

Distribution and Behaviour of Four Polyaromatic Compounds in Oil Facilities in Mexico

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ABSTRACT

The objective of this work is to obtain the distribution and correlation between PAHs present in more than 50 sites evaluated since 2001. The research group on “Remediation of soils and aquifers from Instituto de Ingeniería, UNAM” has characterized soils of various hydrocarbon storage sites, such as refineries, storage tanks, distribution petroleum terminals and pumping plants from PEMEX, in different parts of the country, with the purpose of knowing the contamination by hydrocarbons and petroleum products.

Due to the importance that polynuclear aromatic hydrocarbons (PAHs) represent for human health, this research focused on the presence of PAHs in various soils of the country, through sampling and analysis, in sites with petroleum influence. Soils samples were analyzed, using the EPA 8310 method, from sites associated with leaks of collectors, drains, pumps, ducts and storage tanks.

The compounds benzo(a) pyrene, benzo(a)anthracene, benzo(b)fluoranthene, benzo(k)fluoranthene, have a higher concentration in the states of Oaxaca and Aguascalientes in silty clay soils.

The best relationship was for benzo(b)fluoranthene vs benzo(k)fluoranthene, since it presented a very favorable correlation coefficient and error “xy”, therefore it is considered that the linear equation adapts to the relationship between these two compounds. It was observed that the least favorable ratio was for benzo (a) pyrene vs benzo (a) anthracene since there is no linearity in the relationship between both compounds.

Keywords: Hydrocarbons; PAH; Contaminated Soils

Introduction

Polyaromatic compounds are part of the group of hazardous organic chemical compounds that exist naturally or may be of anthropogenic origin [1]. They have different number and position of their rings; They are generally of high molecular weight, low solubility and volatility and high adsorption capacity. They originate due to incomplete combustion of petroleum products, wood, and a wide variety of industrial processes, such as coal gasification processes, oil refining, coking, etc. [2]. Their physical and chemical properties vary according to their molecular weight and their distribution depends on these characteristics as well as on the properties of the soils as well as their effects on biological systems [3-5]. Due to their toxic, mutagenic and carcinogenic characteristics and the persistence of these compounds in contaminated soils and sediments, the remediation for these cases is very complex due to the presence of recalcitrant molecules that persist in the environment and increase the adsorption of soil organic matter and reduce the chances of biodegradation [3,6-10]. Influence processes in turn depend on the relationship between soil organic matter and clay content in each case [11,12].

Bacteria and fungi are valuable for the biotransformation of PAH [13] and are the path for their transformation [14-16]. However, for its biodegradation to be achieved, it is necessary to take into account the physical, chemical and biological properties. The most important are organic matter in the soil that controls biodegradation [17] and the number of rings. It has generally been shown that biodegradation in PAH of low molecular weight occurs faster than for those of high molecular weight [15].

Bioavailability of sorbed contaminants to microorganisms has been reported to be a major limitation for successful bioremediation of contaminated sites [17-20]. The ability of soil to desorb contaminants determines its susceptibility to microbial degradation, thereby influencing the effectiveness of bioremediation processes. Kinetics for desorption of PAHs from contaminated matrices is biphasic: an initial rapid desorption phase is followed by a slow desorption phase. Microbial activity is an important parameter controlling desorption kinetics during the initial desorption phase. Li et al (2009) [15] reported a biphasic pattern of PAHs during aging and demonstrated that the rapid diffusion of PAH from labile to nonlabile domains was primarily dependent on the distribution of micropore fraction and total organic carbon content. An unavailable PAH fraction may also exist in contaminated soils and sediments: this residual fraction may occur as a result of incorporation of sorbed contaminants into the soil/sediment organic matrix [21] these studies further demonstrate the significant role of organic matter on PAH sorption-desorption and subsequent degradation in contaminated soils.

Table 1 shows the permissible limits for the six PAH selected by Mexican regulations (DOF, 2012) [22]

| PAH | Permissible concentration (mg/kg) |
|------------------------|-----------------------------------|
| Benzo(a)pyrene | 10 |
| Dibenzo(a,h)anthracene | 10 |
| Benzo(a)anthracene | 10 |
| Benzo(b)fluoranthene | 10 |
| Benzo(k)fluoranthene | 80 |
| Indeno(1,2,3-cd)pyrene | 10 |

Table 1: Permissible limits for PAHs on industrial soils

Materials and Methods

The sampling consisted of the extraction of soil cores at each selected site. Pneumatic drilling equipment was used with a 12 HP motor, variable drilling velocity and a pressure of 2000 psi, from the surface to 3 m deep. Soil samples were obtained at 46 sites in 8 states of Mexico: Aguascalientes, Chihuahua, Coahuila, Durango, Estado de México, Monterrey, Oaxaca, Veracruz y Zacatecas, and four PAH, benzo (a) pyrene, benzo (a) anthracene, benzo (b) fluorantene and benzo (k) fluorantene were analyzed.

The frequency of the concentrations for each of the compounds with ArcGIS 10.2.2 was analyzed and a geostatistical analysis of these four compounds and their comparison between them was performed. Table 2 presents the sampling points, identification and location. Table 3 shows the concentrations of each PAH at each sampling point.

| Sample | State | Identification | Ubication (UTM) | |
|-------------|----------------|----------------|-----------------|------------|
| | | | X | Y |
| AG-5 | AGUASCALIENTES | AG-5 | 147615.8 | 2434586 |
| AG-35 | AGUASCALIENTES | AG-35 | 158231.1 | 2435609 |
| AG-36 | AGUASCALIENTES | AG-36 | 165521.1 | 2438039 |
| AVA1B | CHIHUAHUA | AVA1B | 217579.76 | 3098515.64 |
| JIM 1-3B | COAHUILA | JIM 1-3B | 21051.4 | 2993010 |
| CE-1A | COAHUILA | CE-1A | 27826 | 2949300 |
| PER 1B | COAHUILA | PER 1B | 59576.1 | 2927870 |
| PER 4A | COAHUILA | PER 4A | 69068.7 | 2922970 |
| SIBO 2B | COAHUILA | SIBO 2B | 93207.34 | 2841983 |
| SIBO 4A | COAHUILA | SIBO 4A | 115432.4 | 2840396 |
| GOPA 5A | DURANGO | GOPA 5A | 52432.3 | 2854840 |
| MAY 1A | DURANGO | MAY 1A | 33920.69 | 2740311 |
| MAY 2B | DURANGO | MAY 2B | 1276.292 | 2743570 |
| MAY 3A | DURANGO | MAY 3A | 40384.05 | 2722713 |
| MAY 5A | DURANGO | MAY 5A | 55375.36 | 2691426 |
| CATA 1A | EDO. DE MEXICO | CATA 1A | 362354.99 | 2103217.3 |
| ZOQUI 1A | EDO. DE MEXICO | ZOQUI 1A | 397676.935 | 2065910.97 |
| ZOQUI 2A | EDO. DE MEXICO | ZOQUI 2A | 353226.847 | 2100042.29 |
| CEIBA 1 | EDO. DE MEXICO | CEIBA 1 | 410773.837 | 2227047.55 |
| JUAN 1A | EDO. DE MEXICO | JUAN 1A | 448080 | 2143301 |
| POLI 1A | EDO. DE MEXICO | POLI 1A | 459589 | 2111551 |
| TEPE 1A | EDO. DE MEXICO | TEPE 1A | 527455 | 2127823 |
| TEPE 5A | EDO. DE MEXICO | TEPE 5A | 512000 | 2174650 |
| PAI 3A | MONTERREY | PAI 3A | 510000 | 286000 |
| DOR 1A | MONTERREY | DOR 1A | 376000 | 2671770 |
| DOR 2A | MONTERREY | DOR 2A | 377290 | 2640000 |
| DOR 6A | MONTERREY | DOR 6A | 354800 | 2645973 |
| CADE 4A | MONTERREY | CADE 4A | 396317 | 2821367 |
| CADE 5A | MONTERREY | CADE 5A | 398103 | 2831884 |
| CADE 6A | MONTERREY | CADE 6A | 412390 | 2826526 |
| CADE 8A | MONTERREY | CADE 8A | 416954 | 2817001 |
| SC 0+800 1A | OAXACA | SC 0+800 1A | 898931 | 1789860 |
| SC 1+900 1A | OAXACA | SC 1+900 1A | 901153 | 1790600 |
| SC 4+590 1A | OAXACA | SC 4+590 1A | 903481 | 1791440 |
| SC 6+020 1A | OAXACA | SC 6+020 1A | 905281 | 1792080 |
| SC 8+800 1A | OAXACA | SC 8+800 1A | 907926 | 1792930 |
| SC 9+850 1A | OAXACA | SC 9+850 1A | 910043 | 1793880 |
| SC LOLA 1A | OAXACA | SC LOLA 1A | 900931 | 1841200 |
| SC LOLA 3A | OAXACA | SC LOLA 3A | 904463 | 1839420 |
| MAZU 1A | VERACRUZ | MAZU 1A | 885133.3 | 2030864 |
| MAZU 3A | VERACRUZ | MAZU 3A | 886006.4 | 2031076 |
| MAZU 9A | VERACRUZ | MAZU 9A | 886588.5 | 2031235 |
| Z16 | ZACATECAS | Z16 | 75926.65 | 2560448 |
| Z11 | ZACATECAS | Z11 | 91642.46 | 2565686 |
| Z3A | ZACATECAS | Z3A | 110850.7 | 2572671 |
| Z15 | ZACATECAS | Z15 | 131805.1 | 2578492 |

NOTE: ND undetected

Table 2: Sampling points, identification and location

| Sample | Concentration (mg/kg) | | | |
|-------------|-----------------------|--------------------|----------------------|----------------------|
| | Benzo(a)pyrene | Benzo(a)anthracene | Benzo(b)fluoranthene | Benzo(k)fluoranthene |
| AG-5 | ND | 7.26 | 6.42 | ND |
| AG-35 | 5.24 | 18.11 | 2.6 | ND |
| AG-36 | 12.1 | 18.62 | ND | ND |
| AVA1B | ND | 1.11 | ND | ND |
| JIM 1-3B | 0.037 | 0.035 | 0.031 | 0.019 |
| CE-1A | 0.66 | 2.02 | 1.87 | 0.845 |
| PER 1B | ND | 1.315 | ND | ND |
| PER 4A | 0.007 | 0.426 | 0.173 | 0.077 |
| SIBO 2B | ND | 0.7 | ND | ND |
| SIBO 4A | ND | 1.48 | 0.06 | ND |
| GOPA 5A | ND | 0.019 | ND | ND |
| MAY 1A | ND | 0.672 | 0.124 | 0.042 |
| MAY 2B | ND | 0.905 | 0.221 | 0.076 |
| MAY 3A | 0.017 | 0.226 | 0.070 | 0.031 |
| MAY 5A | ND | 0.286 | 0.028 | 0.012 |
| CATA 1A | 0.11 | 0.108 | 0.110 | 0.051 |
| ZOQUI 1A | 0.079 | 0.071 | 0.111 | 0.052 |
| ZOQUI 2A | 0.014 | 0.038 | 0.034 | 0.012 |
| CEIBA 1 | 0.008 | 0.008 | 0.002 | 0.003 |
| JUAN 1A | 0.053 | 0.0734 | 0.054 | 0.028 |
| POLI 1A | 0.004 | ND | 0.15 | 0.022 |
| TEPE 1A | 0.024 | 0.0222 | 0.029 | 0.014 |
| TEPE 5A | 0.041 | 0.0115 | 0.062 | 0.026 |
| PAI 3A | ND | 0.275 | 0.047 | 0.010 |
| DOR 1A | ND | 0.0079 | ND | ND |
| DOR 2A | 0.002 | 0.058 | 0.01 | 0.005 |
| DOR 6A | ND | 0.062 | ND | ND |
| CADE 4A | ND | 0.024 | 0.008 | ND |
| CADE 5A | 0.016 | 0.009 | 0.008 | 0.007 |
| CADE 6A | 0.032 | 0.022 | 0.049 | 0.016 |
| CADE 8A | ND | ND | 0.006 | ND |
| SC 0+800 1A | 0.38 | 0.370 | 0.348 | 0.177 |
| SC 1+900 1A | 0.34 | 0.285 | 0.352 | 0.171 |
| SC 4+590 1A | 0.079 | 0.095 | 0.091 | 0.041 |
| SC 6+020 1A | 15.21 | 16.79 | 17.0 | 8.25 |
| SC 8+800 1A | 0.134 | ND | 0.136 | 0.045 |
| SC 9+850 1A | 2.045 | 2.54 | 2.150 | 1.11 |
| SC LOLA 1A | 0.174 | 0.185 | 0.149 | 0.072 |
| SC LOLA 3A | 2.471 | 3.76 | 3.944 | 1.444 |
| MAZU 1A | ND | 0.21 | 0.075 | 0.039 |
| MAZU 3A | ND | 0.146 | 0.063 | 0.039 |
| MAZU 9A | ND | 0.137 | 0.050 | 0.027 |
| Z16 | ND | 0.733 | 0.545 | ND |
| Z11 | 1.426 | 6.695 | 1.72 | ND |
| Z3A | 0.78 | 0 | 2.35 | ND |
| Z15 | ND | 0.812 | ND | ND |

NOTE: ND undetected

Table 3: Concentrations of each PAH at each sampling point

Results

Benzo (a) pyrene was detected in 27 of the 48 sites, with concentrations between 0.0024 to 15.2 (mg / kg). A frequency of 41 was obtained, corresponding to 89% in concentrations less than 1.5 mg / kg and distributed in 8 states. The frequency histograms, as well as the statistical data are presented in Figure 1. Two samples exceeded the permissible limits, located in Aguascalientes and Oaxaca with respective concentrations of 12.1 and 15.2 (mg / kg), with keys of identification AG-36 and SC 6 + 020 1A.

Figure 2 shows the sampling points grouped into: within the frequency, outside of it and the points with concentrations that exceed the permissible limit (10 mg / kg).

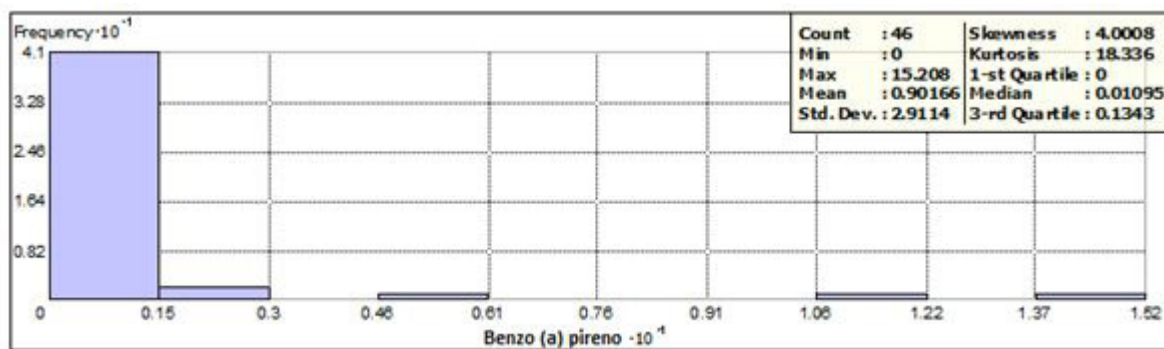


Figure 1: Frequency histogram of benzo(a)pyrene and its statistical data

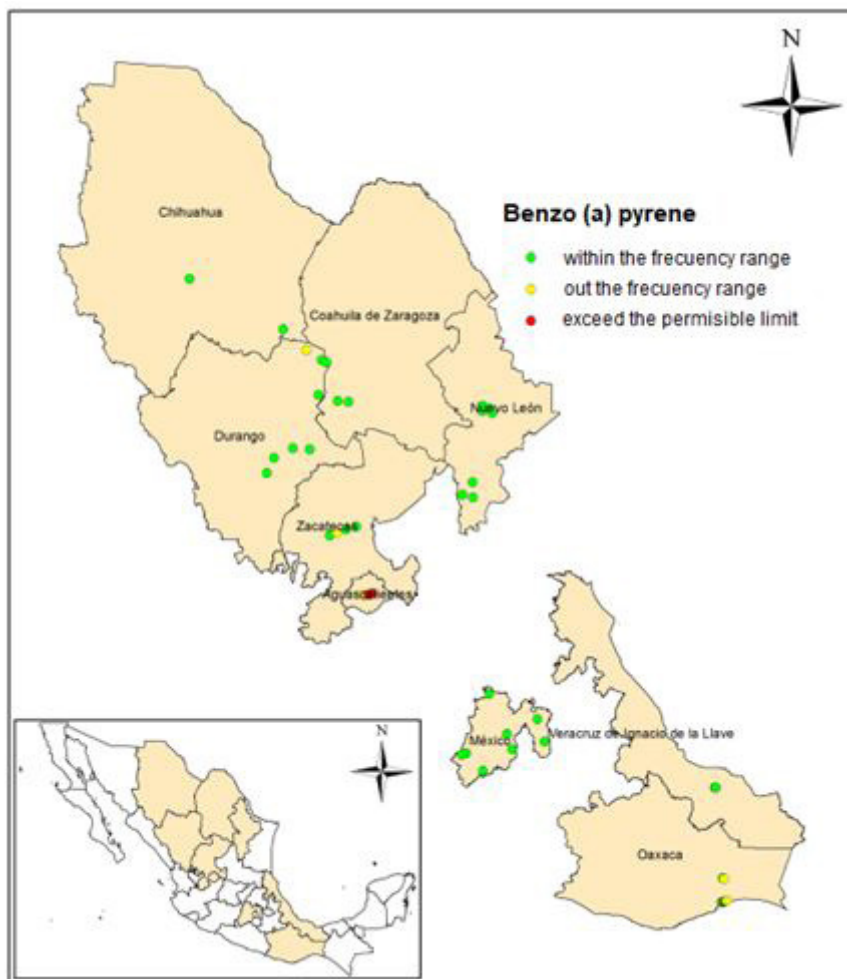


Figure 2: Sampling points that are within the frequency, outside of it and the points with concentrations that exceed the permissible limit

Benzo (a) anthracene was detected in 44 sites, with concentrations between 0.0079 to 18.62 (mg / kg). A frequency of 38 was obtained, corresponding to 82.6% in concentrations less than 1.9 mg / kg and distributed in 8 states. The frequency histogram, as well as the statistical data are presented in Figures 3 and 4. Three samples exceeded the permissible limits, located in Aguascalientes and Oaxaca with respective concentrations of 16.8, 18.1 and 18.6 (mg / kg), with keys of identification AG-35 AG-36 and SC 6 + 020 1A. Two of these samples also had concentration values above the permissible limit in benzo (a) pyrene.

Figure 3 shows the histogram of frequencies and statistical data, and Figure 4 shows the location of the sampling points within the frequency range, outside it and the points at which there is a concentration above of the permissible limit (10 mg / kg).

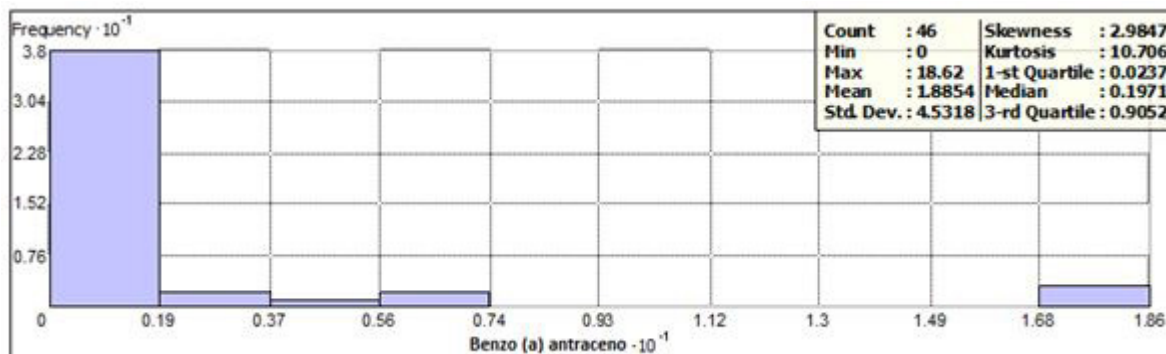


Figure 3: Frequency histogram of benzo(a)anthracene and its statistical data

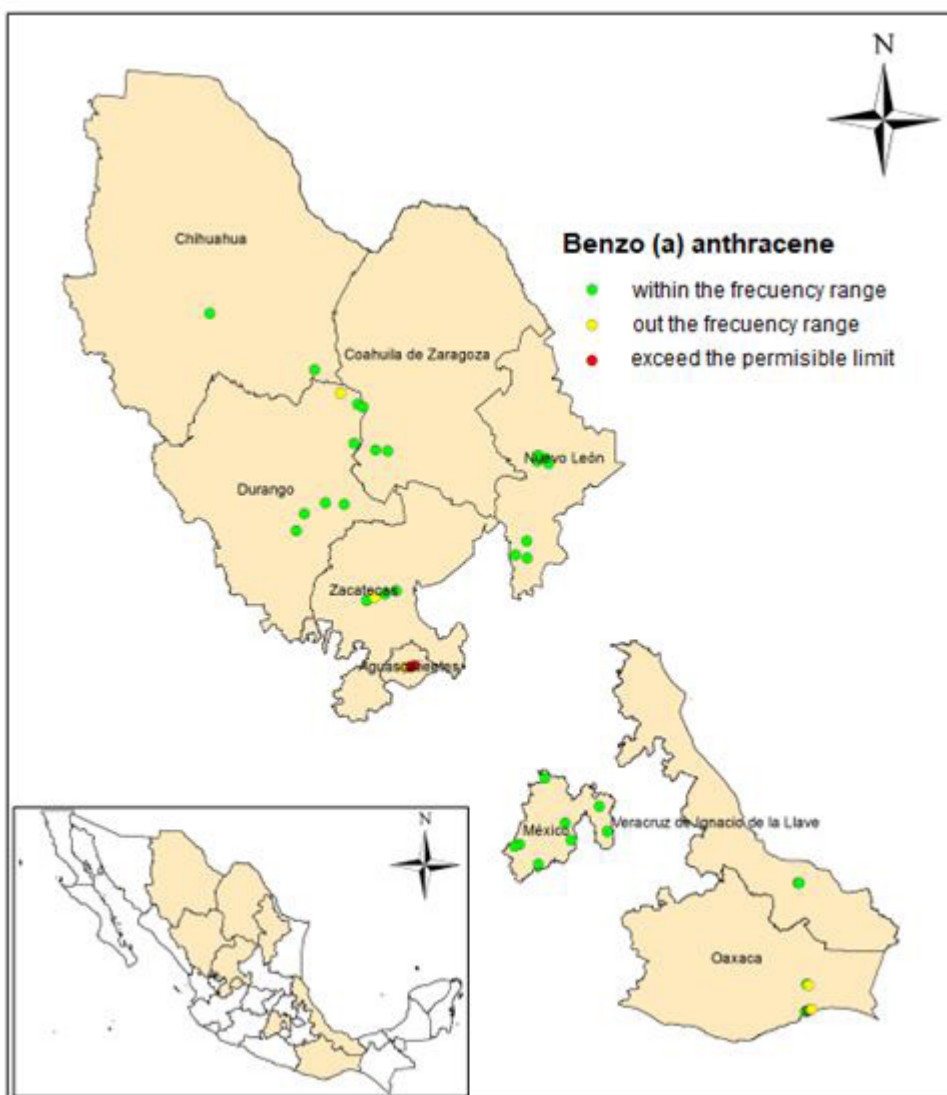


Figure 4: Sampling points that are within the frequency, outside of it and the points with concentrations that exceed the permissible limit

Benzo (b) fluoranthene was detected in 38 sites, with concentrations between 0.0022 to 17 (mg / kg). A frequency of 38 was obtained, corresponding to 82.6% in concentrations less than 1.7 mg / kg and distributed in 8 states. The frequency histogram, as well as the statistical data are presented in figures 5 and 6. Only one sample exceeded the permissible limits, located in Oaxaca with concentration of 17 mg/kg with key of identification SC 6 + 020 1A. The same sample had concentration values above the permissible limit in benzo (a) pyrene and benzo(a)anthracene.

Figure 5 shows the histogram of frequencies and statistical data, and Figure 6 shows the location of the sampling points within the frequency range, outside it and the points at which there is a concentration above of the permissible limit (10 mg / kg).

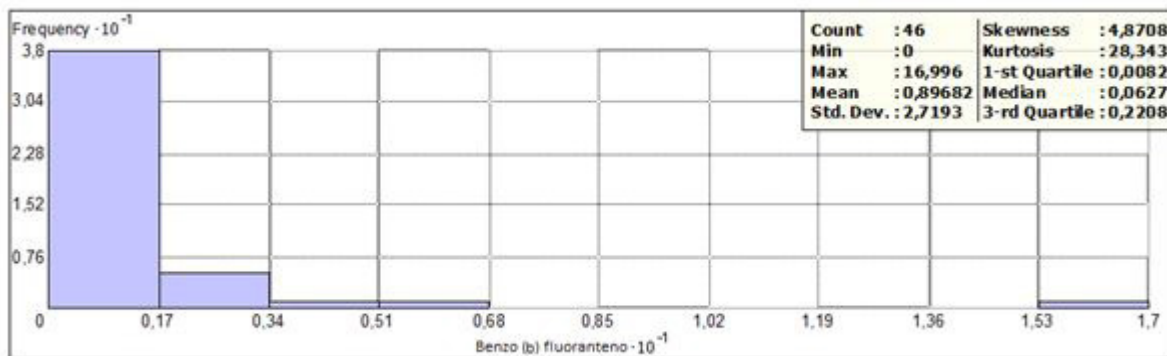


Figure 5: Frequency histogram of benzo(b)fluoranthene and its statistical data

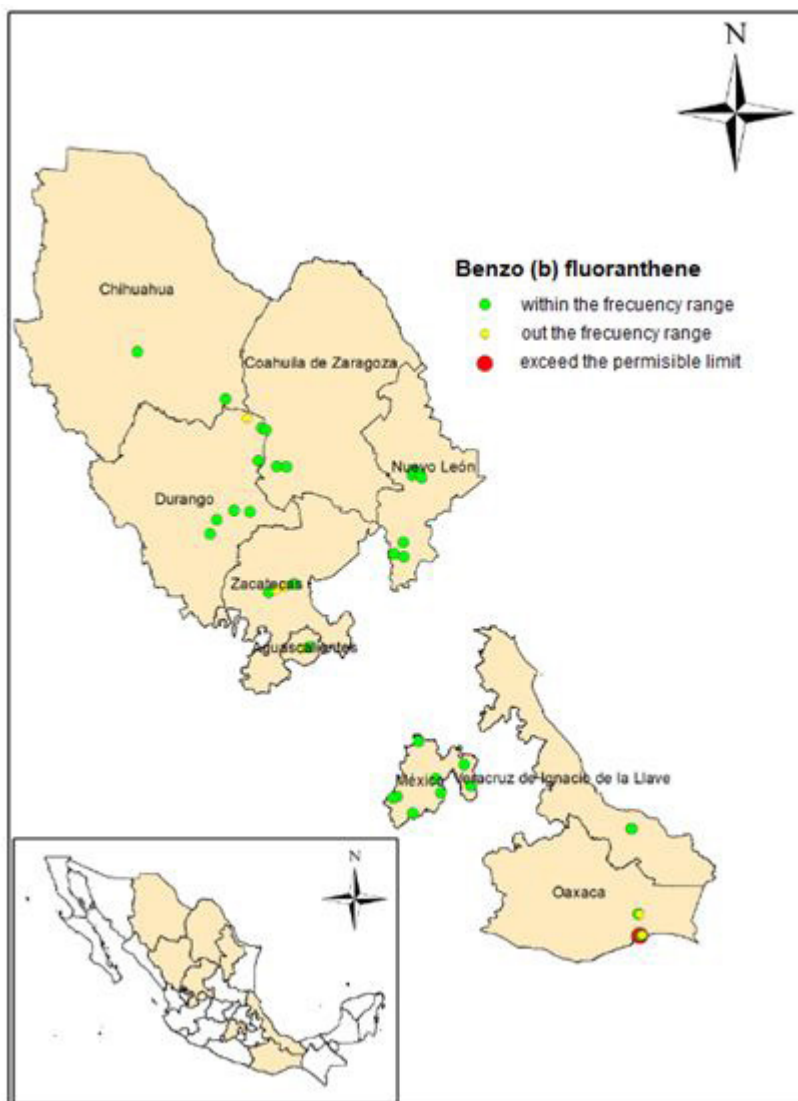


Figure 6: Sampling points that are within the frequency, outside of it and the points with concentrations that exceed the permissible limit

Benzo (k)fluoranthene was detected in 28 sites, with concentrations between 0.0035 to 8.25 (mg / kg), having a frequency greater than the concentration range less than 0.83 mg / kg (Figure 7) corresponding to 91%. The maximum concentration value (8.25 mg / kg) for this compound was found in the same point in Oaxaca, with the identification code SC 6 + 020 1A; however, it does not exceed the maximum permissible limit (80 mg / kg). The frequency histogram, as well as the statistical data are presented in Figures 7 and 8.

Figure 7 shows the histogram of frequencies and statistical data, and figure 8 shows the location of the sampling points within the frequency range, outside it and the points at which there is a concentration above of the permissible limit (10 mg / kg).

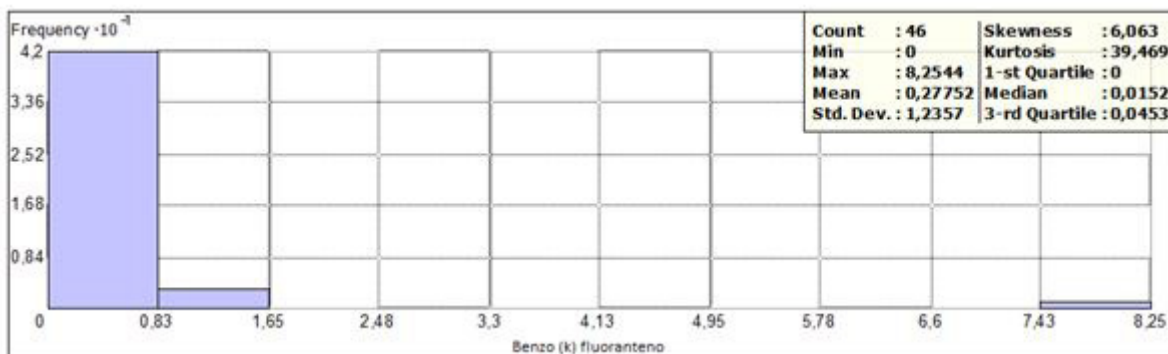


Figure 7: Frequency histogram of benzo(k)fluoranthene and its statistical data

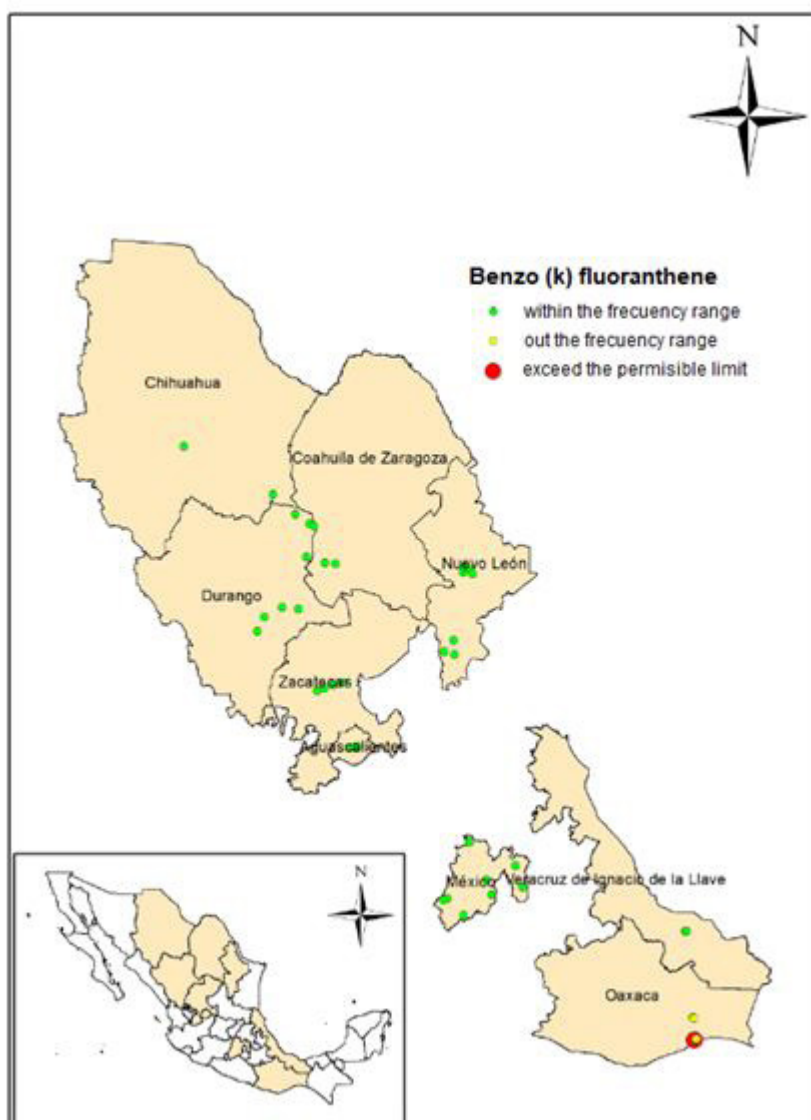


Figure 8: Sampling points that are within the frequency, outside of it and the points with concentrations that exceed the permissible limit

Relationship between the four compounds

A linear trend model was used to analyze the relationship between the 4 PAH studied. Figure 9 shows the analysis of the relationship between benzo (a) pyrene and benzo (a) anthracene and there is no relationship between the concentrations of the different sampling points.

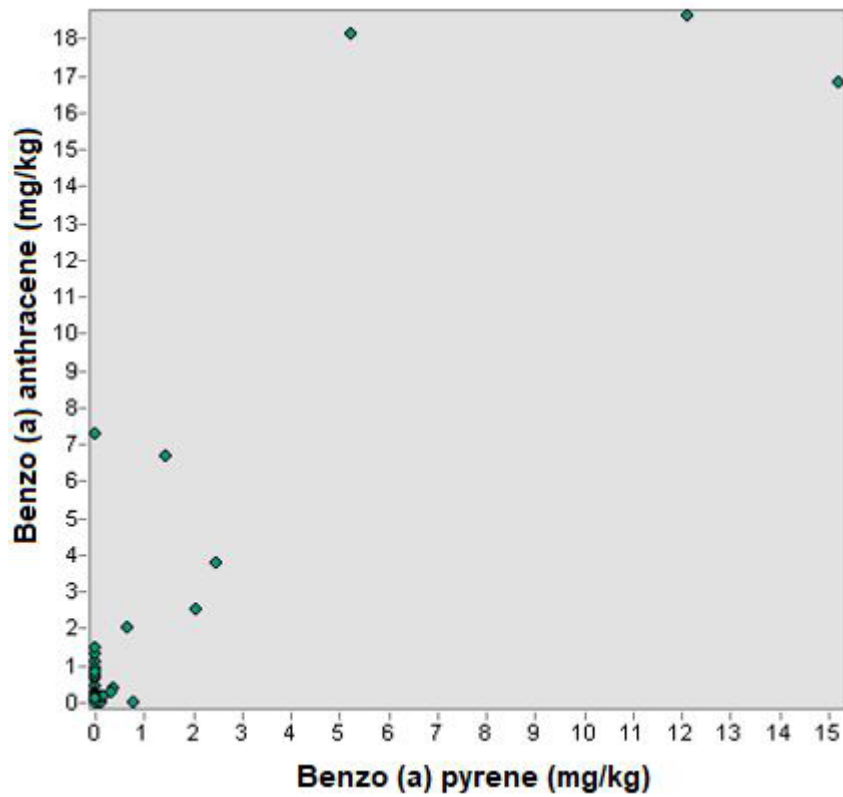


Figure 9: Relation between benzo(a)pyrene and benzo(a)anthracene

The ratio between benzo (a) pyrene and benzo (b) fluoranthene, has a good relationship. According to the trend line the correlation coefficient is 0.9844 and the typical error according to the regression is 36.7%. It can be said that the trend line fits the relationship equation (Figure 10). In this analysis the points of Aguascalientes were not taken into account.

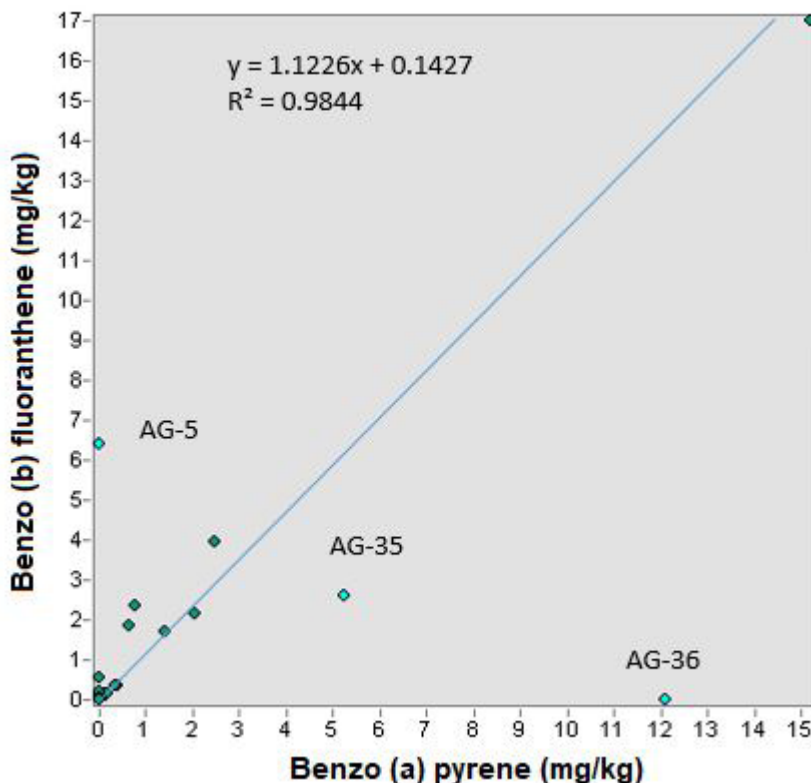


Figure 10: Relationship between benzo(a)pyrene and benzo(b)fluoranthene

Regarding the relationship between the compounds benzo (a) pyrene and benzo (k) fluoranthene, the points of Aguascalientes and Zacatecas were not taken into account because they leave the trend line (Figure 11). In this way there is a correlation coefficient of 0.9934 and a typical “xy” error of 12.3%, which makes this relationship reliable through the linear trend equation ($0.5423x + 0.0145$) and it is concluded that there is a good correlation between these compounds.

In the relationship of benzo (a) anthracene and benzo (b) fluoranthene, there is also linearity; In Figure 12, a correlation coefficient of 0.98 is observed through the generation of the trend line; however, the typical error “xy” was 39.9% which is greater than the ratio of the previous compounds, but it can be said that although this typical error, the linear trend equation is reliable and that there is a relationship between both compounds. The point of Aguascalientes y Zacatecas were not taken into account in this analysis.

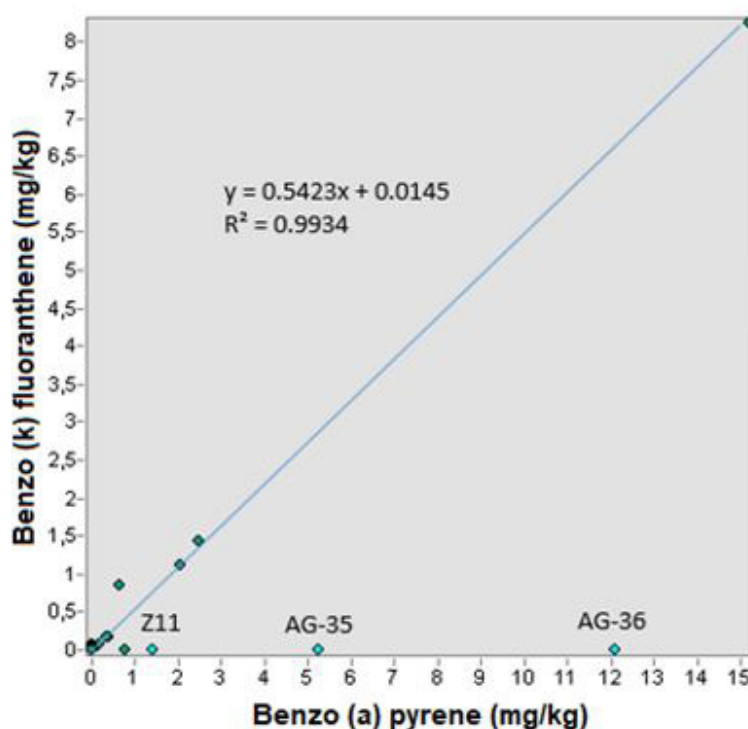


Figure 11: Relationship between benzo(a)pyrene and benzo(k)fluoranthene

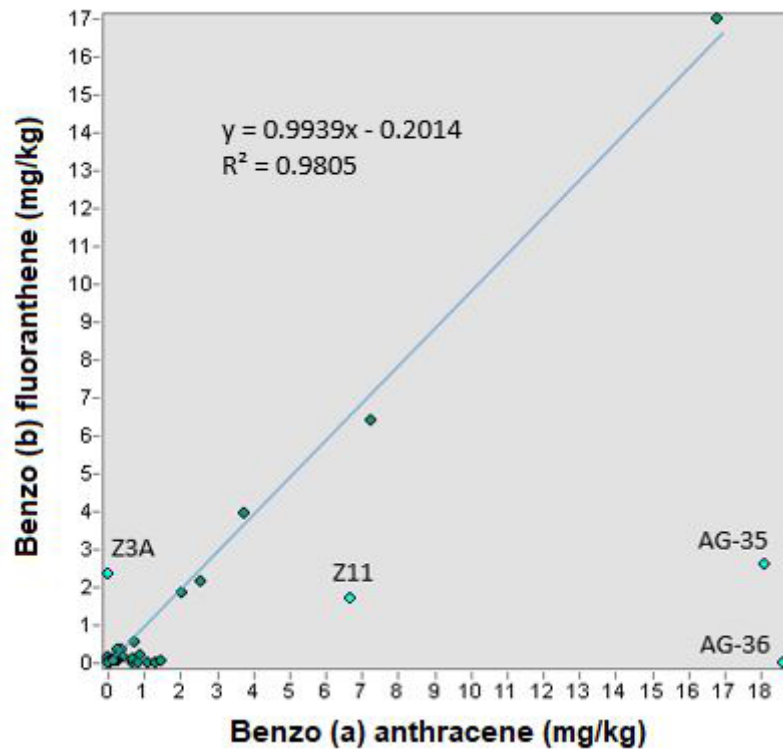


Figure 12: Relationship between benzo(a)anthracene and benzo(b)fluoranthene

In Figure 13 it can be seen that the trend line has a correlation of 0.9777 and a typical error of 20%, so it can be inferred that there is a relationship between both compounds.

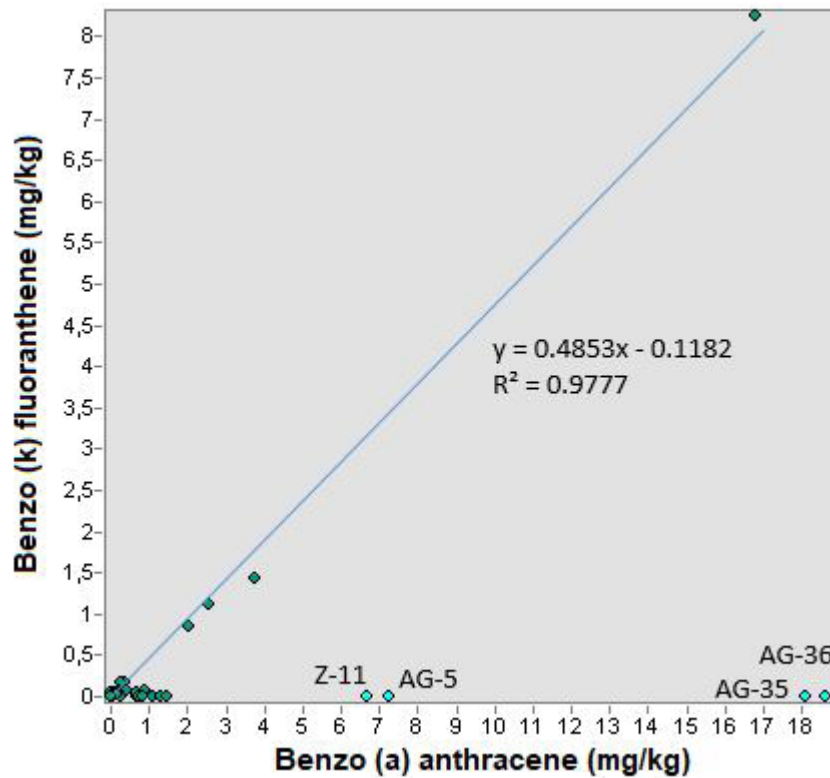


Figure 13: Relationship between benzo(a)anthracene and benzo(k)fluoranthene

Finally the relationship between benzo (b) fluoranthene and benzo (k) fluoranthene is analyzed where it is observed that again the points that come out of the linearity correspond to the states of Aguascalientes and Zacatecas (AG-5, AG-35, Z3A and Z11), but still there is a better linearity than the ratio of the previous compounds. In Figure 14, there is a correlation coefficient of 0.996 and a typical “xy” error of 9.3%, confirming that this relationship between the two compounds is better than those previously analyzed and that the equation $y = 0.4814x - 0.0233$ adapts to the concentration ratio between these compounds [23-26].

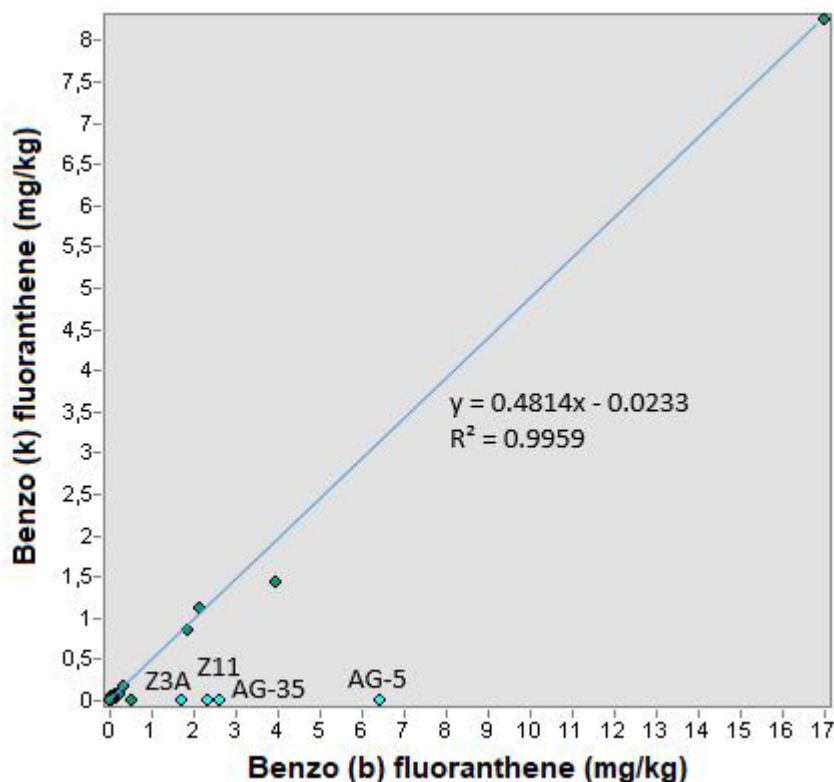


Figure 14: Relationship between benzo(b)fluorantene vs benzo(k)fluorantene

Conclusions

The four PAHs analyzed, have the highest concentration in the states of Oaxaca and Aguascalientes, being the site of SC 0 + 620 1A, in which the highest concentrations were presented. Although for the benzo (k) fluoranthene there were no concentration values above the permissible limit, there are very high values compared to the remaining sampling sites.

Benzo (b) fluoranthene vs benzo (k) fluoranthene had the best relationship, since it has a very favorable “xy” correlation coefficient and error, which concludes that the linear equation adapts well to the relationship between these two compounds. It was also observed that the least favorable relationship was for the relationship between benzo (a) pyrene vs benzo (a) anthracene since no linearity was obtained in the relationship between both compounds.

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