

# Sources Identification and Apportionment of Atmospheric Volatile Organic Compounds (VOCs) using PCA/APCS Receptor Model in an Urban Settlement of Benin City Southern, Nigeria

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**Abstract:** Ambient air Volatile Organic Compounds in Benin City was evaluated for their concentrations using active sampling methods, with the objective of determine the mass contributions of the different sources. Air samples were collected at a human breathing height of 1.5 meters and from nine locations comprising industrial, residential and commercial areas with heavy traffic on a four hourly basis using active sampler at flow rate of  $0.51\text{min}^{-1}$ . The sampling periods cover both dry and wet seasons. Collected samples were desorbed and analyzed for volatile organic compounds using gas chromatography with flame ionization detector. The concentrations of analytes were read from the calibration graph, which was done with standard solution prepared externally. The data of ambient VOCs analysis were subjected to statistical analysis; correlation analysis, factor analysis, principal component analysis/absolute principal component scores (PCA/APCS) model. The average concentrations of the ambient Volatile Organic Compounds (VOC) in the studied city exhibit higher concentrations in dry season months. The results obtained reveal negative or weak correlation coefficients for most Volatile Organic Compounds species, an indication of importance of other VOCs sources in addition to exhaust emission. The results from the PCA/APCS receptor modelling showed that during the dry season, the highest contribution of 53% comes from vehicular emission in the studied area. Meanwhile, evaporative emission contributes the highest during the wet season with 35% for the same locations. High correlation coefficient ( $R^2 > 0.8$ ) between the measured and predicted values suggested that PCA/APCS model is suitable for estimation of sources of VOCs in ambient air.

**Keywords:** Source Identification, Apportionment, Volatile Organic Compounds; Ambient air

## Introduction

The major aim of air pollution modeling is to give a more complete deterministic description of the air quality problem, including an analysis of factors and causes (emission sources, meteorological processes, and physical and chemical changes), and some guidance on the implementation of mitigation measures. In recent time, the use of air pollution models to study the mass contributions of pollutants' sources in the atmosphere of urban cities is becoming popular. Several methods and models have been applied to estimate the source contributions (Song *et al.*, 2001; Zhao *et al.*, 2006; Zhu *et al.*, 2001; Hoopke, 2003) to air pollution. Among the tools that have been used for such studies are chemical mass balance (CMB) (Hellen *et al.*, 2006; Olson *et al.*, 2009), principal component analysis (PCA)/absolute principal component scores (APCS) (Gou *et al.*, 2009, Guo *et al.*, 2006) positive matrix factorization (PMF) (Brown *et al.*, 2007; Sauvage *et al.*, 2009). Most of the studies revealed a significant correlation between the measured and predicted values. The PCA/APCS approach presents a source

apportionment technique which requires a minimum of inputs regarding source characteristics, but provides quantitative information regarding both source profiles and their impacts. Unfortunately, most of the works reported were carried out in America, Europe and Asia.

Air pollution studies in ambient urban environment of Nigeria have so far been rather limited to determination of atmospheric level and variation patterns. Although, various studies have been conducted by several authors at different urban and industrial centers in order to understand the air borne pollutants distribution (Obioh *et al.*, 1994; Baumbach *et al.*, 1995; Ekpo and Udontong, 2004; Ukpebor, *et al.*, 2005; Okuo and Ndiokwere, 2004; Abudl Raheem *et al.*, 2008). Several studies have also shown that air pollution associated to VOCs is threatening to human being and ecosystem in most Nigerian urban centres (Olumayede and Okuo, 2011, Okuo *et al.*, 2012; Ojiodu *et al.*, 2013). Ambient VOCs in urban centres are largely originated from ranges of industrial



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activities and specific sources such as motor vehicle exhaust, paints manufacturing, evaporative losses, petroleum storage/dispensing facilities, wood smoke emission and municipal waste sites (Wieslander *et al.*, 2007). Anthropogenic sources of air pollutants are also known to be prevalent in urban cities of the world (Lee *et al.*, 2002; Srivastava *et al.*, 2004; Barletta *et al.*, 2005). There is a need to better understand both the origin and mass contribution of each source.

Unfortunately, with high volumes of air pollution studies in Nigeria, only few studies have reported sources apportionment or determination of mass contributions of air pollutants in the country's atmosphere using model analysis. Among the few studies, Okuo and Ndiokwere (2004) employed the use of chemical mass balance to the study of sources identification of particulate matters. Recently, Olumayede *et al.*, (2013) employed principal component analysis to identify and apportion VOCs captured in the atmosphere of two urban centres of southwestern, Nigeria. In this study, the PCA/APCS receptor model revealed a high correlation between measured and predicted values.

Benin City is a commercially and industrially active capital city with a number of government parastatals, petroleum and allied industries. It serves as a transit city linking the western and eastern parts of the country through different road networks. Hence, it usually experiences huge traffic flow.

The City lies within the equatorial climatic belt (Af Koppen's climatic classification), which receive adequate rainfall of between 2000mm and 3000mm annually. Its mean monthly temperature and relative humidity are 28°C and 80% respectively. It is about 85 m above the sea level at the highest point with two distinct seasons (wet and dry). The City has a high

relative humidity and intense rainfall occurring between April and September. Meanwhile, ambient concentrations of volatile organic compounds have been reported to be on increase due to increase in the city population and economic activities (Olumayede and Okuo, 2011). There is need to identify and apportion the sources of volatile organic compounds in the city's atmosphere.

The aim of the present investigation was therefore to analyze environmental data gathered on the monitoring of ambient VOCs at nine sampling sites of urban settlement of Benin City, southern, Nigeria during dry and wet seasons. The work is intended to provide more comprehensive information about mass contributions of the identified sources to atmospheric VOCs within studied centres. The study is designed to create a solid basis for the development of air quality management plan in Nigeria cities. It is hoped that data provided would be useful to planners, policy makers, and environmentalists to determine the risk exposure to residential inhabitants and terrestrial life.

## Methodology

### The Study Area

The study was conducted in Benin City, Edo state, Southern Nigeria. Benin City is located between longitude 6.20°N and latitude 5.31°E. It is a city approximately 25 miles north of the Benin River and situated 200 miles by road east of Lagos; the commercial capital of Nigeria. It is one of the urban centers in Southern part of the country with about 1.3 million habitants (NPC, 2009). It is the administrative headquarter of Edo states; hence, an urban settlement with high population and steady traffic density especially during the weekdays. Figure 1 shows the various sampling sites in the studied centre of this work.

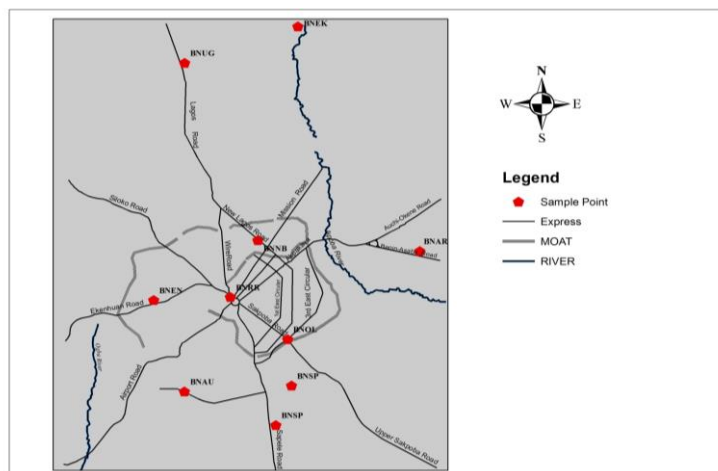


Figure 1: Map of Benin City showing the monitoring sites.

### **Ambient VOC collection and Sampling Routine**

Ambient VOCs collection, sampling devices and sampling routine have been described elsewhere (Olumayede and Okuo, 2011)

### **Extraction**

After sampling, adsorption tubes were extracted with 10ml carbon disulfide (CS<sub>2</sub>) as the extraction solvent as described in standard method (ASTM, 1988). The samples were centrifuged for another 15min, to obtain a clear phase at the top. The extracted samples were stored in a freezer temperature of 4± 1<sup>0</sup>C until they were analyzed.

### **VOCs Analysis and Quantification**

The extracted solutions were analyzed with gas chromatograph (GC; Aligent 6890N) equipped with a selective detector (Aligent 5973 inert Mass Selector Detector). The chromatograph column was HP-INNOWax, 30 × 25mm Internal Diameter × 0.25µm film and total chromatopack software. The carrier gas was helium at 0.1cm<sup>3</sup>/min at 2500C and 36cms<sup>-1</sup> linear velocity with a split ratio of 1:20. The inlet temperature was 2400C. Temperature program was: initial oven temperature 400C, hold for 3 minutes, 400C to 1200C at 50C/min, hold for 1min and the final hold is 15 minutes. Compounds were identified based on their retention times (within ±0.05 minutes of the retention time of calibration standard). The concentrations of the identified compounds were read from the calibration graph, which was done with standard solution prepared externally. The standard is a Volatile Organic Calibration Mix containing 54 VOCs at 2000mg/l in Methanol (Supelco, Bellefonte, U.S.A.). The standard solution was prepared by dilution in CS<sub>2</sub>/methanol for gas chromatography. Ten calibration levels of concentration range of 0.02µg/ml - 20.0µg/ml were prepared from stock standard with CS<sub>2</sub> in a clean vial. They were freshly prepared at the moment of calibration.

The instrumental calibration was performed by analyzing 1µl of the diluted standards. The calibration curve shows good linearity, with determination coefficients (r<sup>2</sup>) greater than 0.999 for all the compounds. The lowest calibration level for each compound was taken as the instrumental quantification limit for that compound.

The quality control includes the determination of instrument repeatability and analyzes of the blank activated carbon tubes, as process blank. The extraction solvent (CS<sub>2</sub>) was also analyzed to determine if there were any contaminations. None of the compounds included in this study were detected in CS<sub>2</sub> and in process blanks. Back – up sections of adsorbent tubes were also extracted and analyzed. VOCs amounts in the back – up sections were below

the detection limits indicating that breakthrough was not a problem. No internal standard was used because of good repeatability shown by the method.

### **Statistical Analysis**

#### **Source analysis and apportionment**

Investigations of the relationships among the VOCs species were made using correlation matrices. Correlation analysis helps reveal the possible source of VOCs (Wang *et al.*, 2002; Na and Kim, 2001). For this purpose, the Pearson's correlations (two-tailed) of the obtained data was evaluated. Correlation coefficient (r<sup>2</sup>) were evaluated into three correlation ranges; positive (0.8 < r<sup>2</sup> < 1.0), moderate (0.5 < r<sup>2</sup> < 0.80), and negative (r<sup>2</sup> < 0.5).

The PCA analysis was applied to the data obtained in all the sampling sites of this study. We followed the PCA method of factor analysis using SPSS Software package (Version 15). The Kaiser's criteria were adopted to decide the appropriate numbers of factors to be retained (only factors with eigenvalues > 1). The results of analysis of the concentrations were obtained and the absolute principal component scores (APCS) calculated based on the PCA identified which was used for source quantification. In the PCA/APCS techniques, the absolute factor score obtained from the PCA analysis are regressed on the observed VOC concentrations to apportion individual VOCs to each source.

### **Results and Discussion**

#### **General characteristic and ambient concentrations of abundant VOCs**

In our previous report (Olumayede and Okuo, 2011), a total of sixteen volatile organic compounds species which were successfully identified and quantified in ambient air of Benin City, among which were five aliphatics, 6 aromatic compounds, 4 halogenated hydrocarbons and 1 Carbonyl. These compounds were used in this study.

#### **Source identification of VOCs**

The major objectives of this work are to identify the sources contributing to VOCs in the areas under study. Therefore, the VOCs data obtained were subjected to correlation coefficient matrix, factor analysis and cluster analysis.

#### **Correlation matrix**

Studies have shown that the major source of aromatic hydrocarbons included gasoline evaporation, solvent evaporation and diffuse emissions and vehicular emissions (Heeb *et al.*, 2000). The results of the correlation coefficient matrix of VOC species are presented in Tables 1-2. As can be seen in the tables, both negative and positive significant correlations exist between benzene, toluene, ethylbenzene, xylenes (BTEX) (indicator of organic compound

from road traffic (Hoque *et al.*, 2008) and other VOC species during dry and wet seasons in the two studied centres. The positive correlation coefficients observed among these species support the hypothesis that vehicular related emission is the major source impacting the atmosphere of the studied centres. The

finding indicates that there might be an exit for the extra sources of benzene, such as evaporation and other forms of combustion. This agrees well with finding of Monod *et al.*, 2001 and Barletta *et al.*, 2005 in atmosphere of Beijing.

**Table 1:** Correlation Coefficient of VOC species in Benin City during Wet Season

	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16
3- Methylpentane (1)	1	-0.436 <sup>*</sup>	-	-	0.15 <sup>*</sup>	0.398	0.228	-0.324 <sup>*</sup>	0.010	0.599	0.163	0.217	-0.374 <sup>*</sup>	-0.180	-0.162	0.078
Isopentane (2)		1	0.104	0.137	0.17 <sup>*</sup>	0.243	0.276	0.579 <sup>*</sup>	0.141	-0.115	-0.117	-0.859	0.156	0.250	0.587	-0.11
Butane (3)			1	0.230	0.15	0.109	0.273	0.066	0.151	0.127	-0.029	-0.253 <sup>*</sup>	0.141	-0.164	0.355	-0.22
2,2 dimethylbutane (4)				1	0.29 <sup>*</sup>	0.365 <sup>*</sup>	0.514	0.684	0.680	-0.125	-0.267	-0.126	0.819	0.740	0.040	-0.36
Undecane (5)					1	0.095	0.166	0.233	0.033	0.226	-0.122	0.300	0.407 <sup>*</sup>	-0.314	0.141	-0.33
Benzene (6)						1	0.114	0.069	0.190	0.434	-0.517	-0.167	0.669 <sup>*</sup>	-0.704	0.510	-0.41
Toluene (7)							1	0.065	0.925	0.686	-0.401	-0.087	0.355	0.407	0.374	-0.18
Ethylbenzene (8)								1	0.217	-0.144	-0.450	-0.540	0.486	0.470	0.040	-0.59
(p,m)-Xylene (9)									1	0.526	-0.404	0.0742 <sup>*</sup>	0.579	0.623	0.320	-0.20
o Xylene (10)										1	-0.440	-0.290	0.289 <sup>*</sup>	-0.090	0.285	-0.25
1,2,4-Trimethylbenzene (11)											1	0.424	0.077	0.134	0.654	0.743
Methylene Chloride (12)												1	0.003	-0.111	0.314	0.277
1,2-dichloroethane(13)													1	0.948	0.075	0.167
Carbon tetrachloride (14)														1	0.209	0.187
Chloroform (15)															1	0.327
Acetone (16)																1

\*\* . Correlation is significant at the 0.01 level (2-tailed).

\* . Correlation is significant at the 0.05 level (2-tailed).

**Table 2:** Correlation Coefficient of VOC species in Benin City during dry Season

	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16
3- Methylpentane (11)	0.211	-0.29	-0.51	0.197	-0.023	-0.125	-0.156	-0.211	0.565	0.169	-0.195	-0.758	-0.58	0.152	-0.12	
Isopentane (2)		1	-0.218	-0.156	-0.176	-0.127	-0.375	0.464	-0.348	-0.288	-0.196	-0.213	-0.298	-0.424	-0.236	-0.193
Butane (3)			1	-0.300	0.672	0.820	0.178	-0.148	-0.511	-0.589	-0.261	-0.036	0.085	0.219	-0.025	-0.41
2,2 dimethylbutane (4)				1	-0.414	-0.376	-0.106	0.689	0.839	-0.113	-0.304	0.173	0.554	0.245	-0.314	-0.21
Undecane (5)					1	0.946	-0.194	-0.215	-0.388	-0.301	-0.259	0.054	-0.133	0.289	-0.184	-0.46
Benzene (6)						1	-0.169	-0.135	-0.449	-0.428	-0.215	-0.024	-0.004	0.364	-0.087	-0.42
Toluene (7)							1	-0.339	0.122	0.047	-0.187	-0.541	0.400	0.167	-0.073	0.278
Ethylbenzene (8)								1	0.433	-0.270	-0.434	-0.145	0.175	-0.128	-0.345	-0.567
(p,m)-Xylene (9)									1	0.299	-0.159	-0.093	0.572	0.354	-0.219	0.060
o Xylene (10)										1	0.704	-0.175	-0.299	-0.216	0.635	0.514
1,2,4-Trimethylbenz (11)											1	0.097	-0.323	-0.144	0.939	0.680
Methylene Chloride												1	-0.270	-0.124	-0.032	-0.15
1,2-dichloroethane													1	0.840	-0.336	0.220
Carbon tetrachloride														1	-0.21	0.269
Chloroform (15)															1	0.511
Acetone(16)																1

\*. Correlation is significant at the 0.05 level (2-tailed).

\*\*. Correlation is significant at the 0.01 level (2-tailed).

**Principal Component Analysis (PCA)**

The results of PCA analysis for wet and dry seasons in the studied centre are summarized in Tables 3-4. The PCA of the variability in wet season concentrations were dominated by six factors that

together accounted for 90.82% of the total variance between concentrations (Table 3). The two strongest factors accounted for 52.38% of the variance. Four other factors accounted for 17.41%, 10.66%, 8% and 7.37% of the variance respectively.

**Table 3:** Factor loadings estimated by PCA analysis at Benin City during Wet Season

	Component					
	1	2	3	4	5	6
3- Methylpentane	-.313	.159	-.200	.721	-.354	.018
Isopentane	.096	.016	.963	-.201	.022	.008
Butane	-.040	-.018	.114	.039	.984	-.071
2,2 dimethylbutane	.874	-.327	.062	-.128	.233	-.111
Undecane	-.209	-.132	-.160	.078	-.083	.947
Benzene	.660	-.561	-.091	.313	.034	-.355
Toluene	.519	-.269	-.197	.714	.280	-.169
Ethylbenzene	.467	-.525	.567	-.235	-.153	-.097
(P,m) –Xylene	.753	-.266	-.130	.538	.190	.036
o Xylene	-.092	-.238	.077	.949	.091	.124
1,2,4-Trimethylbenzene	-.035	.000	-.136	-.202	-.018	.932
Methylene Chloride	.008	.907	-.236	-.173	-.143	.210
1,2 –dichloroethane	.956	.092	.036	-.156	-.114	-.199
Carbon tetrachloride	.204	.941	.188	.062	-.160	-.081
Chloroform	.020	.685	-.200	.582	.299	.104
Acetone	.008	.860	-.180	-.010	-.182	-.296
% variance	28.73	23.65	17.41	10.66	8.00	7.37
Possible source						
	Exhaust emissions	Emission from Waste dumpsite	Gasoline Evaporative emissions	Industrial solvent/ Solvent usage	LPG	Biomass burning/Bio genic

Extraction Method: Principal Component Analysis.  
Rotation Method: Varimax with Kaiser Normalization.

The strongest factor (factor 1) accounted alone for 28.73% of the variance and was highly correlated with 3-methylpentane, undecane, 2,2 – dimethylbutane, toluene, benzene, Xylenes, trimethylbenzene. The VOCs associated with factor 1 were typical for motor vehicle (both gasoline and diesel powered) emissions (Chan *et al.*, 1991 Weisel

*et al.*, 1992, Wixtrom and Brown, 1992, Watson *et al.*, 2001).

The second strongest factor (factor 2) has cumulative variance of 52.38% together with factor 1. Factor 2 accounted for 23.65% of the variance in VOC concentrations and was identified as acetone

emissions from waste disposal (Environmental Australia, 2001).

Factor 3 correlated strongly with butane, isopentane, benzene, ethylbenzene. However, as the more volatile compounds of traffic emissions were strongly present in factor 3 it was concluded that this factor represented more recent traffic emissions.

Furthermore, factor 4 accounted for 10% of the variance and had strong association with toluene, ethylbenzene, xylenes and along with the halogenated hydrocarbon (chloroform). It is well known that toluene, ethylbenzene, and xylenes have large portion in solvents (Brocco *et al.*, 1997). Furthermore, halogenated hydrocarbons are used as aerosol and paints. Therefore, the source might be attributed to solvent usage.

High loading of butane in factor 5 implied the factor is petroleum products (LPG). N-butane has been used as marker of Gas leakage. Factor 6 is highly loading in undecane and 1, 2-dimethylbenzene. This source is attributed to biomass emission. However, the presence of volatile alkane (undecane) in this factor indicating additional emissions from indoor sources

such as cleaning products, fragrances, consumer products, particleboard etc.

The variability in dry season VOC concentrations was dominated by five factors that together accounted for 72% of the total variance (Table 7).

Factor 1 correlated highly with the VOC study compounds isopentane, 3-methylpentane, 2,2-dimethylbutane, benzene along with Xylenes. All of these compounds are indicator of gasoline Vehicular exhaust. Previous studies support this identification (Ulman and Chilmonezyk, 2007; Knöppel and Schauenburg, 1989, Vega *et al.*, 2001, Wallace, 1991, Fellin and Otson, 1994, Cooper *et al.*, 1995). Factor 2 correlated highly with toluene, methylene chloride, o-Xylene and Chloroform, and was identified as Solvent Usage. Factor 3 was highly loaded in butane and was also identified as emission from liquidified petroleum gas (LPG) leakage. Factor 4 is highly loaded in Acetone. This may be attributed to emission from waste dumpsite. However, high loading in 123- trimethylbenzene and in Factor 5 is an indication of emission from diesel source. The emitters are likely to be diesel trucks coming from Warri.

**Table 4:** Factor loadings estimated by PCA analysis at Benin City during dry season

	Component				
	1	2	3	4	5
3- Methylpentane	.071	.119	.017	.017	.207
Isopentane	.725	.215	-.470	-.248	.219
Butane	-.374	-.207	.748	.139	-.001
2,2 dimethylbutane	.698	-.287	-.329	-.276	-.203
Undecane	-.047	-.143	-.058	.076	.959
Benzene	.977	-.079	-.170	-.038	.013
Toluene	-.190	.707	-.058	-.195	.561
Ethylbenzene	-.141	-.403	.364	-.783	.089
pm -Xylene	.906	-.100	-.332	.007	.092
o Xylene	-.263	.681	.408	.205	.169
1,2,4-Trimethylbenzene	-.141	-.123	-.080	.048	.968
Methylene Chloride	-.018	-.022	-.014	.756	-.993
1,2 -dichloroethane	.857	-.174	.356	.125	.290
Carbon tetrachloride	.361	.060	.321	.197	.163

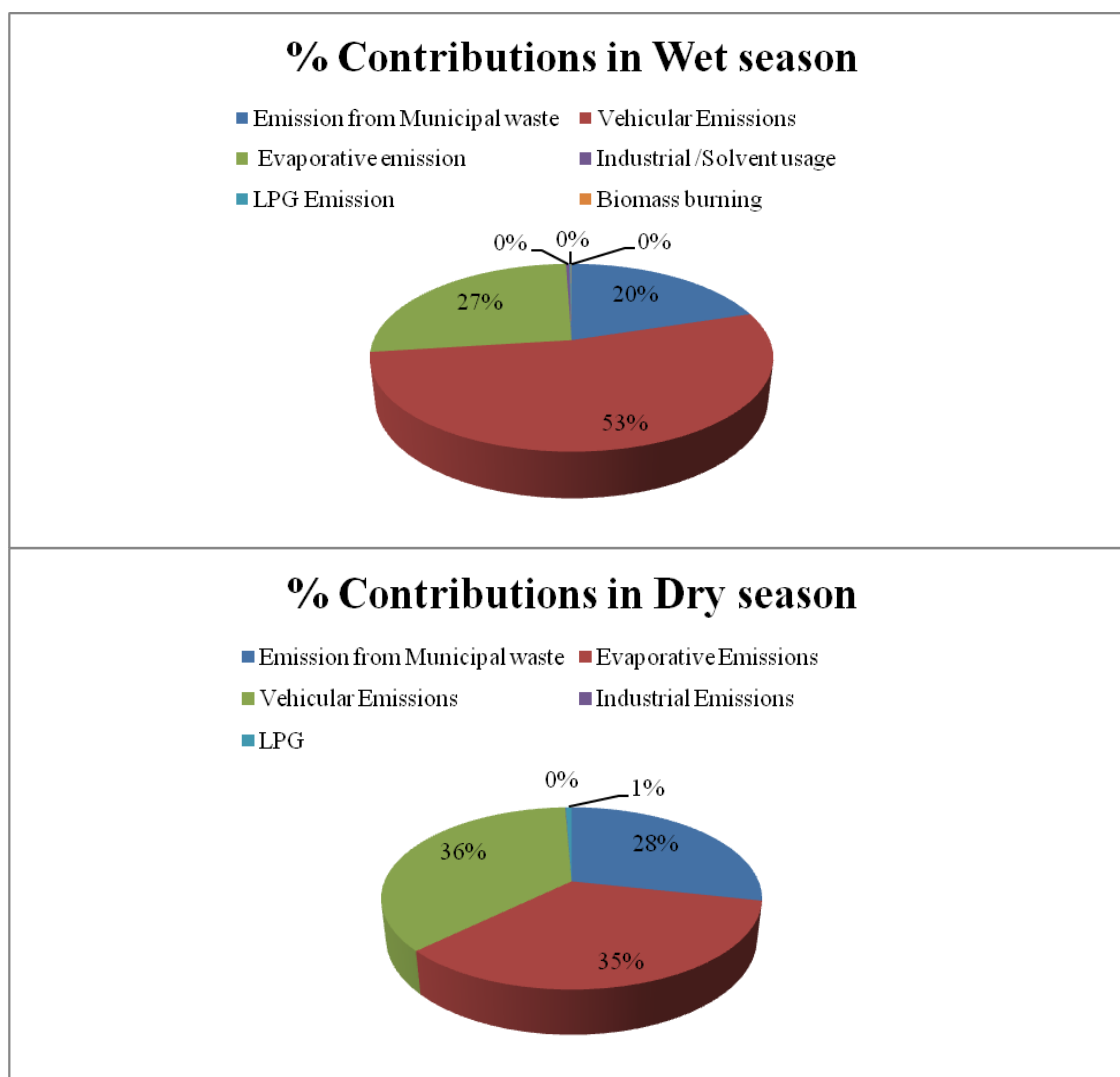
Chloroform	-0.014	.906	-.111	.032	-.025
Acetone	-.423	351	-.106	.698	.194
% Variance	40.68	24.35	10.66	8.09	6.43
Possible Sources	Gasoline / Exhaust emissions	indoor sources/ Solvent usage	LPG	Emissions from Municipal Waste dumpsite	Biomass /Diesel combustion

Extraction Method: Principal Component Analysis.  
Rotation Method: Varimax with Kaiser Normalization.

### Source contributions

The seasonal apportionment of VOCs to sources at the studied centres is presented as shown in Figures 3a and 3b. The average contribution of vehicle related

source was highest in dry season and lowest in wet season in all the Centres, probably due to the fact that traffic is the main mode of transportation in Nigerian urban centres.



**Figure 3:** Seasonal Trends of Contributions of Source factors in the VOCs Samples in Benin City



## Conclusions

In this study we present the data on the measurements of VOCs in the urban center of Benin City, southern, Nigeria over the dry and wet seasons of 2009 to 2010. Sixteen species of VOCs were detected in all the sampling sites of the center. From the study, vehicle exhaust emission from petrol driven light duty vehicles are the greatest contributor in all centre. Although other sources such as evaporative emission; industrial solvent and waste dump make contributions but the highest come from vehicular emission.

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