ratio resulted half than that obtained in outdoor sites. In this work, BTEX concentrations depend both on number of vehicles, on vehicular characteristics (age, emission control technology, fuel quality etc.) and on factors related to the characteristics of parking facilities (volume of the monitored areas, indoor or open facilities, ventilation systems, size and maintenance). However, it was found that the external contribution (intrusion of pollutants from outdoor and from the other floors) and the influence of mixing air (removal of pollutants by ventilation and air exchange) on BTEX concentrations were not significant if inside the multi-storey car park there was a strong source (due to the number of vehicles) such happened in the first and second floors and along the ramp that connected them.

Key words: BTEX ratios, garage, indoor environment, number of vehicles, VOC

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1. Introduction

Volatile Organic Compounds (VOC) are considered ubiquitous pollutants and so they are used as important compounds to evaluate indoor and outdoor air quality as discussed in different studies (Amodio et al., 2011; Bruno et al., 2006, 2008a; Caselli et al., 2010; Postolache et al., 2013; Revel et al., 2014).

The most important sources of these compounds in urban areas are represented by vehicles emissions, filling stations, chemical plants and petroleum refineries. In particular BTEX compounds (benzene, toluene, ethylbenzene and xylenes) and methyltertbutylether (MTBE) are pollutants related to traffic emissions (Hun et al., 2011). In and around urban areas, anthropogenic emissions of these VOCs are usually more significant, in fact for this reason they are considered ideal tracers for gasoline-related exposures. The vehicular emissions of BTEX come from different contributions: exhaust emissions (cold and hot), evaporative emissions, and emissions from brake and tyre wear (Franco et al., 2013). Transport-related emissions are important factors in determining air quality in many urban regions, depending on the altitude and thus the dispersion pattern of emissions. In most urban areas, air pollution is now badly affecting the quality of life and the improvement of air quality has become a priority for most cities in

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developed countries. Therefore, many public administrations adopted different strategies to reduce the number of vehicles operating in urban areas, to improve road traffic conditions and to promote the use of public transport. These actions caused the creation of many multi-storey car park well as in urban areas, even in the peripheral areas of a large number of city. In these environments high VOC concentrations can be due to the number of cars that daily attend them (Graham et al., 1999, 2004; Lansari et al., 1996; Murphy et al., 1997; Noseworthy and Graham, 1999; Thomas et al., 1993; Tsai and Weisel, 2000). Exhaust emissions and evaporative emissions produced by vehicles, powered by gasoline or diesel, are the main sources of VOCs in enclosed parking garages (Rakha and Ding, 2003). The exhaust emissions are commonly referred to as "tail-pipe" emissions and are the results of the combustion of fuels within the vehicle's engine.

The operation mode of the vehicle affects the rate of VOCs emissions: in general, they are less during hot starts than cold starts and are lowest for hot stabilized operation mode (Singer et al., 1999). Instead the escape of hydrocarbons from the fuel storage and delivery system represents the evaporative emission. In general very important in the definition of the motor vehicle emission rate of VOCs are parameters such as vehicle characteristics and fuel properties (Krarti and Ayari, 1999).

Air quality in indoor environments also depends on factors related to the characteristics of parking facilities (indoor or open facilities, ventilation systems, size and maintenance).

In this work, a monitoring campaign was carried out inside and outside a multi-storey car park in order to assess VOC concentrations in a confined environment. Moreover the main factors related to the structural characteristics of the parking facility (ventilation rate, climatic conditions, air mixing, size of the environments, number and size of the openings) and number and strength of the sources (number of cars) were evaluated. In detail, an equation was provided and applied to describe how and which factors contribute to indoor VOC concentrations in different investigated environments (parking areas and ramps).

2. Material and method

2.1. Sampling sites

The monitoring campaign was carried out inside and outside a multi-storey car park from 06 to 21 September 2013. Forty 3-day samples were collected in six indoor and four outdoor sites. The multi-storey car park is situated inside of a stone rock mountain located on the Amalfi coast highway (province of Salerno, southern Italy) (Fig. 1). The entrance to the garage is located near a road tunnel and on the side of the Amalfi coast highway, a very busy street that connects two resort towns of the south of Italy as Atrani and Amalfi (on the right and on the left in Fig. 1, respectively). The structure has two exits; the exit for the vehicles is on the opposite site to the entrance, while the second one connects the parking area with the central square behind the garage that can be reached on foot from the garage itself. Inside the parking area, there were 204 car parking spaces and 30 motorcycle spaces spread over four floor. The ground floor has an area of approximately 2150 m^2 and a height of 2.70 m. The first and the second floor have an area of about 2100 m^2 and a height of about 2.60 m, and the third floor, at the top of the building, has an area of 1750 m^2 and a height of about 4.00 m. The entire parking structure is equipped with a ventilation system that draws air from the outside and distributes it within the structure. An additional system directs indoor air outside the building through a pipe located at about one meter below the road surface.

Fig. 1. Position of monitored structure on the Amalfi coast highway

Four monitored sites were selected in outdoor (Site 1 - 4). In detail, site 1 was located next to the vehicles access, adjacent to the road tunnel, on the Amalfi coast highway. Site 2 was positioned next to the exit of vehicles from the parking area on the Amalfi coast highway as well. Site 3 was located in proximity of the aeration system pipe of the parking area. Furthermore, an outdoor site (Site 4) was positioned in the square behind the parking structure.

Six indoor sites were selected inside the rock multi-storey car park. In particular, four indoor sites (Site 5 - 8) were positioned in the center of each level at about 2 m above the floor. Sites 9 and 10 were located instead on the two ramps that respectively connect the third with the second floor and the second with the first floor. The choose of indoor and outdoor sampling sites was performed in order to consider the dimensions of monitored area and so that each of them could sample a homogeneous area.

2.2. Sampling and analytical method

VOC monitoring was performed according to the previously validated procedure (Bruno et al.,

2005, 2008a). VOC were sampled exposing Radiello® diffusive samplers containing graphitised charcoal (Carbograph 4) as adsorbent beds. Then, collected cartridges were thermally desorbed (Markes International, Llantrisant, UK) and a gas chromatography-mass spectrometry analysis of VOCs (GC-6890 PLUS, MS- 5973 N; Agilent Technologies, Santa Clara, California, USA) was carried out (UNI EN ISO 16017-2, 2007). For quantitative analysis, standard solutions were prepared by injecting into cartridges successive dilution in methanol of a VOC standard mixture at 2,000 µg/mL (Cus-5997 Ultra Scientific, Bologna, Italy). Atmospheric concentration of each detected VOC was calculated in accordance with Bruno et al. (2005, 2008b).

3. Results and discussion

Twenty-two volatile organic compounds were detected in indoor and outdoor sites during the monitoring campaign. Average VOC concentrations and variability over the sampling period are listed in Table 1.

Data analysis showed that indoor concentrations were higher than outdoors one for all monitored sites, with the exception of the parking area on the third floor. The indoor concentrations in this area were comparable to those detected in outdoor sites. BTEX were the most abundant compounds among detected VOCs both in indoor and outdoor sites. In addition, a significant contribution of alkanes (on average 16% compared to the sum of VOCs detected) was observed. In detail, the ratios BTEX/VOCs were on average equal to 68% both in indoor sites (ranging from 67% to 69%) and outside (ranging from 65% to 76%). However, VOC percent compositions observed in indoor parking areas and outdoor sites were different. This result confirmed that a different pattern of VOCs characterized traffic emission source in indoor environments.

Toluene was the most abundant compound in all monitored sites, but difference in percentage composition was observed among the investigated sites. In fact in outdoor sites, Toluene accounted on average for 56% and 38% of the composition of BTEX and detected VOCs, respectively. These percentages were higher than those observed in parking areas (35% and 24%) and long ramps (26% and 19%) of the multy-storey car park. On the contrary, higher percentage composition of xylenes were found (X/BTEX percentage = 37%; X/VOCs percentage = 25%) in indoor air. Same finding was found for ethylbenzene: its indoor percentage compositions (E/BTEX percentage = 11%; E/VOCs percentage = 10%) were double than those for outdoor sites.

These evidences suggested that vehicle exhaust emissions (when the vehicles are moving) and evaporative emissions (when the vehicles are parked) in a confined environments can be affected by diffusion or degradation phenomena different from those that occur in urban areas or in general in outdoor sites.

This evidence was further confirmed from the higher variability over the sampling periods obtained for indoor concentrations, as showed in Fig. 2 for benzene concentrations.

Several studies conducted in urban areas demonstrated that the ratios among BTEX are diagnostic to indicate that emissions are predominantly from vehicles (Amodio et al., 2011; Bruno et al., 2006, 2008a; Caselli et al., 2010). In detail, Benzene/Toluene (B/T) (values between 0.26 and 0.50) ratios are commonly used as indicators of the predominance of the vehicular traffic emission, while xylenes/ethylbenzene (X/E) ratios give an indication of the photochemical aging of the emission and thus of the proximity of the emission source. Average indoor and outdoor diagnostic ratios obtained for the monitoring campaign are listed in Table 2.

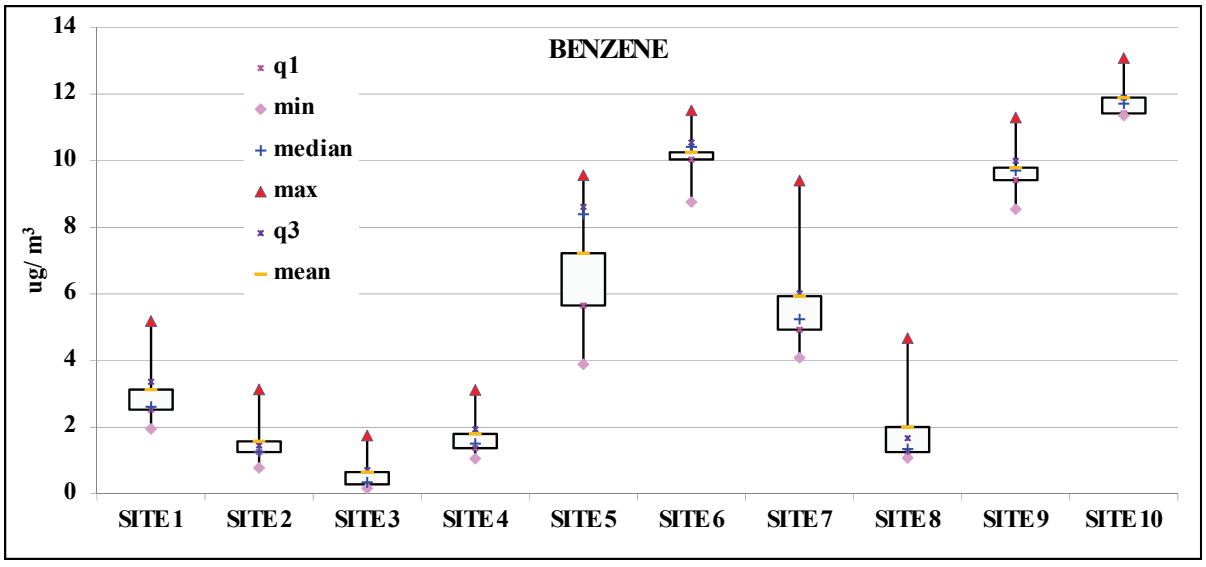

Fig. 2. Trend of the levels of benzene during the monitoring campaign in outdoor sites (1-4) and indoor sites (5-10)

Table 1. Average VOC concentrations and variability over the sampling periods

	Site 1		***Site 2***		***Site 3***		***Site 4***		***Site 5***		***Site 6***		***Site 7***		***Site 8***		***Site 9***		***Site 10***	
	Outdoor		***Outdoor***		***Outdoor***		***Outdoor***		***Ground floor***		***First floor***		***Second floor***		***Third floor***		***Ramp***		***Ramp***	
	ug/m^3	SD	ug/m^3	SD	ug/m^3	SD	ug/m^3	SD	ug/m^3	SD	ug/m^3	SD	ug/m^3	SD	ug/m^3	SD	ug/m^3	SD	ug/m^3	SD
MTBE	0.2	0.1	0.21	0.12	0.12	0.03	0.27	0.08	2.24	2.95	0.9	0.2	0.77	0.37	0.3	0.23	0.93	0.25	1.61	1.66
ETBE	1.56	1.34	0.66	0.61	0.44	0.06	1.66	1.07	2.62	2.55	3.51	1.12	1.82	1.19	0.71	0.78	3.03	1.07	5.18	1.36
1,1,1-trichloroethane	0.04	0.02	0.04	0.02	0.05	0	0.054	0	0.03	0.04	0.05	0.03	0.04	0.02	0.04	0.03	0.053	0	0.056	0
Benzene	3.13	1.26	1.58	0.9	0.54	0.68	1.8	0.91	7.22	2.37	10.25	1	5.94	2.06	2	1.51	9.79	1	11.89	0.7
Heptane	5.25	2.17	1.78	0.84	0.83	0.67	2.84	1.58	9.94	8.3	13.22	9.66	6.01	2.55	1.82	0.71	10.8	3.7	15.53	4.61
1,1,2-trichloroethane	0.033	0	0.033	0	0.03	0.01	0.033	0	0.04	0.02	0.06	0.06	0.07	0.09	0.04	0.02	0.09	0.12	0.1	0.12
Toluene	9.57	1.19	6.54	1.51	3.71	1.56	8.22	1.27	13.79	2.1	16.51	1.43	13.72	2.3	8.48	2.04	15.8	1.61	18.86	2.16
Octane	0.24	0.05	0.17	0.06	0.1	0.05	0.2	0.04	0.55	0.24	0.91	0.46	0.58	0.54	0.2	0.12	0.84	0.54	1.08	0.67
Tetrachloroethylene	0.08	0.06	0.09	0.05	0.08	0.04	0.13	0.06	0.08	0.06	0.11	0.1	0.12	0.14	0.08	0.06	0.15	0.17	0.21	0.21
N-butyl acetate	0.06	0.08	0.06	0.08	0.01	0.01	0.04	0.04	0.09	0.15	1.04	2.17	1.96	4.34	0.1	0.16	2.45	5.33	8.41	10.7
Ethylbenzene	1.5	0.49	0.67	0.4	0.19	0.25	0.96	0.37	3.71	1.67	7.08	2.63	4.21	3.37	1.18	0.96	6.82	3.5	11	5.2
M/p-xylene	3.79	0.96	1.93	1.26	0.62	0.83	3.09	1.02	8.66	2.83	12.78	2.6	8.21	4.6	2.93	2.28	12.46	3.12	15.6	3.48
Styrene	0.14	0.03	0.14	0.05	0.09	0.04	0.14	0.03	0.46	0.13	0.6	0.2	0.36	0.17	0.32	0.09	0.54	0.11	0.63	0.16
O-xylene	2.07	0.53	1.03	0.74	0.3	0.45	1.67	0.55	5.81	2.69	9.82	3.02	5.54	4.35	1.51	1.24	9.5	3.28	12.42	3.77
Nonane	0.14	0.03	0.14	0.05	0.11	0.02	0.13	0.03	0.43	0.33	0.69	0.74	0.41	0.47	0.14	0.04	0.38	0.22	0.33	0.14
Alpha-pynene	0.24	0.08	0.26	0.13	0.22	0.03	0.3	0.09	0.24	0.09	0.32	0.11	0.26	0.09	0.23	0.09	0.24	0.05	0.36	0.09
Camphene	0.15	0.14	0.16	0.17	0.15	0.16	0.24	0.18	0.19	0.22	0.19	0.18	0.22	0.2	0.17	0.19	0.19	0.2	0.18	0.17
1,2,4-trimethylbenzene	0.89	0.62	0.98	1.3	< LOD	-	0.94	0.97	3.73	2.71	3.58	4.17	7.03	3.02	0.47	0.19	4.78	2.6	2.8	1.62
Decane	0.12	0.01	0.14	0.06	0.12	0.03	0.12	0.03	0.22	0.15	0.49	0.62	0.27	0.29	0.13	0.03	0.2	0.1	0.18	0.06
1,4-dichlorobenzene	0.024	0	0.026	0	0.025	0	0.028	0	0.025	0	0.025	0.01	0.025	0	0.024	0	0.025	0	0.025	0
Limonene	0.16	0.01	0.19	0.07	0.16	0.02	0.26	0.07	0.18	0.04	0.24	0.14	0.22	0.09	0.15	0	0.16	0.01	0.51	0.09
Undecane	0.17	0.01	0.19	0.06	0.18	0.04	0.19	0.03	0.2	0.04	0.37	0.3	0.27	0.19	0.18	0.02	0.22	0.04	0.23	0.04
ΣVOCs	29.57	9.19	17.01	8.51	8.08	4.98	23.34	8.42	60.46	29.7	82.72	30.9	58.06	30.5	21.19	10.8	79.45	27	107.19	37
Σ BTEX	20.07	4.43	11.75	4.82	5.37	3.77	15.75	4.13	39.19	11.7	56.44	10.7	37.62	16.7	16.1	8.02	54.38	12.5	69.77	15.3
Σ Alifatics	5.93	2.27	2.42	1.07	1.35	0.81	3.49	1.7	11.35	9.06	15.69	11.8	7.55	4.04	2.47	0.92	12.44	4.6	17.35	5.51
Others	3.57	2.49	2.84	2.63	1.37	0.39	4.1	2.59	9.93	8.96	10.59	8.49	12.9	9.74	2.62	1.83	12.63	9.91	20.07	16.2

Table 2. Average indoor and outdoor diagnostic ratios

Diagnostic ratios	*Outdoor sites*	*Parking area*	*Ramps*
B/T	0.23	0.54	0.63
X/T	0.47	0.99	1.44
MTBE/B	0.14	0.17	0.12

B/T ratios confirmed that exhaust vehicle emissions affected mainly indoor and outdoor air quality. This evidence was further highlighted plotting benzene versus toluene concentrations (Fig. 3): the high linearity confirms the presence of a common source of two pollutants, while the different slope of the straight lines indicates that pollutants may have suffered different degradation or diffusion phenomena in outdoor sites and in indoor sites.

BTEX compounds were characterized by a reactivity that consists in two main depletion reactions: with OH radicals in presence of light and with NO_3 radicals in the absence of light. The rate constants for the reactions of BTEX with OH radicals are so higher than those with NO_3 that with OH can be considered as predominant. Xylenes are the most reactive compounds with respect to the OH reaction, followed by ethylbenzene, toluene, and benzene. In indoor environments, and in particular on the ramps characterized by lower lighting, this reaction can result lower with respect to that expected in open atmosphere. This phenomenon can explain the higher X/T ratios obtained in parking areas and long the ramps. In addition, the ratio of methyltertbutylether (MTBE) (used as an additive in gasoline) to benzene indoor concentrations can be used to examine if these compounds originated from gasoline vapors or car exhaust. Low MTBE/benzene ratios (0.40) indicate that tailpipe emissions are dominant because during combustion the amount of MTBE decreases while benzene is enriched due to toluene and xylene dealkylation (Zielinska et al., 2006).

Alternatively, high MTBE/benzene ratios (up to 50) suggest a significant contribution from evaporative emissions from hot soak and/or fuel system leakage. MTBE/B ratio obtained in this study (0.15) confirmed car exhaust emissions as main source of VOCs both in indoors and outdoors.

Data analysis showed that different levels of BTEX were detected in monitored environments of the multi-storey car park. Several studies showed that pollutant concentrations in an indoor environment depend not only on chemical degradation, but on several factors such as number and strength of the sources, ventilation rate, climatic conditions, air mixing, building characteristics, and possible sinks (e.g. sorption by surfaces and furnishings) (Graham et al., 2004; Spengler and Chen, 2000).

To study the impact of these factors on detected VOC concentrations, Eq. (1) describing the relationship among them in the multi-storey car park is reported.

$$C = \left(\frac{K_1 \times N}{V} \right) + K_2 - K_3 \quad (1)$$

where: $K_1 \times N$ represents the contribution due to the number of vehicles that entered inside the parking area during each monitoring period; V is the volume of the parking area in each floor, expressed in m^3; K_2 represents the external contribution due to intrusion of pollutants from outdoor and from the other floors; K_3 represents the contribution due to removal of pollutants by ventilation and air exchange.

In detail, the first term of the equation in brackets describes both the increase in the concentration linked to the number of vehicles and the possible decrease due to the dilution of pollutants in the volume of the environment. In order to immediately display which factors had a greater effect in the different environments monitored, benzene concentrations per site versus the number of vehicles are showed in Fig. 4.

The lack of correlation between benzene concentration and number of vehicles on the ground floor (Site 5) can be explained by considering the characteristics of the environment.

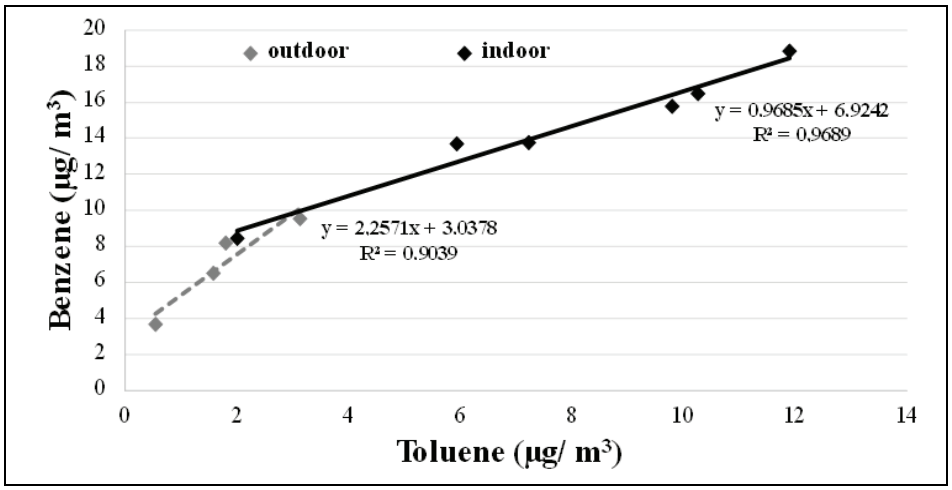

Fig. 3. Benzene concentrations versus toluene concentrations in indoor and outdoor sites

The presence of wide openings for access and exit of vehicles produced a dispersion of pollutants variable in time (high contribution of K_3). High correlation was found at first and second floor (Site 6 and Site 7), respectively. A higher correlation revealed in Site 6 (R^2 = 0.965) can be explained considering the greater number of vehicles that reached the first floor than the second one. Therefore, in this case K_2 and K_3 can be considered negligible. The third floor (Site 8) was characterized by a volume greater than the other floors and it was reached by a lower number of vehicles. These factors explain the lower correlation found in this parking area (R^2 = 0.853).

The highest benzene and VOC concentrations were detected along the two ramps (Site 9 and Site 10) that connect the third with the second floor and the second with the first floor, respectively. In detail, the highest correlation (R^2 = 0.962) was obtained in Site 10 as it was crossed by cars that reached the second and third floors. Therefore, the concentrations detected in this environment were mainly due to the number of cars (high contribution of K_3). However, the small size of the environment (small volume) and the absence of openings (low contribution of K_2) may have contributed to detected levels.

To evaluate indoor air quality (IAQ) in the multy-storey car park, indoor VOC concentrations were compared with others monitored in non-residential indoor environments by using the same methodology (Amodio et al., 2014; Bruno et al., 2008b, 2009; de Gennaro et al., 2013).

It was found that, although in the multi-storey car park there was a continuous source of volatile compounds, the levels of BTEX and in particular of benzene obtained in this study were comparable and, in many cases, lower than those monitored in businesses, schools and offices, environments characterized by the small size and reduced air exchange (Table 3).

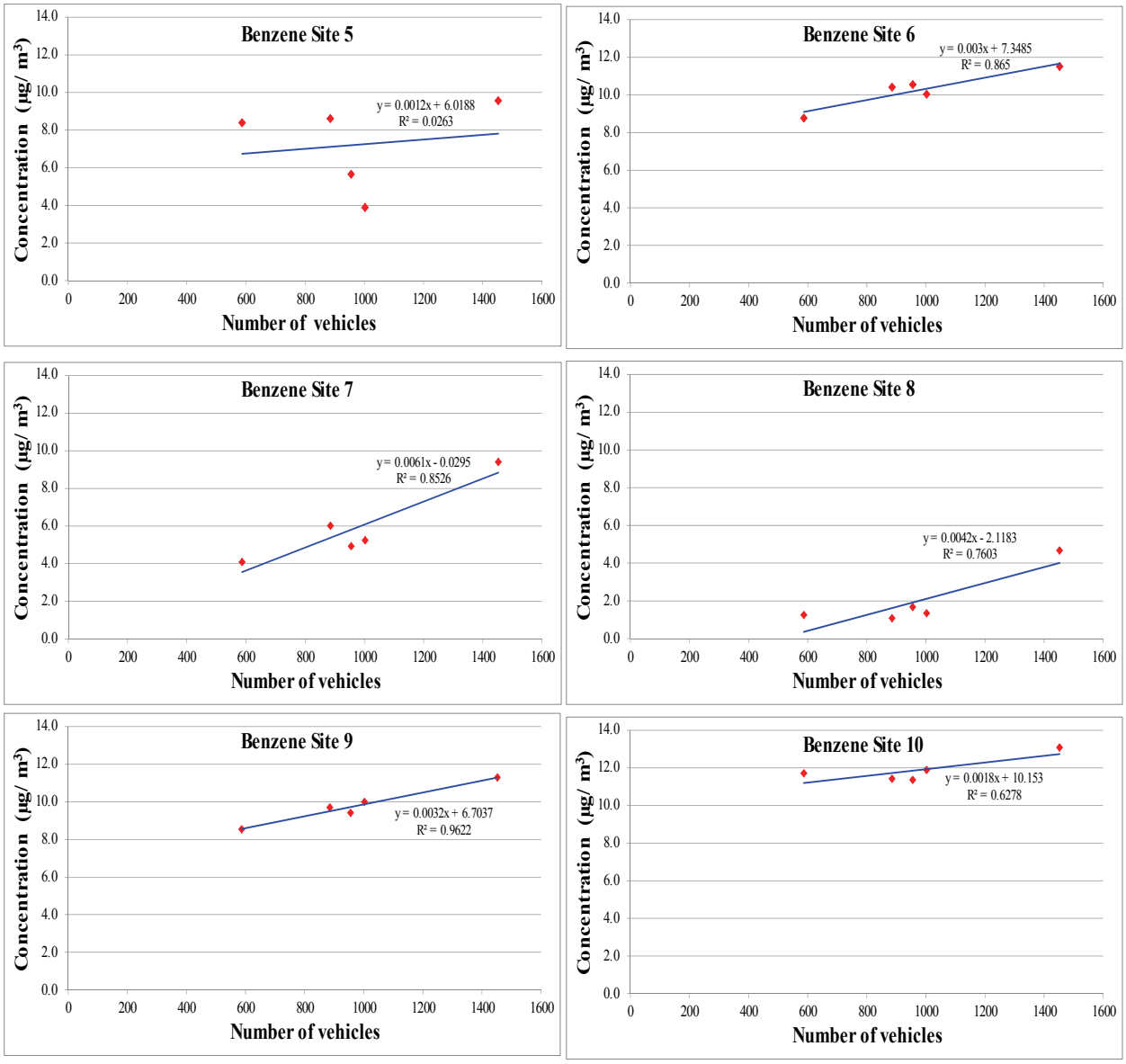

Fig. 4. Benzene concentration in indoor sites vs the average number of vehicles in each monitoring periods

Table 3. Indoor concentration of BTEX and benzene in several not-residential indoor environments

	BTEX (μg/m^3)	***Benzene (μg/m^3)***	***References***
Coffee shops	271	5	Bruno et al. (2008)
Libraries	38	2	Bruno et al. (2008)
Copy centers	545	10	Bruno et al. (2008)
Pharmacies	834	7.5	Bruno et al. (2008)
Newspaper stands	690	3	Bruno et al. (2008)
Offices	26	3.3	Bruno et al. (2008)
Gymnasiums	69	1.7	Bruno et al. (2008)
Hairdressing salons	150	2.7	Bruno et al. (2008)
Restaurants	79	2.5	Bruno et al. (2008)
Supermarkets	90	2.2	Bruno et al. (2008)
School Buildings	4.2	0.04-6	de Gennaro et al. (2013)
Multi-storey shopping mall	18	0.7-9	Amodio et al. (2014)
Multi-storey car park	45.6	7.9	This study

4. Conclusions

The study of VOC concentrations, the reactivity of BTEX, the main structural parameters and the number of cars that characterized the multi-storey car park highlighted that, in an indoor environment, diffusion or degradation phenomena are different from those that commonly occur in urban areas. Furthermore, a comparison with other non-residential indoor environments showed that the structural characteristics of the buildings, such as air exchange systems and the high volumes of the environments could have a considerable impact on the indoor air quality even when a strong emission source is present.

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