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Polycyclic aromatic hydrocarbons in ambient air particles in the city of Las Palmas de Gran Canaria

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Abstract

Concentration levels, seasonal variation and winter/summer ratios of 11 polycyclic aromatic hydrocarbons (PAHs) in the TSP of Las Palmas de Gran Canaria city were determined. The study area is under the influence of heavy traffic (80,000 vehicles/day). Pyr/BaA, Pyr/Flt, B(ghi)P/I(cd)P and B(b + k)F/B(ghi)P ratios were calculated; the values of these two last suggest a strong influence of diesel fuel burning. Levels of TSP and TSP-bound PAHs were also established. The hazard potential of PAHs in terms of carcinogenicity of BaP is acceptable. Potential risk on the basis of TEFs is also studied.

Keywords: Polycyclic aromatic hydrocarbons; Carcinogenicity; Health risk

1. Introduction

There is growing evidence on the noxious effects of atmospheric pollutants on human health. Thus, the results of several epidemiological studies (Nielsen, 1996; Doll and Peto, 1981; Speizer, 1986) suggest that 1–10% of lung tumours are due to the presence in the atmosphere of chemical species from anthropogenic sources. Especially prominent among such species, on account of the facts that they originate from various combustion processes and pose severe health risks, are polycyclic aromatic hydrocarbons (PAHs).

PAHs occur in both gaseous and particulate form in the atmosphere; both types of phases can come into contact with living beings, either externally (through the skin) or internally (through the lungs). This makes them hazardous or even toxic species. In fact, some PAHs are major mutagenic and carcinogenic agents (Miguel, 1998; IARC, 1991; EPA, 1992; Nauss, 1995; Petry et al., 1996) according to the classification of the International Agency for Research on Cancer (IARC). The body organs most markedly affected by the action of PAHs are the skin, lungs and bladder (Moorcroft, 1999). In addition to their mutagenicity and carcinogenicity, PAHs have been found to exert many other toxic effects on experimental animals (Nebert et al., 1980; Payne, 1958; White et al., 1985).

Automobile exhaust is one of the main sources of urban PAHs emissions. The amount of PAHs released into the atmosphere has grown hand-in-hand with the global trend to suppressing lead as a petrol additive since preserving an acceptable fuel knocking conditions in this situation entails the production of increased amounts of PAHs (Lee et al., 1995; Pedersen et al., 1980).

PAH emission profiles vary among engine types. Thus, diesel engines are the principal source of low-molecular weight PAHs, whereas petrol engines release the greatest amounts of high-molecular weight PAHs such as benzo(a)pyrene and dibenzo(a,h)anthracene (Nielsen, 1996; Miguel, 1998; Rogge et al., 1993). For this reason, some authors (Daisey et al., 1979, 1986; Daisey and Lioy, 1981; Li and Kamens, 1993) recommend using them as tracers for vehicular traffic-related pollution or particulate matter in the atmosphere.

The city of Las Palmas de Gran Canaria, with a population density in the region of 3520 per square kilometer, lies on the northeast end of the Gran Canaria Island (one of the seven that constitute the Canary archipelago). The city bears heavy automobile traffic as a result not only of the large vehicle base on the island (more than 450,000 units in 1999), but also of the fact that, because of the way the road network is arranged, every vehicle coming from or going to the main

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tourist resorts (in the south) must inevitably cross it, so they probably constitute the main source of anthropogenic pollution in the area.

Because automobile emissions are among the principal sources of mutagenicity in urban atmospheres (Alfheim et al., 1983)—which has aroused especial interest among scientists in both particulate emissions from diesel engines (Clark et al., 1982; Rapaport et al., 1980) and PAH emissions from petrol engines (Löfroth, 1981; Westerholm et al., 1988; Alsberg et al., 1985)—a city area that might be used as a reference in studying such emissions was chosen as vehicles are the only major source of anthropogenic pollution in it. The specific area studied was Bravo Murillo, a downtown street with heavy motor vehicles traffic (80,000 vehicles/day), of which, a high proportion are diesel propelled (taxis, buses and trucks), and the main roadway connecting the downtown and uptown districts. This is a typical downtown street with many office blocks and virtually uninterrupted human traffic from 07:00 am to 09:00 pm.

In this work, the concentration levels of 11 PAHs including the most mutagenic and carcinogenic congeners present in ambient air particles collected in the sampled street were determined, as was their seasonal variation, with a view to assessing the aggressiveness of this environment to living beings. Because BaP and most PAHs with mutagenic properties, except BaA, occur as particulate matter (Menichini, 1999), the study was restricted to this phase.

2. Experimental

2.1. Sampling and analysis

Airborne particulate matter was collected in 24-h periods starting at 08:00 am, by filtration through glass microfiber filters (Whatman GF/A 20 × 25 cm, kiln fired at 400 °C overnight) using Hi-Vol sampling system (CAV-P; MCV, Collbató, Spain). After sampling, all filters were wrapped in aluminium foil (Halsall et al., 1993; Gustafson and Dickhut, 1997) and stored at −20 °C (Allen et al., 1996; Baek, 1988) until analysis in the laboratory. A total of 28 samples (seven samples/season) between July 8, 1999 and June 1, 2000.

Three extractions of 30 min each for every filter was made ultrasonically (Bransonic, model 2510) with dichloromethane (Nielsen, 1996; Escrivá et al., 1991; Fromme et al., 1998). The extracts were concentrated to 2 ml in a rotary evaporator at 35 °C and 800–810 mbar. The concentrated extracts were fractionated by column chromatography. A column filled with 2 g silica was used. The PAH were collected in the second fraction, nitrogen concentrated to almost dryness and redissolved with dichloromethane (20%) and acetonitrile (80%). Recovery factors were determined by spiking filters with standard (NIST SRM 1647c). The recovery factors of this extraction and fractionation process were in the order of 90–100%.

The samples were analyzed by GC coupled to mass spectrometer (GC-MS). These analyses were performed with a Shimadzu GC/MS system consisting of an GC-17A gas chromatograph and Shimadzu QP-5000 mass spectrometer. A 30 m × 0.25 mm i.d. HP-5MS (film thickness 0.25 μm) was used. The GC analyses were performed with an oven temperature program from 50 to 300 °C at 8 °C/min; injector and transfer line temperatures of 300 and 230 °C, respectively; and helium was the carrier gas. The injector was in the splitless mode.

Compound identification was based on the GC-MS data and co-injection with authentic standards. Quantitation was performed from the GC profiles using the external standard method. The aromatic hydrocarbons were quantitated with a standard containing naphthalene (Naph), acenaphthene (Ace), acenaphthylene (Acy), fluorene (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flt), pyrene (Pyr), benzo(a)anthracene (BaA), chrysene (Cry), benzo(b)fluoranthene and benzo(k)fluoranthene (B(b + k)F), benzo(a)pyrene (BaP), dibenz(a,h)anthracene, benzo(g,h,i)perylene (B(ghi)P), indeno (1,2,3-cd)pyrene (I(cd)P). Benzo(b)fluoranthene and benzo(k)fluoranthene with similar elution times were quantitated together.

3. Results and discussion

The concentrations of primary atmospheric pollutants depend markedly on the position of the sampling station and on the particular weather conditions. This makes it rather difficult to compare results from the different studies especially considering that only 28 samples were available. This general constraint was even greater in this study because of the haze present during four of the seven sampling days of wintertime, which had a marked effect on the total suspended matter (TSP) levels recorded and hence on all related parameters.

3.1. Total suspended matter

Table 1 lists the average TSP concentrations obtained each season, as well as the overall average value for the studied period as a whole. As can be seen, the highest concentrations occurred in winter, consistent with previous

Table 1
Total suspended particulate (TSP) concentration in ambient air of Bravo Murillo street

Season	TSP concentrations (μg/m^3)	
	Range	Mean
Winter	57.44–513.80	232.60
Spring	31.60–78.61	55.41
Summer	50.58–67.26	72.39
Autumn	32.68–67.26	49.72
		102.53

findings in other cities with heavy traffic such as Barcelona and Cartagena (both in Spain) (Aceves and Grimalt, 1993; Moreno Grau et al., 1999) or in highly industrialized cities such as Katowice (Poland) and Hamburg (Germany) (Pastuszka et al., 1993). The high value for winter means observed in Las Palmas appears to be due, among others, to enrichment of the urban aerosol with K from the sea, which is blown away by Trade Winds from the first and fourth quadrants of compass card, directions NW, N, NNE, NE (Vera Castellano, 1992; Blanchard, 1983), which introduces soil dust from the rest of islands into the city. However, such a high value obtained in this work, 232.6 µg m^{-3}, was no doubt also the result of the presence of haze aerosols (natural origin) coming from Sahara Desert and common during the winter months in episodes lasting 1–5 days.

The average TSP value, 102.5 µg m^{-3}, is lower than those reported for other urban areas such as Taichung (165 µg m^{-3}) (Fanf et al., 2000), but very similar to the values obtained by Steiger et al. (1990) in industrial German cities. The significance of our result lies in the fact that, as shown by previous studies (Vera Castellano et al., 2002), roughly one-half of TSP in the area consists of particles less than 2 µm in size (mass median diameter 0.33–0.39 µm), which is a rough estimate of its hazardous potential.

3.2. PAH concentrations

Neither acenapthylene nor dibenzo(ah)anthracene were detected among the PAHs studied. Also, naphthalene and acenaphthene were only occasionally encountered, so their presence in the studied atmosphere was assumed to be insubstantial because mainly gas-phase and the two congeners, which are considered to be irrelevant to the ensuing discussion, were found in ambient air. Table 2 shows the seasonal and overall average concentrations of the 11 PAHs quantified.

The heavier PAHs (MW > 200) were found to be the most abundant in all seasons; this was particularly the case with B(b + k)F and, except in spring, B(ghi)P, which is consistent with the previous results of Nielsen (1996) and Li and Kamens (1993). BaP concentrations ranged from 0.160 to 0.605 ng m^{-3} and were thus considerably lower than those in 1989/1990 in Barcelona (1.6–6.54 ng m^{-3}) (Bayona et al., 1994). Also, they were lower than the limit imposed by the German Federal Environment Agency (Baek et al., 1992).

As can be seen, pyrene concentrations (0.381 µg m^{-3}) are much more lower than the other four-ring PAHs. Ratios between Pyr/BaA, 0.83, and Pyr/Flt, 0.36, are smaller than the found in particulate aerosol of cities such Barcelona 1.07–1.39 (Bayona et al., 1994), Copenhagen 1.78–1.30 (Nielsen et al., 1996), Birmingham 1.59–2.01 (Harrison et al., 1996) and several Greek cities (Papageorgopoulou et al., 1999). Taking into account that, as shown elsewhere (Murray et al., 1974), the proportion of some PAHs in the gas phase is 100 times higher at 30 °C than at 10 °C, the pyrene concentrations found in the Jinámar Valley might be the result of a highly favourable distributions into such a phase (at levels well above the usual for other places as a consequence of the mild temperatures that prevail on the island of Gran Canaria throughout the year).

Li and Kamens (1993) reported B(ghi)P/I(cd)P ratios over the ranges 3.5–3.8 for petrol engines and 1.1–1.2 for diesel engines. Our ratios were 1.16 in winter and 1.78 in summer, which suggests a stronger influence of diesel fuel burning; these values are consistent with those of Greenberg and Darak (1985) for New Jersey in both winter (0.74–1.13) and summer (0.27–1.67), and with those found by Nielsen (1996) in Copenhagen in winter in a street with heavy traffic (1.77) and in one with bus traffic (0.2–1.67). If the B(b + k)F/B(ghi)P ratios reported by Cretney et al. (1985) are subtracted from those of Li and Kamens (1993), a value of 0.33 for petrol engines and 1.60 for diesel engines results. Our ratios were 1.49 in winter and 1.23 in summer, which again reflect the influence of diesel fuel burning.

There were no specially large differences among seasonal concentrations. The lowest overall PAH levels were recorded in spring and the highest in autumn—particularly for Phe, Chr, B(b + k)F, BaP and B(ghi)P. This is consistent with previous findings in other areas on the Gran Canaria Island (López Cancio et al., 2002) but departs from the results

Table 2
Mean concentrations (ng m^{-3}) ± standard deviations of PAH's at Bravo Murillo over the whole period and each season and ratios winter/summer

Variable	Mean	Winter	Spring	Summer	Autumn	W/S
Flr	0.208 ± 0.065	0.306 ± 0.252	0.167 ± 0.125	0.134 ± 0.063	0.223 ± 0.186	2.28
Phe	0.205 ± 0.059	0.236 ± 0.168	0.147 ± 0.065	0.166 ± 0.068	0.272 ± 0.121	1.42
Ant	0.047 ± 0.031	0.069 ± 0.075	0.006 ± 0.002	0.084 ± 0.066	0.027 ± 0.028	0.82
Pyr	0.381 ± 0.144	0.358 ± 0.161	0.270 ± 0.112	0.272 ± 0.091	0.622 ± 0.225	1.14
Chr	0.633 ± 0.301	0.705 ± 0.133	0.228 ± 0.185	0.536 ± 0.184	1.063 ± 1.279	1.32
Flt	1.049 ± 0.323	1.076 ± 0.569	0.637 ± 0.250	0.948 ± 0.613	1.536 ± 0.831	1.67
BaA	0.457 ± 0.104	0.580 ± 0.250	0.361 ± 0.128	0.347 ± 0.125	0.541 ± 0.317	1.32
B(b + k)F	2.197 ± 0.623	2.466 ± 1.320	1.508 ± 1.225	1.729 ± 0.421	3.085 ± 1.610	1.43
BaP	0.341 ± 0.163	0.316 ± 0.218	0.160 ± 0.035	0.284 ± 0.085	0.605 ± 0.392	1.11
I(c,d)P	0.826 ± 0.417	1.416 ± 0.171	0.238 ± 0.121	0.786 ± 0.216	0.864 ± 0.662	1.80
B(ghi)P	1.436 ± 0.486	1.653 ± 1.385	0.538 ± 0.026	1.403 ± 0.560	1.789 ± 1.297	1.18

obtained in other parts of the world; in fact, winter levels are usually higher as a result of increased photochemical degradation during the summer. According to Aceves and Grimalt (1993), however, the reason for this departure may be that climatic differences between the two seasons on the island are minimal.

3.3. Winter/summer ratios

The summer/winter ratios for total PAHs (ΣPAH) and each individual congener are listed in the last column of Table 1. As can be seen, except for Flt and I(cd)P, they ranged from 1 to 2; the sole value below unity was that for anthracene, which is consistent with previous results of Escrivá et al., 1991 for Valencia and reflects a distinct behaviour in this PAH. As can also be seen, the W/S ratio for ΣPAH was 1.37 and hence much lower than those found by Papageorgopoulou et al. (1999) in the Greek cities of Moudania and Kilkis (12 and 12, respectively), but similar to that found in Edessa, a city that bore heavy vehicular traffic in the sampled zone during the summer. The W/S ratio for carcinogenic PAHs on the whole was 1.52 and slightly higher than that found in Edessa.

3.4. TSP-bound PAH levels

Table 3 shows the individual and global levels of particle-bound PAHs. As can be seen, the highest levels were recorded in autumn. Overall, the individual levels are consistent with those reported by Papageorgopoulou et al. (1999). The highest values among the PAHs with MW < 228 and MW>228 were those for fluoranthene and B(b + k)F, respectively. The winter/summer (W/S) ratio for ΣPAH, 0.37, was very low; on the other hand, the autumn/summer (A/S) ratio was somewhat higher (2.28) and consistent with the values found in the abovementioned Greek cities (Papageorgopoulou et al., 1999). The shift in the maxima from winter to autumn, and the value of the W/S ratio can be ascribed to the presence of winter haze. This also explain the minimum level for the B(ghi)P bound to TSP during wintertime.

Table 3
Contents of TSP-bound PAHs (μg/g)

Variable	Mean	Winter	Spring	Summer	Autumn
Flu	2.598	1.316	2.736	1.850	4.490
Phe	2.788	1.015	2.376	2.290	5.470
Ant	0.487	0.176	0.072	1.160	0.540
Pyr	5.276	1.539	3.294	3.760	12.510
Chr	7.899	2.743	0.072	7.400	21.380
Flt	14.748	4.627	10.386	13.090	30.890
BaA	6.171	2.494	6.498	4.790	10.900
B(b + k)F	29.714	10.604	22.320	23.880	62.050
BaP	4.747	1.045	3.920	3.920	12.170
I(c,d)P	7.785	5.719	10.850	10.850	14.500
B(ghi)P	15.107	3.500	19.380	19.380	35.980
ΣPAH	97.320	34.778	51.246	92.370	210.900

Table 4
B(a)P$_{eq}$ concentrations using Nisbet and LaGoy's (1992) TEFs

Variable	pg m^{-3}	Factor of Nisbet and LaGoy	B(a)P$_{eq}$ (ng m^{-3})
Flu	208	0.001	0.208
Phe	205	0.001	0.205
Ant	47	0.01	0.47
Pyr	381	0.001	0.381
Chr	633	0.01	6.33
Flt	1049	0.001	1.049
BaA	457	0.1	45.7
B(b + k)F	2197	0.1	219.7
BaP	341	1	341
I(c,d)P	826	0.1	82.6
B(ghi)P	1346	0.01	13.46
			711.1

3.5. Toxicological considerations

As with other tumour-inducing agents, no threshold for a dose–response relation appears to exist for PAHs, so no absolutely safe levels could be established.

The hazard potential of PAH can be determined in terms of BaP, which is an indicator for carcinogenicity (Menichini, 1999). In fact, some European countries have established target annual mean values of BaP from 0.7 to 1.3 ng m^{-3} (Pérez Ballesta et al., 1999). Accordingly, our BaP levels were about 50% of the previous upper limit, so they can be considered acceptable.

This form to measure the hazard potential assume to ignore the rest of PAH, some of them with a carcinogenic potency as high as benzo(a)pyrene and would be an inadequate evaluation of the risk of hydrocarbons altogether. In order to avoid this shortcoming, Toxic Equivalency Factors (TEFs) have been established. The most widely known TEFs are those reported by Chu and Chen (1984), Clement (1988), and Thorslund and Farrar (1990), and the most recent ones those of Nisbet and LaGoy (1992)—these last may be more accurate reflections on currently available knowledge on the relative potency of PAHs.

Table 4 shows concentrations of different carcinogenic PAH calculated in terms of benzo(a)pyrene equivalents, B(a)P$_{eq}$, using the TEF of Nisbet and LaGoy and total sum. From the results, it can be concluded that the potential risk on the Las Palmas de Gran Canaria city population, 0.711 ng m^{-3} of B(a)P$_{eq}$, is slightly higher than the lower limit established by European legislation.

The studies of Petry et al. (1996) and Menichini (1999) based on TEF in different occupational environments and in the atmosphere of a sampling site in a city agglomeration have shown that the contribution of the carcinogenic potency of B(a)P alone is in the range of 27–67% of the activity of the different PAH mixtures. This result underlines and confirms the importance of B(a)P as a surrogate compound for PAH mixtures in air. As can be seen, the B(a)P concentration in our case represents 48% of the

$B(a)P_{eq}$, which is consistent with previously reported results.

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