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Analysis of Gasoline Used by Motorbike-Taxi Drivers in Cotonou

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Abstract

In 2009, samples from itinerant stations were taken in Cotonou. Official reference gasoline stations (French and boundary African country) were also studied. The analysis was done by gas chromatography coupled to a mass spectrometer (GC-MS). The results showed appreciate difference between itinerant and official gasoline samples: ETBE (Ethyl Tert-Butyl Ether) was found in the composition of the French reference but not in the African gasolines composition. Benzene was detected at yields less than 1% in the French reference gasoline whereas in the gasoline made in the African countries benzene yields were between 2.36 and 4.39%. The ratio of toluene/benzene varied between 1.27 and 10.25 in studied gasoline and was much lower in ambient air. For major components (aromatics and paraffin compounds), we observed another heterogeneity for itinerant gasoline (probably due to numerous hands making and transport).

Keywords: ETBE, toluene/benzene ratio, gasoline, motorbike-taxi drivers, Cotonou

1. Introduction

All the way from Cotonou, to approximately 3 km of the international airport Bernardin GANTIN, it is easy to note an exposition to gasoline carboys along the roadside and to observe above the different crossroads, a thick layer of smoke which wraps the population. These gasoline carboys contain unleaded gas coming from Nigeria (Avogbe et al., 2011) and frequently crossing the countryside all the way to Cotonou without taking any precaution.

Nearly 90% of Cotonou's population gets its supplies of gasoline from these exposed carboys. This preference is due to the low income and the low costs of this gasoline coming from Nigeria, supported by the currencies exchanges on the unofficial market (currently 300 FCFA in 2009 against 490 FCFA in the official stations). This phenomenon is of big concern because it contributes to the deterioration of the environment and a worrying atmospheric pollution.

In such situation, a pilot study has been conducted to examine the quality of the ambient air (Avogbe et al., 2005; Ayi-Fanou et al., 2006). Our results showed that the ambient air in Cotonou contained many air pollutants including volatile organic compounds (VOCs) like benzene and its derivatives, ultrafine particles (UFPs) associated with polycyclic aromatic hydrocarbons (PAHs) at high concentrations. The benzene reaches an average concentration of 76 $\mu g/m^3$, a value higher than the admissible concentration of 5 $\mu g/m^3$ recommended by the WHO; PAHs and UFPs reach 103 ng/m^3 and 166816 particles/m^3 respectively, against 1.55 ng/m^3 and 5276.5 particles/m^3 in a pilot site located at 80 km to Cotonou (Sohon village).

Moreover, aren't any petrochemical industries in Benin that use benzene, a product classified as a carcinogenic compound (International Agency for Research on Cancer [IARC], 1974).

At the moment, Benin has adopted the unleaded gas, and new investigations have been led to know the potential origin of the pollutants which have been encountered in the air. The main purpose was to know if the gasoline sold in Benin (carboys along the road and official stations) respected the international standard specification with a benzene concentration of 1% in the gasoline (Kolmetz & Gentry, 2007) similar to that sold in the developed countries such as France. Indeed, the unleaded gasoline is mainly a mixture of hydrocarbons and oxygenated compounds such as Ethyl Tert-Butyl Ether (ETBE) (Linnekoski, Krause, & Rihko, 1997; Reich, Cartes, Wisniak, & Segura, 1999; Vinuesa, Mirabel, & Ponche, 2003) which increase the octane number of the gasoline (Dagaust, 2007) and reduce the concentrations of aromatic compounds in the mixture (Nadim, Zack, Hoag, & Liu, 2001). This additive can be added to a total value of 15% in the gasoline in France. The ethanol seems to decrease the steam pressure of the gasoline, reducing the losses by evaporation (da Silva, Cataluna, de Menezes, Samios, & Piatnicki, 2005). In contrast, the addition of ethanol in gasoline reduces by 10% the emission of carbon monoxide (Poulopoulos, Samaras, & Philippopoulos, 2001). Furthermore, an American study carried out within the framework of American Car/Oil Program showed that the addition of ETBE in the gasoline leads to a reduction in the carbon monoxide emissions (Kivi, Niemi, Nylund, Kuto, & Orre, 1992; Reuter et al., 1992; Noorman, 1993) and hydrocarbons (Guibet, 1998; de Menezes, Cataluña, Samios, & da Silva, 2006).

Thus, samples of gasoline were taken in Cotonou from the official stations and carboys along the roadside. The main objective was to determine their composition in benzene and its derivatives and in ETBE and to compare their concentrations to those in French unleaded gasoline label “Sans Plomb 98” and motorbike-taxi drivers (MBTD) exposition followed for few years (Avogbe et al., 2011).

2. Method

2.1 Sampling Campaign

In August 2009, gasoline samples were taken in Cotonou (Figure 1) and Nigeria. Samples were convoyed for analysis in Dunkirk, France. The samples were selected as follow: one French gasoline from Total France sampled in Dunkirk and thirteen African gasolines originated from Nigeria and Benin. The latter were sampled in official stations (Oryx, Texaco, Total, Sonacop, and Pegaz) and carboys along the roadside (at Vedoko, Opke-oluwa, St Michel, Zongo and red Star site “Place Etoile rouge”). Samples were collected in 2 mL sealed vials (3 replicates) only one was analysed and referred in the following way: gasoline from France: SP 98, gasoline from Benin: samples 1 to 10 and gasoline from Nigeria: samples 11 to 13. All the samples were stored in the freezer at -20 °C before analysis.

2.2 Analytical Conditions

Volatile and semi-volatile hydrocarbons analysis were based on the analytical methods described by Caplain et al. (2006). 1 µl of each sample was injected (split ratio of 20:1) in a gas chromatograph (VARIAN 3800) coupled to a mass spectrometer (VARIAN 1200 TQ). The capillary column is a Factor four VF-5 ms (30 m x 0.25 mm x 0.25 µm) and helium used as carrier gas. The total time for this analysis was 60 mins; 40 °C for 5 mins, 5 °C/min up to 300 °C and 310 °C for 3 mins. The parameter of mass detector was impact electronic ion current = 70eV and temperature of source 280 °C. The samples were analyzed between 40 and 350 mass unit. Benzene and ETBE were identified based on their characteristic ions: m/z = 78 for benzene and m/z = 57 and 89 for ETBE. The total composition of the gasoline was given by using the mode TIC (Total Ion Current).

2.3 Identification of Compounds

Compounds were identified by comparing retention times of chromatographic peaks from gasoline samples with those from standard mixtures and by comparing mass spectra with those contained in NIST and/or WILEY libraries. The percentage of each compound was determined using TIC surface areas.

2.4 Study of the Exposure of Motorbike-Taxi Drivers to Benzene and Toluene

Benzene and toluene ambient concentrations of personal exposure were determined by using Radiello diffusive samplers consisting of an active carbon cartridge enclosed in a porous polyethylene body (Fondazione Salvatore Maugeri, Paduva Italy). The Radiello tube has an uptake of 80 m^3 per min at 25 °C and 1 atm. Benzene, toluene, ETBE, paraffin and aromatic compounds were analysed by GC-MS using the method described by Sørensen, Skov, Autrup, Hertel, and Loft (2003). The ratio of toluene/benzene was used to evaluate the variability and origin of fuel (Guo, Lee, Louie, & Ho, 2004).

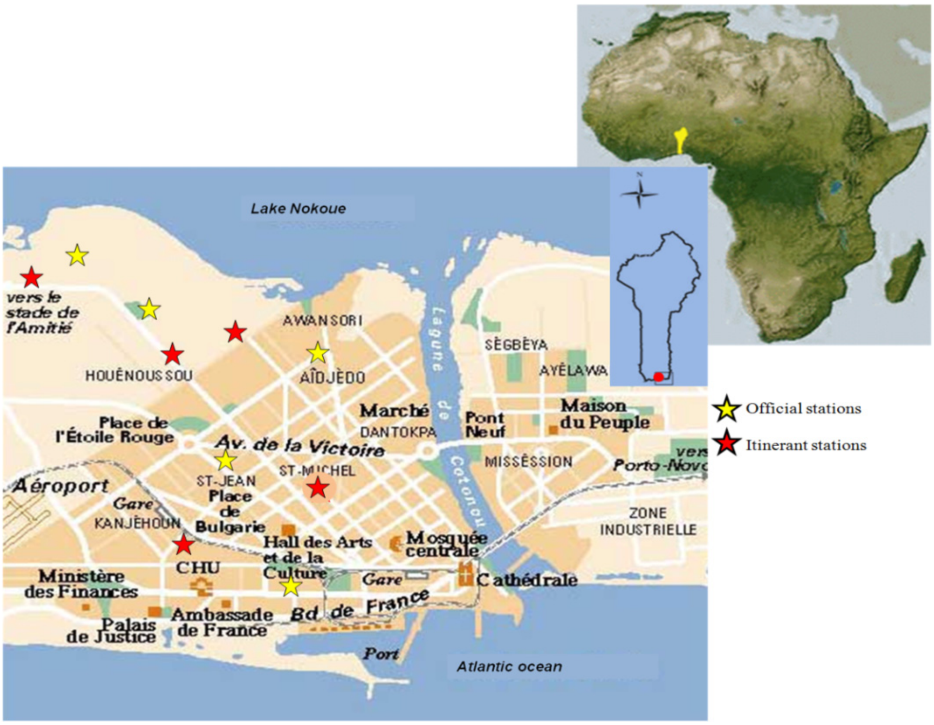

Figure 1. Geographical location of the sampling site in Cotonou

3. Results and Discussion

Alkyl-lead compounds are replaced by oxygenated compounds which have become increasingly important components in automotive gasoline, thus improving octane rating (Nadim et al., 2001) and reducing the emission of carbon monoxide (de Menezes et al., 2006). One of the most widely used oxygenated compounds in gasoline is ETBE (van Wezel, Puijker, Vink, Versteegh, & de Voogt, 2009). We will show in this section the main results of gasoline compositions (Benzene, toluene, ETBE, paraffin and aromatics) present in gasoline using by motorbike-taxi drivers of Cotonou. The Table 1 show the compositions of gasoline (benzene, ETBE) determined by GC-MS analysis.

Table 1. Benzene and ETBE amounts (percent) in the samples of examined gasoline

	Sample	Benzene	Median	Mean ± SD	ETBE
SP 98	SuperPlus	0.69			13.68
Official stations Benin	2	4.15	3.58	3.44 ± 0.91	n.d.*
	3	2.92			n.d.*
	6	3.58			n.d.*
	8	2.18			n.d.*
	10	4.39			0.75
Official stations Nigeria	11	2.36	3.16	3.21 ± 0.87	n.d.*
	12	4.10			n.d.*
	13	3.16			n.d.*
Itinerant stations	1	1.98	2.84	2.81 ± 0.64	0.29
	4	2.84			n.d.*
	5	3.19			n.d.*
	7	3.63			n.d.*
	9	2.42			0.65

* n.d.: not detected

For these studied gasoline, we can see that the compositions of all thirteen samples are relatively similar for gasoline originating from both official stations and itinerant stations (African stations). ETBE are not detected in all samples except Pegaz of official stations, Vedoko and Etoile of itinerant stations who have respectively 0.75, 0.29 and 0.65% of ETBE whereas it was 13.68% in SP 98 of France. According to french regulations, ETBE concentration in gasoline can reach 15% (Norme NF EN 228, 2008). Westphal, Krahl, Brüning, Hallier, and Bünger (2010) and Kim, O'Shea & Cooper (2012) had similar results. There was no difference concerning the percentage of benzene in all examined gasolines. Official and itinerant stations showed a remarkably higher percentage of benzene. The highest percentage (4.39%) was observed in official stations whereas in SP 98 it was less 1% either three to six times as high as french unleaded gasoline. The benzene concentration level in the gasoline is 1% in international standard specification (Kolmetz & Gentry, 2007). Some studies showed that the gasolines respect this regulation (Simon et al., 2004; Adami et al., 2006; Bonfim, Alves, & Filho, 2012). Table 2 lists some average ETBE, aromatic compounds, paraffin and ratio of toluene/benzene for various countries.

Table 2. Summary of average ETBE, aromatic compounds, paraffin and ratio of toluene/benzene in gasoline for different countries

Cities/Countries	ETBE	Aromatics	Paraffin	Ratio T/B	Benzene	Reference
Germany	10%	-	-	-	-	Westphal et al., 2010
USA	15.7%	-	-	-	-	Kim et al., 2012
Italy	-	29.02-41.77%	36.68-44.75%	6.59-15.54	0.39-1.06%	Adami et al., 2006
Brazil	-	-	-	-	0.30-0.60%	Bonfim et al., 2012
Brazil (Sao Paulo)	-	-	-	1.68	16.7µg/m^3	Gee et al., 1998
Chile (Santiago)	-	-	-	2.01	14.8µg/m^3	Gee et al., 1998
Chine (Hong Kong)	-	-	-	7.07	10.05µg/m^3	Lee et al., 2002
Spain	-	64%	-	-	-	Montells et al., 2000
Scotland	-	30.5%	-	-	-	Sentenac et al., 2012
Brazil	-	26.90-38.50%	10.10-13.70%	-	-	de Menezes et al., 2006
USA (Atlanta)	-	-	-	5.14	0.55%	NESCAUM, 1995
USA	-	-	-	3.56	11.50 mg/mL	Kaplan et al., 1997
Brazil	-	-	-	3.33	30 g/L	Montells et al., 2000

(-) not available

ETBE is also used in gasoline formulation to decrease the concentration of aromatics (Pumphrey, Brand, & Scheller, 2000; Nadim et al., 2001).

Table 3 shows trends of paraffin and aromatic hydrocarbons found in all three types of gasolines.

Table 3. Values (percent) of paraffin and aromatic hydrocarbons in the three types of gasoline

	Paraffin	Median	Mean ± SD	Aromatic	Median	Mean ± SD
SP 98	5.41			44.70		
Official stations	5.07	5.57	5.57 ± 0.46	39.91	44.26	43.92 ± 3.87
Benin	6.25			40.14		
	5.57			48.51		
	5.23			46.80		
	5.71			44.26		
Official stations	6.15	2.37	3.62 ± 2.19	51.51	51.56	51.68 ± 0.26
Nigeria	2.34			51.56		
	2.37			51.98		
Itinerant stations	8.54	6.88	5.91 ± 2.26	39.40	41.39	41.12 ± 2.18
	6.88			41.39		
	7.04			41.49		
	3.75			38.91		
	3.32			44.42		

In this study, total concentration of aromatics was equivalent in studied gasoline (Mean: 41.12 to 51.68%). These concentrations were lower than the aromatics level in unleaded gasoline in Spain (Montells, Aceves, & Grimalt, 2000) and higher than the level in Scotland (Sentenac, Ayeni, & Lynch, 2012), Brazil (de Menezes et al., 2006) and Italy (Adami et al., 2006) (Table 2). This high level of aromatics in examined gasolines could be probably due to the weak rate of ETBE. Furthermore the total paraffin values of official stations of Benin ranged from 5.71 to 6.25% against 3.32 to 8.54% for itinerant stations and 2.34 to 6.15% for official stations of the border between Benin and Nigeria. French unleaded gasoline showed 5.41 percent of paraffin. These concentrations of paraffin were lower than those reported in Brazil (de Menezes et al., 2006) and Italy (Adami et al., 2006). The means and medians obtained show a better homogeneity resulting from the three types of gasoline as well as for total paraffin and total aromatic hydrocarbons. Some authors used concentration ratios of toluene/benzene in order to evaluate different types of fuel and rate of diesel or gasoline vehicles driven in different urban sectors (Guo et al., 2004).

Table 4 present toluene/benzene ratio in examined gasoline.

Table 4. Ratio of toluene and benzene in the three types of gasoline

	Benzene (%)	Toluene (%)	Toluene/benzene	Mean ± SD
SP 98	0.69	7.03	10.25	
Official stations	4.39	5.59	1.27	2.76 ± 1.38
Benin	4.15	6.30	1.52	
	2.92	11.38	3.89	
	3.58	9.79	2.74	
	2.18	9.56	4.38	
Official stations	2.36	6.44	2.73	2.83 ± 0.26
Nigeria	4.10	10.83	2.64	
	3.16	9.88	3.13	
Itinerant stations	1.98	11.45	5.77	3.76 ±1.30
	2.84	12.02	4.24	
	3.19	10.42	3.27	
	3.63	8.84	2.43	
	2.42	7.51	3.10	

The mean toluene/benzene (T/B) ratio of unleaded gasolines of official stations in Benin, Nigeria and itirenant stations were respectively 2.76, 2.83 and 3.76 (Table 4). These results showed a similarity between these different stations. The toluene/benzene ratio in gasoline is in the order of 9 (Institut National de l'Environnement Industriel et des Risques [INERIS], 2010). This ratio was 10.25 in unleaded gasoline SP98 of France. On the other hand the value of this ratio was low in Atlanta city (5.14 while considering percent of toluene and benzene, Northeast States for Coordinated Air Use Management [NESCAUM], 1995), USA (3.56 while considering average (mg/mL) of toluene and benzene, Kaplan, Galperin, Lu, & Lee, 1997) and Brazil (3.33 while considering concentration (g/L) of toluene and benzene, Montelles et al., 2000). This ratio varied from 6.59 to 15.54 in examined gasoline in Italy considering the percentage of toluene and benzene (Adami et al., 2006). The values obtained in this study vary from 2.76 to 3.76. The differences observed between values obtained in African and developed countries may be due to differences in the composition of gasoline and/or concentration of compounds. The results showed a low ratio of toluene/benzene in sampled gasolines which indicates a high concentration of benzene in gasolines.

The main source of benzene and toluene is vehicle exhaust gases and Lee, Chiu, Ho, Zou, and Wang (2002) described that the ratio of toluene/benzene increases with traffic. Meuwese-Mulder (2006) show clearly that at remote urban sites ratios of toluene to benzene are often below 1, the well-known standard values ranging from 3 to 5 corresponding to a city's background. This result is also confirmed by the paper of Gelencsér, Siszler, and Hlavay (1997), where the ratio of toluene/benzene was used as a tool for charactezing the distance from vehicles emission sources (1 to 1.5). For Xu, Pereira, Miller, Grgicak-Mannion, and Wheeler (2010), benzene is predominantly emitted from traffic whereas toluene is generated from both traffic and solvents. The ratio between toluene and benzene (T/B) can act as an indicator of traffic emissions when this ratio is within the range of 1.5 to 4.3 as repeated by previous studies (Hoque, Khillare, Agarwal, Shridhar, & Balachandran, 2008; Liu et al., 2009). For ratios > 4.3 solvent source compounds likely. For fresh plumes ratio of toluene/benzene was 2 to 2.4 and aged to < 1 (Bahreini et al., 2012). In this study, the results of personal exposure monitoring are presented in Table 5.

Table 5. Summary of personal exposure monitoring for one-week

Compounds	motorbike-taxi drivers					
	1	2	3	4	5	Mean ± SD
Benzene (μg/m^3)	14.05	4.65	24.57	44.79	87.27	35.07
Toluene (μg/m^3)	0.82	3.34	5.51	2.90	2.86	3.09
Ratio T/B	0.06	0.72	0.22	0.06	0.03	0.22

The benzene concentration of MBTD over one-week average exposure was more ten times as high as toluene concentration. Therefore the ratio of T/B was between 0.03 and 0.72. These results indicated much lower ratio of T/B within MBTD. This ratio in air is in the order of 5 (INERIS, 2010). In Toulouse City, Simon et al. (2004) have found ratio of T/B equal to 5.54, 2.90 to 3.40, 2.20, 3.80 to 4.40, 8.60, and up to 10 were observed in Paris (Vardoulakis, Gonzalez-Flesca, & Fisher, 2002), Copenhagen (Hansen & Palmgren, 1996), Mortsel (Buczynska et al., 2009), Ren-Wu (Hsieh, Yang, & Chen, 2006), Hong Kong (Lee et al., 2002), respectively. Buczynska et al. (2009) suggested the same common source (traffic exhausts) of toluene and benzene as an explanation for such low values of T/B ratio. In this work, this ratio was much smaller because of high benzene concentration level (Mean: 35 μg/m^3). Benzene concentration in Cotonou ambient air was seven times as high as the European limit value (5 μg/m^3). It suggests the existence of other sources than traffic exhausts which could result using itinerant gasoline along the roadside and different crossroads. This could be the proof of an excessive manipulation of gasoline (successive transfer and/or evaporations).

In conclusion, Benin unleaded gasolines doesn't contain ETBE which increase the octane number of the gasoline (Dagaust, 2007) what promote a reduction of concentrations of aromatics (Nadim et al., 2001), carbon monoxide emissions (Noorman, 1993) and hydrocarbons (de Menezes et al., 2006). Moreover, benzene is found in high level in the gasoline. The same conclusions have been found in the gasoline of the border Benin-Nigeria. It confirms that Benin gets a stock in gasoline to Nigeria neighboring country. These results demonstrate that the ambient air in Cotonou contains high levels of benzene, carcinogenic compound by IARC (1974) which come as well as traffic exhaust that gasoline.

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