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Multivariate Statistical Study of Spatial Patterns of Volatile Organic Compounds in an Urban Atmosphere in Nigeria

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ABSTRACT

In complex environment of an urban atmosphere, variability in concentrations of pollutants from site to site within a city is often observed, hence there is need to investigate the quality parameters responsible for the variations. For this purpose, ambient VOCs (volatile organic compounds) measured on four hourly bases at different days of the week and at six days interval in Benin City, southern Nigeria were collected (from June 2009 to May 2010) at nine different microenvironments, selected to represent local activities in the city. The samples were collected at the breathing zone of 1.5meter height, extracted with carbon disulphide and were analyzed using gas chromatographic system. A total of 15 VOCs were captured during sampling period at nine urban sites and among the VOCs species detected are four alkanes, six aromatic compounds, four chlorinated hydrocarbons and one ketone. To determine the patterns of VOCs, multivariate analysis of variance (MANOVA) and factor analysis techniques were applied for evaluation of spatial variation and interpretation of complexity of data in the urban atmosphere. The result showed that the nine sites are significantly different in terms of total VOC concentrations. Factor analysis (FA) results revealed that six factors explained 95.00% of the total variance while spatial Discriminate Analysis (DA) showed that seven discriminate function (DFs) were found to discriminate the nine sites. Wilk"s Lambada test showed that only the first two functions are statistically significant. The present study shows the usefulness of multivariate statistical techniques for analysis and interpretation of complex data sets, and identifies probable source components in order to explain the atmospheric behaviors of pollutants.

Key words: Volatile Organic Compounds, MANOVA, Factor analysis, urban atmosphere

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INTRODUCTION

Assessment of variability in concentrations of pollutants from site to site within a city is of important interest for air toxics data analysis. The aim of such analysis is to understand how representative a given site is with respect to air toxics concentrations in a city. Understanding the variability of pollutants is an important factor in determining human exposure to the chemicals. Recently, the use of environmetric techniques, especially multivariate analysis, to study the variation of pollutants in the atmosphere of urban cities and drawing meaningful information is becoming popular [1-3]. One common approach is the use of multivariate techniques to pattern the spatial and temporal variation of the pollutant concentrations. Multivariate analysis offers techniques for classifying relationships among measured variables. The two most common multivariate analyses are principal components analysis (PCA) and factor analysis (FA). Notable studies of such approach in environmental chemistry include evaluation of spatial and temporal variations in river waters [4-7] and in air quality [8 - 10]. In most of these studies, it was concluded that application of environmetric methods revealed adequately information of the spatial variability of the large and complex quality data. Unfortunately, most of the works reported were carried out in America, Europe and Asia.

VOCs are carbon-based compounds that have vapor pressure to significantly vaporize and enter the atmosphere [11, 12]. Ambient VOCs in air are largely originate from mobile and industrial sources, including industrial operations, vehicle emissions, and the use of all kinds of artificial panels such as plywood, laminated wood floor by some furniture industries [13-15], hence they are commonly encountered by people as they go about their daily routine. Meanwhile, ambient concentrations of volatile organic compounds have been reported to be on increase in many cities of the world [16-20] and some Nigerian urban centers [21- 23]. In Nigeria, concentrations of total VOCs up to 26.42± 6.82µgcm^{-3} have been reported by Olumayede and Okuo [21] in atmosphere of urban centers of Benin City, Akure and Ado – Ekiti. Similarly, Okuo et al [23] reported that concentration up to 919.01± 19.48µgcm^{-3} was observed in a polluted environment of Lagos, Nigeria. Recent epidemiological studies have strengthened the evidence that there are short-term VOCs effects on mortality and respiratory morbidity [24]. There is also growing evidence that chronic exposure to VOCs have adverse health effects to human [25 - 26]. The short term adverse effects include conjunctive irritation, nose and throat discomfort, headache and sleeplessness, allergic skin reaction, nausea, fatigue and dizziness. While the long term adverse effects include loss of coordination, leukaemia, anaemia, cancer and damage to liver, kidney and central nervous system[27,28].

Air quality studies have been reported in most urban centers of Nigeria [29- 32]. Most of these studies reported that the mean levels of most pollutants showed significant spatial and seasonal variations. Unfortunately, of all the air pollution studies in Nigeria, only few studies have employed the use of multivariate approach to account for the variability of pollutants in the cities atmosphere. Although, Okuo and Ndiokwere[31] applied factor analysis to the study of sources identification, but it was not used in explaining the pollutant trends. Only Abdul-Raheem et al [32] employed combination of multivariate statistical methods to evaluate the influence of seasons on the concentrations of ozone, sulfur (IV) oxide, and oxide of nitrogen in ambient air of two Nigerian cities of Lagos and Ilorin. In the study, the PCA revealed that three distinct groupings during the day for all data, which is a reflection of different factors contributing to the atmospheric chemistry of these cities.

In the present study, we applied multivariate analysis to captured VOCs data of nine sampling sites in Benin City, Nigeria, to characterize the patterns of the Volatile organic Compounds pollution. The principal components and discriminate analyses were applied to understand the source of spatial pattern of VOCs species and to identify the various anthropogenic activities associated with the variation. The aim of such analysis is to understand how representative a given site is with respect to air toxics concentrations in a city.

MATERIALS AND METHODS

Sampling Locations

Benin City, southern Nigeria, is located between longitude 6.20^{0}N and latitude 5.31^{0}E. It is situated within the equatorial climatic belt (Af Koppen‟s climatic classification) [35] and is one of the urban centers in Southern part of the country with about 1.3 million habitants [36]. It is the administrative headquarter of Edo states; hence, an urban residential area with high population and steady traffic density especially during the weekdays. The City lies within such areas which receive adequate rainfall of between 2000mm and 3000mm annually. Its mean monthly temperature and relative humidity are 28°C and 80% respectively. Benin City is about 85 m above the sea level at the highest point [37].

Sampling Device and Routine

This study involves a field study which was carried out between May, 2010 and June, 2011 at nine selected sites within the City (Figure 1). The study sites were carefully chosen to reflect the different pattern of human activities dominating in the area. The detail descriptions of sampling device, sampling sites and routine procedure have been discussed elsewhere [22]

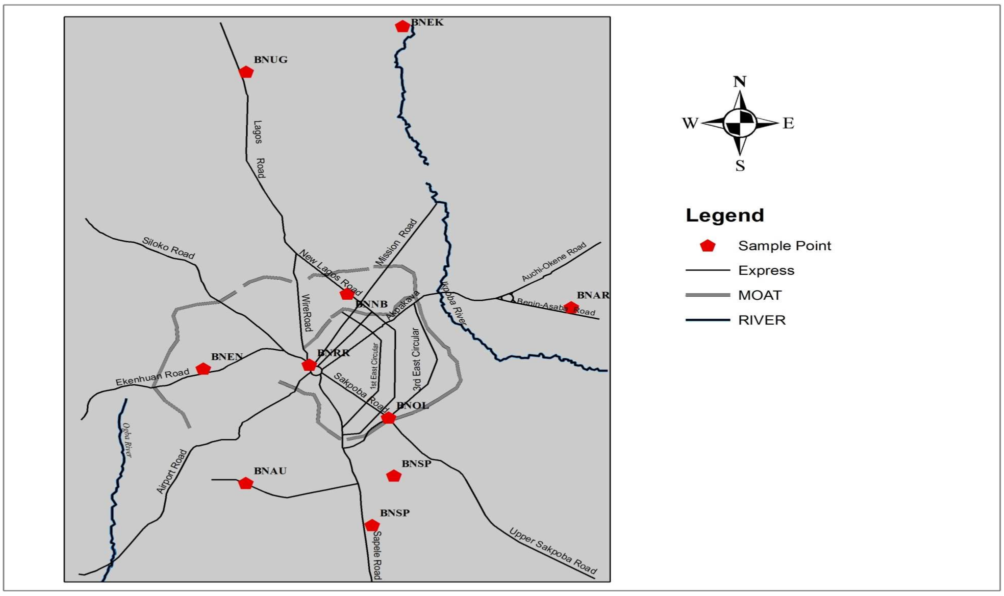

Figure 1: Map of Benin City showing the nine monitoring sites

Extraction and Analysis of VOCs

After sampling, adsorption tubes were extracted with 10ml carbon disulfide (CS2) as described in standard method (ASTM, 1988) with slight modification as described elsewhere [23]. 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± 1^0C until they were analyzed. The quantification and quality control processes have also been described earlier [22].

Data analysis

The raw data on ambient VOCs captured in the city atmosphere were taken through multivariate analysis of variance (MANOVA). MANOVA was used where several dependent variables were measured for each sampling unit instead of one variable. The objective of MANOVA was to investigate whether the mean vectors of several groups were the same, and if not, which means the variables were differed significantly from group to group. Principal Component analysis (PCA) and discriminate analysis (DA) are multivariate tools for establishing cluster of data was used to measure and compare differences in the sites. Discriminate analysis is used to describe or elucidate the differences between two or more groups and identifying the relative contribution of all variable to separation of the groups. Second aspect is prediction or allocation of observations to group in which linear or quadratic functions of the variable (classification functions (CFs)) are used to assign an observation to one of the groups [38, 39]. All data analyses were done with SPSS 13 software [40].

RESULTS AND DISCUSSION

Ambient levels of abundant VOCs

In our earlier report [22], a total of fifteen volatile organic compounds species were successfully identified and quantified in ambient air of Benin City, among which were four alkanes, 6 aromatic compounds, 4 chlorinated hydrocarbons and 1ketone. Figure 2 presents the contributions of each species to the total VOCs at the various sampling sites. As can be seen, aromatic group of VOCs has the highest total mass contribution (between 10% and 86%) in all the sampling sites. This was followed by aliphatics, with mass contribution in range of 0% and 9%. These observations have earlier been explained and have been attributed to factors such as economic activities, poor conditions emitter and proximities to emission sources [22, 23].

Patterns Recognition

According to Baumbach [24] factors responsible for spatial variation in urban air pollution which could also be responsible for VOCs distribution in this study include; emission rate, emission strength, emission conditions and atmospheric dispersion. To investigate these assumptions, we pattern the sites and VOCs species using multivariate approaches.

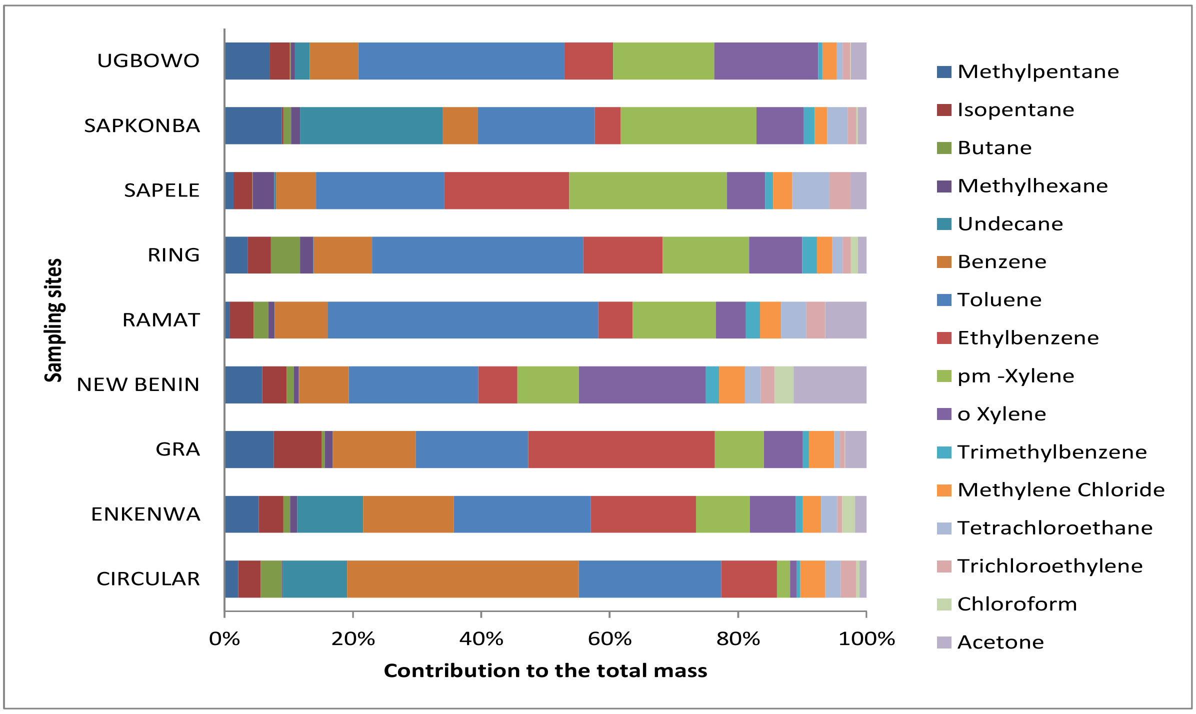

Figure 2: Plot of the percentage contribution of each VOC capture to the total VOCs at the various sampling sites.

Recognition of Spatial Patterns

The results of multivariate analysis of variance (MANOVA) are presented in (Table 3). According to obtained data, the nine sites are significantly different in terms of mean VOCs. This indicates that the quality status of air in the site, emission strength and fate of the VOCs are different between the sites.

Table 3. Multivariate test (MANOVA) for VOCs in the nine sampled sites of Benin City

Effect		Value	F	Hypothesis df	Error df	Sig.
Intercept	Pillai's Trace	.768	2.580(a)	9.000	7.000	.112
	Wilks' Lambda	.232	2.580(a)	9.000	7.000	.112
	Hotelling's Trace	3.317	2.580(a)	9.000	7.000	.112
	Roy's Largest Root	3.317	2.580(a)	9.000	7.000	.112

a Exact statistic

b Design: Intercept

Principal component analysis of sites

Understanding of the cluster pattern of the VOCs captured in each site of this study will help gain further insight into the behaviors of the pollutants so as to measure the quality status of the sites. The Hierarchical Cluster Analysis (HACA) was performed on the VOCs data collected to evaluate spatial variation among the sampling sites. This analysis resulted in the grouping of sampling stations into three clusters/groups (Fig. 3).

Cluster 1 (Enkenwa, GRA and Sapele Road) represents the light traffic frequency areas, other sources of pollution in these areas include emission from residential houses and waste dumpsites in the areas. Cluster 2 (Ramat, Ring, Sakponmba Junction and Ugbowo) represents the moderate traffic frequency areas and pollution sources in these sites include evaporation of petroleum products from dispensing stations and small- scale industries located close to these sites. Cluster 3 (New Benin and circular road) represents dense traffic (bus stops) areas with other possible pollution sources from markets around the bus stop.

This result shows that traffic frequency is the dominant factor affecting spatial variation of pollutants in this city. In addition, poor dispersion of pollutants due to clustering of buildings in the city might play a significant role in the spatial variation. This observation agrees well with the earlier reports of Ukpebor et al [30] on another pollutant (CO). Furthermore, the result implies that for rapid assessment of atmospheric quality, only one station in each cluster is needed to represent a reasonably accurate spatial assessment of the VOCs atmospheric pollution for the whole network.

Factor analysis of the captured VOC species

Table 4 presents the result of factor analysis of the data carried out to identify the sources of the variation. Table 4 summarized the eigen-value, percentage of variance accounted and the component loading. The result shows that six factors were extracted and explained 95.00% of the total variance.

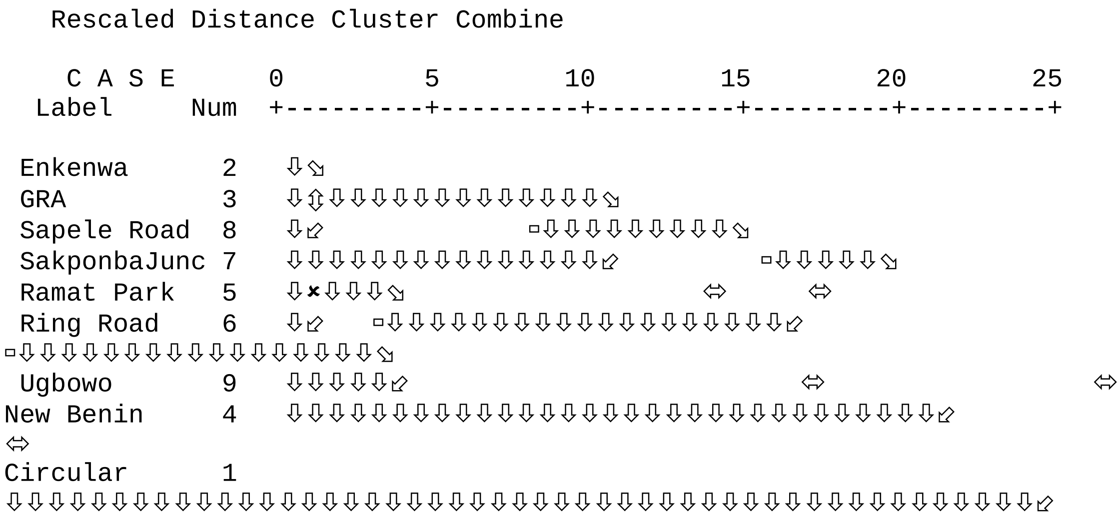

Figure 3: Result of Hierarchical Cluster Analysis of sampling sites

TABLE 4: Summary of the eigen-value, percentage of variance accounted and the component loading

Component	Initial Eigenvalues			Extraction Sums of Squared Loadings		
	Total	% of Variance	Cumulative %	Total	% of Variance	Cumulative %
1	4.624	28.900	28.900	4.624	28.900	28.900
2	2.953	18.454	47.353	2.953	18.454	47.353
3	2.447	15.296	62.649	2.447	15.296	62.649
4	2.261	14.131	76.780	2.261	14.131	76.780
5	1.818	11.360	88.140	1.818	11.360	88.140
6	1.098	6.861	95.001	1.098	6.861	95.001

Extraction Method: Principal Component Analysis.

The first (F1), second (F2), third (F3), fourth (F4), fifth (F5) and sixth (F6) factors accounted for 28.9, 18.55, 15.29, 14.13, 11.36 and 6.86 % respectively of the total variance.

F1: This factor is highly loaded in methylhexane, ethylbenzene and p,m -xylene. VOCs like these compounds have been associated with petrol powered vehicle emission [41]. Therefore, factor 1 most likely corresponds to emission from vehicular source.

F2: Factor 2 is loaded in methylpentane, undecane and p,m -xylene. These chemical have been associated with combustion of diesel related compounds [42]. Factor 2 is thus an indication of emission from heavy vehicle transportation.

F3: this is highly loaded in O-xylene, chloroform and acetone. These chemicals are associated with contribution by uncontrolled solvent usage

F4: Factor 4 is loaded in methylene Chloride, tetrachloroethane and 1, 1, 1 trichloroethylene. The chemicals are also associated chemicals which are contributed by anthropogenic activities of solvents from refrigeration repair shops in the area.

F5: Toluene is highly loaded in this factor. This compound is commonly use as solvent in paints hence F5 is likely solvent loss from paints factory or from painted buildings.

F6: F6 is loaded in butane and trimethylbenzene.

Table 5: Result of factor analysis for ambient VOCs

	Component					
	1	2	3	4	5	6
Methylpentane	.055	.877	.158	-.369	-.058	-.075
Isopentane	-.216	-.872	-.114	-.272	-.222	-.087
Butane	-.118	-.009	-.216	.040	.207	.935
Methylhexane	.982	.012	-.107	.017	-.027	.059
Undecane	.081	.960	-.188	.072	-.046	.097
Benzene	-.559	.049	-.608	.441	-.128	.203
Toluene	.149	.043	.047	-.125	.920	.323
Ethylbenzene	.498	-.435	-.429	-.282	-.480	-.238
P,m-xylene	.793	.522	.062	-.007	.268	-.097
O-xylene	.033	.298	.786	-.275	.132	-.138
Trimethylbenzene	.531	.309	.328	.014	.239	.627
Methylene Chloride	-.085	.006	.030	.921	-.245	.051
Tetrachloroethane	.770	.233	-.077	.539	.096	-.062
111-Trichloroetylene	.507	-.023	-.006	.796	.301	-.028
Chloroform	-.239	-.007	.642	-.016	-.494	.424
Acetone	-.090	-.196	.888	.340	-.003	-.063

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

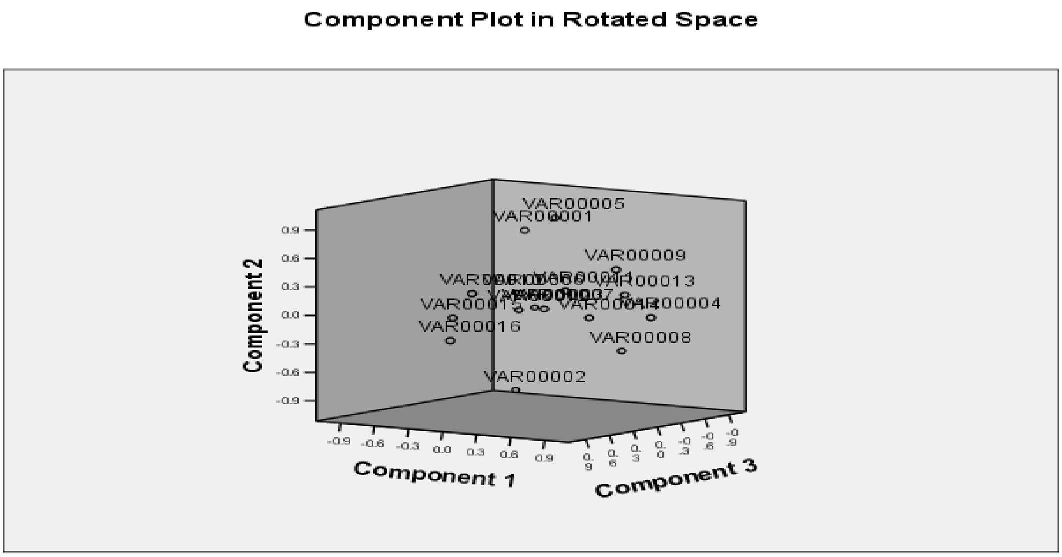

Figure 4: Component plot in rotate space

Key to the rotated component variables in Figure 4

1. 3-methylpentane 3. 22-methylbutane 5. Undecane 7. Toluene
2. Butane 4. Isopentane 6. Benzene 8. Ethylbezene
9. pm – Xylene 10. O -xylene 11. 1,2, 4 – Trimethylbenzne
12. Methylene Chloride 13. 1,2 –dichloroethane 14. 1,1,1- trichloroethyne
15. Chloroform 16. Acetone.

Discriminate analysis of the captured VOC species
Discriminate analysis (DA) was also applied on the raw data of volatile organic compounds species to further evaluate the spatial variation. Two discriminate functions (DFs) were found to discriminate the nine sites as shown in Tables 6 and 7. Wilk's Lambada test showed that only the first two functions are statistically significant (Table 6). The first DFs explained 87.6% of the total spatial variance, and the second DFs explained 10.3%. The relative contribution of each parameter is given in Table8. BTEX exhibited strong contribution in discriminating the nine sites and account for most of the expected spatial variation, while less contribution exhibited from other VOCs.

Table 5: Eigenvalues

Function	Eigenvalue	% of Variance	Cumulative %	Canonical Correlation
1	.392(a)	89.7	89.7	.531
2	.045(a)	10.3	100.0	.208

a First 2 canonical discriminant functions were used in the analysis.

Table 6: Wilks' Lambda

Test of Function(s)	Wilks' Lambda	Chi-square	df	Sig.
1 through 2	.687	1.500	10	.999
2	.957	.177	4	.996

Table 7: Standardized Canonical Discriminant Function Coefficients

	Function	
	1	2
Undecane	5.887	.867
o-xylene	25.913	.792
11-dChloroethene	-12.269	.526
Chloroform	-10.941	-1.451
Acetone	-7.021	.221

Table 8: Structure Matrix

	Function	
	1	2
Toluene(a)	-.932(*)	-.004
Isopentane(a)	-.694(*)	-.263
124-Trimethylbenzene(a)	-.662(*)	.001
Ethylbenzene(a)	.562(*)	.311
Methylene Chloride(a)	.398(*)	.115
22-dimethybutane(a)	.325(*)	-.035
o-xylene	.028(*)	.009
11-dChloroethene	-.027	.522(*)
pm-xylene(a)	-.017	.521(*)
Chloroform	.005	-.511(*)
111-trichloroethylene(a)	.118	.502(*)
2-methylpentane(a)	.118	.413(*)
Benzene(a)	.056	-.165(*)
Acetone	-.001	-.086(*)
Undecane	-.002	-.004(*)

Pooled within-groups correlations between discriminating variables and standardized canonical discriminant functions
Variables ordered by absolute size of correlation within function.
** Largest absolute correlation between each variable and any discriminant function*

CONCLUSION

In this study, multivariate statistical techniques were used to evaluate spatial variation in ambient VOCs of Benin City. Discriminate analysis gave the best result for spatial variability and identified only two parameters to discriminate between nine sites with 92.7% correct assignations. Therefore, discriminate analysis helped to identify and understanding the source of spatial variation. Thus, the application of multivariate statistical techniques has been proved to be an effective tool for analyzing a huge and complex environmental data matrix.

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* * * * * * H I E R A R C H I C A L   C L U S T E R   A N A L Y S I S * * * * * *


 Dendrogram using Average Linkage (Between Groups)

                        Rescaled Distance Cluster Combine

    C A S E      0         5        10        15        20        25
  Label     Num  +---------+---------+---------+---------+---------+

  Methylhexane        4   ⇩⇘
124 trimethyl    11   ⇩▫
 111,trichloro     4  ⇩▫
  Chloroform     15   ⇩▫
  Butane          3   ⇩▫
  Isopentane      2  ⇩▫
 Methylchoride    12  ⇩⇧⇩⇘
12dichloroethen   13  ⇩▫ ▫⇩⇩⇩⇘
  Acetone        16   ⇩⇙ ⇔    ▫⇩⇩⇩⇘
  Methylpentane   1  ⇩⇩⇩⇙    ⇔   ▫⇩⇘
  0-Xylene       10   ⇩⇩⇩⇩⇩⇩⇩⇩⇙   ⇔ ▫⇩⇩⇩⇩⇩⇘
  Undecane        5   ⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇙ ⇔     ⇔
  Benzene         6   ⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩×⇩⇙     ▫⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇘
  Ethylbenzene    8   ⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇙      ⇔                       ⇔
  Pm-Xylene       9   ⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇙                       ⇔
  Toluene         7   ⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇩⇙
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Figure 5: Result of Hierarchical Cluster Analysis of sampled VOCs

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