



Health risk assessment of polycyclic aromatic hydrocarbons (PAHs) adsorbed in PM_{2.5} and PM₁₀ in a region of Arequipa, Peru

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Abstract

The concentrations of PM_{2.5} and PM₁₀, as well as those of the PAHs bound to these particles, were quantified at four sites in the region of Arequipa, Peru, during the year 2018. These samples were collected with high volume samplers, and the concentrations of the PAHs were quantified by liquid chromatography (HPLC). The values found for PM_{2.5} and PM₁₀ at all the sampling sites in Arequipa exceeded the norms established in Peru (50 µg m⁻³ annual average value for PM₁₀ and 25 µg m⁻³ annual average value for PM_{2.5}), with the industrial site presenting the highest values of particulate matter (PM₁₀ max = 235.1 µg m⁻³; PM_{2.5} max = 218.4 µg m⁻³). With respect to seasonality, in the cold season (winter), the concentration of particles was higher compared to the other seasons. Concerning the PAHs, it was found that these had the highest concentrations at the industrial site, followed by the site with high vehicular traffic, with both these sites differing significantly from the rural sites. In addition, at the industrial and high traffic sites, there was a predominance of PAHs with 5 and 6 rings, whereas at the rural sites, PAHs with fewer rings predominated. Finally, the calculated values of lifetime lung cancer risk also revealed a difference between sites with marked emission sources, where irrigation was considered moderate, and the rural sites, where irrigation was considered low. This demonstrated that people living at sites with mobile sources and/or industries had a higher cancer risk compared to the inhabitants of rural sites.

Keywords Particular matter · Polycyclic aromatic hydrocarbons · PM_{2.5} · PM₁₀ · Arequipa · Peru · Health risk assessment

Introduction

In the atmosphere, there is a mixture of suspended solid and liquid particles of different sizes, which have a very characteristic composition depending on the location. Generally, in this particulate matter (PM), we can find sulfates, nitrates, and

ammonium, as well as other inorganic ions such as sodium, potassium, calcium, magnesium, and chloride ions, and also organic and elemental carbon, bark material, water agglomerated in particles, metals (including cadmium, copper, nickel, vanadium, and zinc), and polycyclic aromatic hydrocarbons (PAHs) (WHO 2013). This particulate material is called inhalable when it is less than 10 µm in size and can be deposited in the upper respiratory tract (PM_{2.5-10}) or penetrate the lungs (PM_{2.5}), as well as being able to reach the circulatory system (Sun et al., 2015). According to a large number of toxicological and epidemiological studies (Kim et al., 2015), increases in the levels of particulate matter may have an adverse impact on the health of the population, which is related to factors concerning the chemical composition of the inhalable particulate matter, such as the presence of polycyclic aromatic hydrocarbons (PAHs) and their derivatives adhered to this material (Harrison and Yin, 2000). In fact, PAHs are widespread environmental pollutants, which are mostly formed during the incomplete combustion or pyrolysis of organic matter, but may also be of biogenic origin (Orecchio et al., 2010; Yin and Xu 2018).

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In the environment, PAHs can originate during combustion processes such as forest fires and volcanic eruptions (Rajput and Lakhani, 2009). However, the main source of PAHs is the incomplete combustion of organic products such as crude oil, coal, and natural gas (Ravindra et al., 2008; Slezakova et al., 2010), with the presence of these compounds being of concern in urban atmospheres, mainly due to vehicle traffic emissions (Gaga et al., 2012; Mohanraj et al., 2011), as well as originating from other stationary sources such as domestic heating, industrial processes, waste incineration, and energy production systems, among others (WHO 1998). In addition, the composition and concentration of PAHs may vary depending on the season and region and also on the weather conditions (Longhin et al., 2013), with it having been reported that solar intensity, ambient temperature, humidity, wind speed, and precipitation might have a significant influence on PAHs, depending on the particle concentrations (Tham et al., 2008; He et al., 2014).

Due to their mutagenic and carcinogenic characteristics, many of these compounds are considered to be priority pollutants by several government agencies, including USEPA, IAEA, and IARC. Benzo(a)pyrene (BaP) (listed as a Group 1 carcinogen by the International Agency for Research on Cancer (IARC)) can be used to evaluate the risk of exposure to PAHs, because it has a well-established carcinogenic effect (Callén et al., 2014) that allows us to calculate the value of the lifetime lung cancer risk (LLCR) at different study sites, thereby providing an estimate of the risk to which the population is exposed at particular sites of the study area (Wiriyā et al., 2013; Yu et al., 2008).

The objectives of the present study were the following: (i) to determine the levels of particulate matter (PM_{2.5} and PM₁₀) and the concentrations of PAHs absorbed into the atmosphere at a region of the Peruvian Andes (Arequipa), where no previous information on these compounds is available; (ii) to study the influence of climatological parameters, as well as seasonal variations on the concentrations of PM_{2.5}, PM₁₀, and PAHs; and (iii) to compare the different study sites and evaluate the potential health risk (risk of cancer by inhalation) to the populations living at these sites.

Materials and methods

Area of study

The present study was carried out at a region of the Peruvian Andes in the south of Peru, in the city of Arequipa and its surroundings, which has population of approximately 1 million inhabitants distributed in this mountainous area. The average altitude is 2335 m above sea level, with a minimum of 2041 m and a maximum of 2810 m above sea level. This irregular topography, together with the fact that the city is

located in a valley surrounded by volcanoes, favors the formation of frequent thermal inversions that generally occur in autumn and winter. The climate of the region is desertic and has an average annual rainfall of 100 mm concentrated mainly in summer. Winds occur mainly at night and during the early hours of the day, and there are mountain breezes that present a northeast direction, with valley breezes predominating in a southwest direction in the course of the day. The wind speed throughout the day fluctuates between 1.5 m s⁻¹ and 2.5 m s⁻¹ (Chana, 2003). In the present study, four sampling sites with different environmental characteristics were selected in the city of Arequipa, Peru (Fig. 1), with samples of PM_{2.5} and PM₁₀ being taken.

The selected sites were as follows:

Industrial site (Ind): Located in the northwest of the city in the industrial park of Rio Seco and characterized by the presence of a large number of tanning industries, companies of borate products (whose finished products are intended for agricultural or industrial use), and also laboratories of chemical and pharmaceutical products, among others.

Downtown site (Dow): This sampling site is located at the University of San Agustín, which is in a densely populated area with the presence of public buildings, shops, and businesses. The sampling site is 30 m from Independencia Avenue, which is one of the busiest in the city and has a large amount of both public and private vehicular traffic.

Rural site 1 (R1): Located at Tingo Grande, which is in the southwest of the city. In this area, the inhabitants are predominantly engaged in agriculture, with it being customary to incinerate biomass. This neighborhood is adjacent to a mining company that exploits copper.

Rural site 2 (R2): Located in the Yarabamba neighborhood in the south of the city, with the main economic activity being the manufacture of bricks.

Sample collection

PM_{2.5} and PM₁₀ samples were collected for 24 h in quartz filters (WHATMAN-UK Grade: QMA) using two high volume samplers (HI-vol 3000, Ecotech). Before use, these filters were placed in a stove (Memmert mod. UN) for 4 h at 200 °C and then conditioned for 24 h in a desiccator provided with a constant temperature and humidity control system (20 ± 1 °C and 50 ± 5% relative humidity), after which the filters were weighed three times using an analytical balance (Mettler Toledo, XSE205DU). The weight of the particulate matter was then calculated from the difference between the weight of the filters before and after sampling. Filters were collected from January 8, 2018, to November 15, 2018 (Table S1), with a total of 80 samples of PM_{2.5} (19 samples from the Ind site, 22 samples from the Dow site, 19 samples from the R1 site, and 20 samples from the R2 site) and a total of 75 samples of PM₁₀ being obtained (18 samples from the Ind site, 20

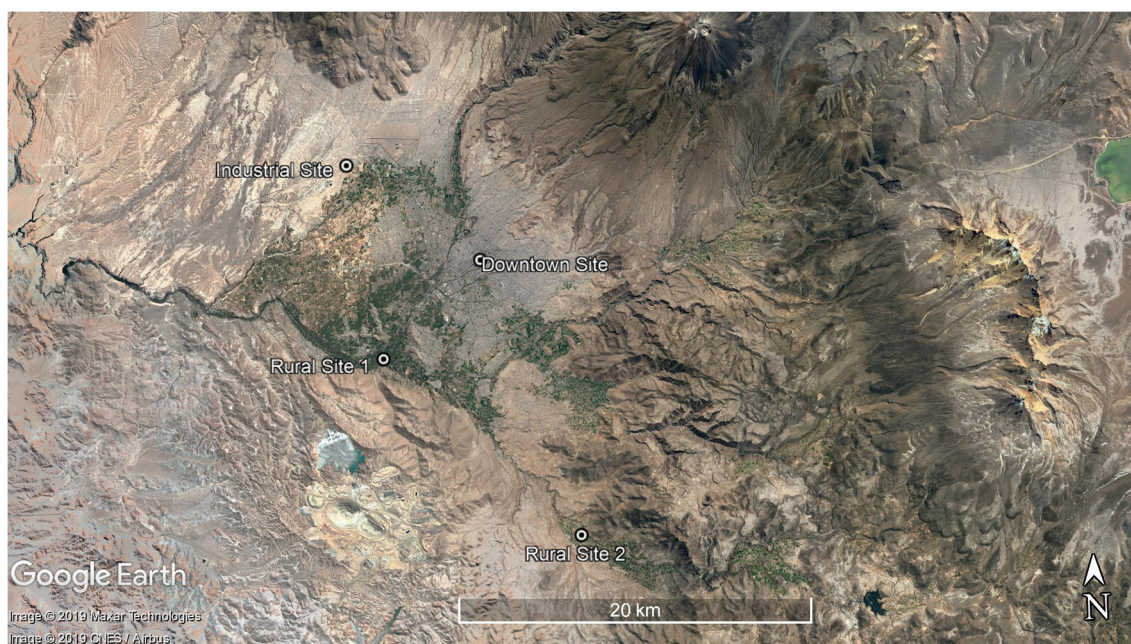


Fig. 1 Study sites in the region of Arequipa, Peru

samples from the Dow site, 18 samples from the R1 site, and 19 samples from the R2 site). Regarding seasonality, 40% of the samples were taken in summer, 22 % in autumn, 14 % in winter, and 24 % of the samples in spring.

Quantification of the PAHs

All the chemicals in this study were used in the form purchased without the need for any additional purification. The solvent (acetonitrile) was of HPLC grade, and the water was purified using a Milli-Q system (Millipore Corp., Bedford, MA). A standard solution of polycyclic aromatic hydrocarbons (PAH calibration Mix, CRM47940) at $10 \mu\text{g mL}^{-1}$ was purchased from Supelco (Sigma Aldrich Argentina), which contained naphthalene (Naph), acenaphthylene (Acpy), acenaphthene (Acp), fluorene (Flu), phenanthrene (Pha), anthracene (Ant), fluoranthene (Fl), pyrene (Pyr), benzo(*a*)anthracene (BaA), chrysene (Chr), benzo[*b*]fluoranthene (BbF), benzo[*k*]fluoranthene (BkF), benzo[*a*]pyrene (BaP), dibenzo[*a,h*]anthracene (DBA), indeno[1,2,3-*c,d*]pyrene (IPY), and benzo[*g,h,i*]perileno (BghiP). The acenaphthylene was not measured because it has no fluorescence. In addition, concentrations of BaA and Chr are reported together as the column was unable to separate the compounds. More diluted solutions in acetonitrile and water (60:40, v/v) were prepared from the commercial solution, which were used to obtain a calibration curve ($0.05\text{--}50 \text{ ng mL}^{-1}$).

Extraction was performed using a method of Santos et al. (2016). First, each quartz filter was cut (5 cm^2) and transferred to an amber glass vial with 1 mL of ACN, after which it was placed in the sonicator (ultrasound TESTLAB TB10, power = 400 W, frequency = 40 kHz) for two periods of 30 min. The

extraction solution was then filtered in a vial using a PTFE syringe filter (porosity of $0.2 \mu\text{m}$), with the extract being analyzed by HPLC (Thermo Fisher, model Ultimate 3000) coupled to a multi-wavelength fluorescence detector. The liquid chromatograph was equipped with an inverse phase column (NC-03 ChromBudget, Germany, 25 cm long \times 3 mm in diameter) at a temperature of $35 \text{ }^\circ\text{C}$, using $25 \mu\text{L}$ as the injection volume and a flow rate of 0.4 mL min^{-1} . The chromatographic run lasted 34 min, starting with a concentration of 70% ACN and attaining 100% ACN at 28 min. Retention times as well as the excitation and emission wavelength values are presented in Table S2, with the values for PAH detection limits being included in the range $0.002\text{--}0.099 \text{ ng mL}^{-1}$. In addition, a recovery percentage was determined for each analyte, by adding a known amount of PAHs to the unused filters and extracting them using the same method. As it was observed that the recovery rate for Naph was very low (45.2%), this compound was excluded from all analyses, while for the other PAHs, their percentages were found to be between 75.4 and 114.4% (Table S2).

Meteorological parameters

The daily values of temperature (T) (maximum, minimum, and mean) ($^\circ\text{C}$), dew point (DP) (maximum) ($^\circ\text{C}$), maximum relative humidity (HR) (%), and maximum wind speed (WS) (km h^{-1}) were obtained at Arequipa Rodriguez Ballón airport (Table S1), in order to examine the influence of these meteorological factors on the atmospheric concentrations of PM_{10} and $\text{PM}_{2.5}$. During sampling days, there was no rainfall, so we were not able to evaluate this meteorological variable.

Health risk assessment

The calculation of equivalence factors was carried out by expressing the toxicity of all PAHs in relation to BaP, which is the most toxic (Nisbet and La Goy, 1992). In this way, a BaP_{eq} is obtained by multiplying the concentration of each PAH by its corresponding TEF (Table S1). In addition, by summing the BaP_{eq} (Σ BaP_{eq}) of all the compounds quantified at a site, the total equivalent toxic concentration of the mixture for that site can be obtained (Nisbet and La Goy, 1992). Then, by multiplying the total BaP_{eq} at each study site by the risk factor of inhalation of BaP (IUR_{BaP}), the lifetime lung cancer risk (LLCR) associated with each site can be determined.

The inhalation risk used was defined by the World Health Organization (WHO, 2000) as 8.7×10^{-5} ng m⁻³ (IUR_{BaP}) for BaP, which implies an incidence of 8.7 cases per 100,000 individuals from chronic inhalation exposure to 1 ng m⁻³ of BaP over a lifetime of 70 years (based on an epidemiological study of coke oven workers in Pennsylvania, Office of Environmental Health Hazard Assessment) (OEHHA, 1994, 2005, 2011, Callén et al., 2014). According to the USEPA, cancer risks can be categorized as very low (LLCR < 10⁻⁶), low (10⁻⁶ ≤ LLCR < 10⁻⁴), moderate (10⁻⁴ ≤ LLCR < 10⁻³), high (10⁻³ ≤ LLCR < 10⁻¹), and very high (LLCR > 10⁻¹) (Zhang et al., 2019).

Statistical analyses

Statistical analyses were performed with InfoStat software coupled to R (Di Rienzo et al., 2018). ANOVA was used for the comparison between sites (spatial) and between the different seasons of the year (temporal), considering a value of $p < 0.05$ to be statistically significant. The assumptions of normal distribution and homoscedasticity were checked before the analysis was carried out. In cases where heteroscedasticity was found, this was modeled in order to be incorporated into ANOVA, with the post hoc LSD Fisher test used to compare the mean values. In the tables, the standard deviations of the concentrations are presented, but not the standard errors reported in the ANOVA model. The Pearson correlation coefficient was used to evaluate the relationship between PM₁₀ and PM_{2.5} and the meteorological parameters.

Results and discussion

Concentration of PM_{2.5} and PM₁₀

At all the sampling sites, the values found for PM_{2.5} and PM₁₀ in Arequipa exceeded the quality standards of these pollutants established in Peru (50 µg m⁻³ annual mean value for PM₁₀ and 25 µg m⁻³ annual mean value for PM_{2.5}) according to the

supreme decree no. 003-2017 of the Ministry of Environment. In fact, 75% of the total samples collected from PM_{2.5} were above the daily mean concentration established in Peru (50 µg m⁻³), and with respect to the PM₁₀ concentration, 48% of the samples collected were above the daily mean concentration established (100 µg m⁻³).

The ANOVA revealed the highest concentrations at the Ind site, which differed significantly from the other sites for both fractions. In addition, for the PM₁₀ fraction, the Dow site differed significantly from the R1 and R2 sites (Table 1). The values of PM_{2.5} and PM₁₀ reported in the present study for the Ind site were higher than those found in some previous studies carried out in Peru. For example, in an urban area of Huancayo, values of 65 ± 31 µg m⁻³ of PM₁₀ and 35 ± 15 µg m⁻³ of PM_{2.5} were reported (Suarez-Salas et al., 2017), whereas at urban sites of the city of Tacna, maximums of 34 and 62 µg m⁻³ were obtained for PM_{2.5} and PM₁₀, respectively (Miranda and Merma, 2017). Moreover, at a zone of high traffic in the city of Cuzco, an average value of 57 µg m⁻³ for PM₁₀ was reported (Vara Licon, 2017).

In the present study, at the Ind site of Arequipa, the values found in the fractions of particulate matter are comparable to those found by other authors in certain large cities, such as in the city of Beijing (100 and 169 µg m⁻³ of PM_{2.5} and PM₁₀, respectively) (Zhang et al., 2008), in the city of Taiyuan (193 µg m⁻³ of PM_{2.5}) (Meng et al., 2007), and in the city of Lahore (190 µg m⁻³ of PM_{2.5}) (Raja et al., 2009). These levels of PM_{2.5} and PM₁₀ may be explained by the presence of a large number of industries including leather tanneries, chemical and pharmaceutical companies, as well contributions afforded by high vehicular traffic at the industrial site.

It was observed that in winter the concentrations of PM_{2.5} and PM₁₀ were significantly higher than in the other seasons. Related to this, there are other studies that have attributed the greater use of winter heating to the presence of higher concentrations of particulate matter in winter (Rasheed et al., 2015, Zhou et al., 2016), especially in European cities where the use of wood for heating can contribute up to 50–70% of the aerosol organic mass during the heating period (Lanz et al., 2007). However, this could not be the cause of the differences in particulate matter in Arequipa, since the minimum temperatures recorded during the winter are approximately 9 °C (Table S3), so it is unusual to use heating in this region.

Effect of meteorological parameters on PM₁₀ and PM_{2.5}

Table 2 presents the correlation analysis between the particle content in the atmosphere of Arequipa and some meteorological parameters (temperature, dew point, relative humidity, and wind speed), where it can be observed that the concentrations of fine particles (PM_{2.5}) correlated negatively with the dew point, with coarse particles also correlating negatively

Table 1 Mean concentrations (\pm standard deviation) of PM_{2.5} and PM₁₀ measured at the different sites in Arequipa, Peru during the year 2018

Site	Summer	Autumn	Winter	Spring	Mean ($\mu\text{g m}^{-3}$)	ANOVA (<i>p</i> value)
PM_{2.5} ($\mu\text{g m}^{-3}$)						
- Ind	115 \pm 82	104 \pm 19 A	171 \pm 24 A	---	128 \pm 67 A	
- Dow	82 \pm 32	50 \pm 11 B	---	73 \pm 13	72 \pm 23 B	
- R1	78 \pm 38	35 \pm 10 B	102 \pm 26 B	72 \pm 13	69 \pm 30 B	
- R2	43 \pm 26	37 \pm 6 B	69 \pm 4 B	59 \pm 25	49 \pm 21 B	
ANOVA (<i>p</i> value)	ns	<i>p</i> < 0.0001	<i>p</i> = 0.0006	ns	<i>p</i> < 0.0001	
Mean + SD ($\mu\text{g m}^{-3}$)	83 \pm 57 b	50 \pm 27 b	126 \pm 52 a	71 \pm 15 b		<i>p</i> = 0.0002
PM₁₀ ($\mu\text{g m}^{-3}$)						
- Ind	150 \pm 37 A	182 \pm 58 A	193 \pm 21 A	---	167 \pm 40 A	
- Dow	113 \pm 12 B	97 \pm 10 A	171 \pm 54 A	93 \pm 9	116 \pm 41 B	
- R1	72 \pm 20 C	52 \pm 19 C	95 \pm 4 B	106 \pm 10	83 \pm 27 C	
- R2	61 \pm 10 C	46 \pm 18 C	55 \pm 5 B	119 \pm 12	64 \pm 28 b C	
ANOVA (<i>p</i> value)	<i>p</i> < 0.0001	<i>p</i> < 0.0001	<i>p</i> = 0.005	ns	<i>p</i> < 0.0001	
Mean + SD ($\mu\text{g m}^{-3}$)	104 \pm 44 b	85 \pm 51 b	146 \pm 63 a	102 \pm 13 b		<i>p</i> = 0.004

Different letters indicate significant differences; ANOVA between seasons in small letters; ANOVA between sites in capital letters. *ns* not significant

with the dew point, as well as with relative humidity and wind speed (Fig. 2). A recent study has mentioned that wind speed correlates with PM levels in the atmosphere in different seasons of the year (both cold and warm) and that an average increase in wind speed decreases the concentration of particles in the atmosphere due to a washing effect.

In Arequipa, the little rain that falls (approximately 100 mm per year) occurs in late spring and summer, so this time of the year is the only time where a washing of particles from the atmosphere can take place due to the effect of these precipitations. Thus, a higher relative humidity is observed in summer, which decays toward winter (Table S3). This may explain the higher concentrations of particulate matter found

in the winter season and the negative correlations between the relative humidity and the dew point with respect to the content of particulate matter in the atmosphere. Related to this, it is known that precipitation favors the wet deposition of particles in the atmosphere, favoring air purification (Wang et al., 2013). According to Zheng et al. (2014), for example, a rain-fall event led to a 56% decrease in PM_{2.5} content in Beijing.

Concentrations of PAHs bound to PM_{2.5} and PM₁₀ at different sampling sites

The concentrations of PAHs bound to PM_{2.5} and PM₁₀ and the ANOVA between sites are presented in Table 3. The highest concentrations of all the PAHs analyzed were found at the industrial site, with the total concentration of PAHs linked to PM_{2.5} and PM₁₀ being significantly higher at the industrial site (Ind), followed by the high-traffic site (Dow), which differed significantly from the rural sites (R1 and R2); these differences between concentrations of PAHs at different sites can also be seen in the study area (Fig. 3).

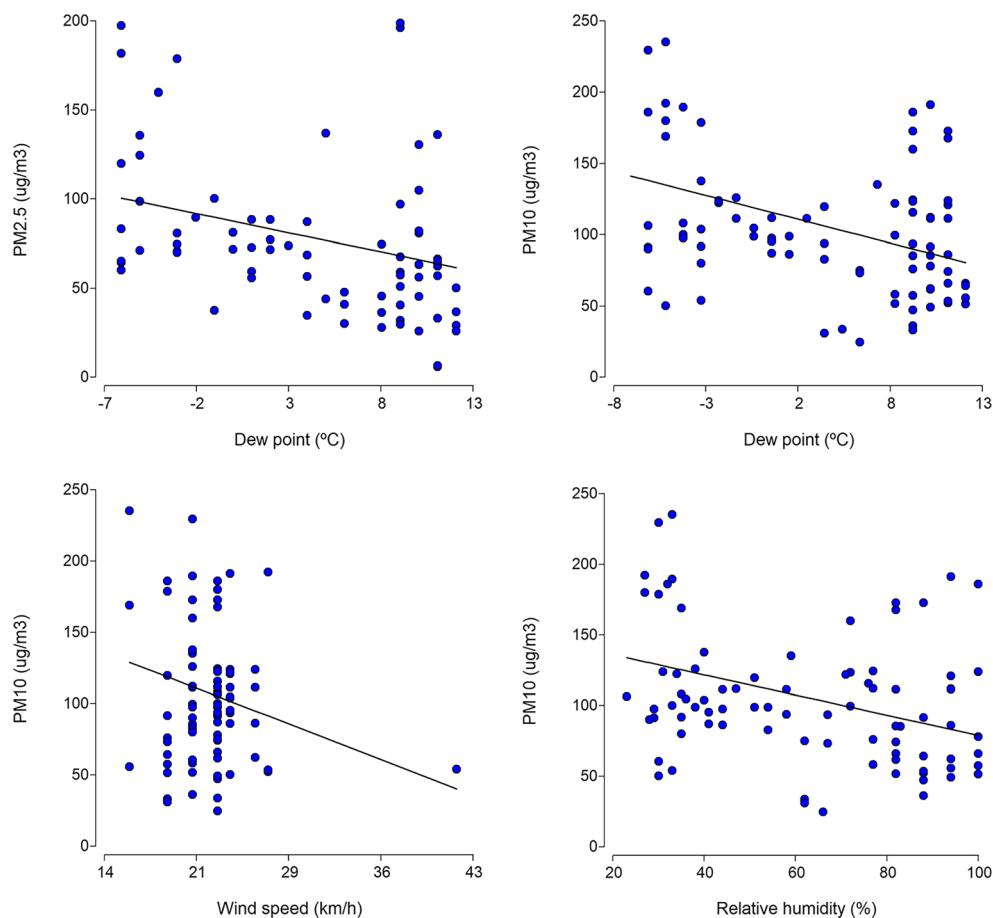
The smallest fraction (PM_{2.5}) comes from fuel processes and vehicle emissions, since, for example, approximately 92% of diesel engine particle emissions are less than 1 μm in diameter. Table 3 shows that for this fraction, the most abundant compound in sites with vehicle sources (Dow and Ind) was BghiP, which is identified as a marker of diesel or gasoline emissions (Teixeira et al., 2013).

Table 2 Pearson correlation coefficients between meteorological parameters and concentrations of PM_{2.5} and PM₁₀ in the atmosphere of the region of Arequipa, Peru, during 2018

	PM _{2.5}	PM ₁₀
T (max)	0.029	0.133
T (mean)	0.016	0.071
T (min)	0.077	- 0.021
Dew point (max)	- 0.280**	- 0.411***
RH (max)	- 0.200	- 0.360***
WS	- 0.004	- 0.215*

T temperature; RH relative humidity; WS wind speed. (**p* < 0.05, ***p* < 0.01, ****p* < 0.001)

Fig. 2 Relationship between particulate matter and meteorological parameters that presented significant correlation in the atmosphere of the region of Arequipa, Peru, during 2018



With respect to the concentration of BaP, the average value quantified during 2018 for the industrial site exceeded the standard value established by EPA (1 ng m^{-3}), both in the $\text{PM}_{2.5}$ fraction (1.9 ng m^{-3}) and in the PM_{10} fraction (2.3 ng m^{-3}). In fact, the values of BaP in the fraction of PM_{10} found in the industrial area were greater than those found in the city of Lima (which is the most important city in Peru in terms of number of inhabitants), where concentrations were reported ranging from 0.3 to 1.4 ng m^{-3} (Del Aguila López, 2018). Other investigations carried out in some Latin American cities have also reported values above the standard set by EPA, such as in Puente Aranda, Bogotá, and Colombia, where concentrations of BaP of up to 5.4 ng m^{-3} in the PM_{10} fraction were obtained (Caballero López and Alvarado Díaz, 2006), and also in the city of Santiago de Chile, with values of 11.1 ng m^{-3} of BaP in the $\text{PM}_{2.5}$ fraction being reported (Kavouras et al., 1999). In our study, the ANOVA showed that the total values of PAHs bound to the $\text{PM}_{2.5}$ and PM_{10} fractions were significantly higher at the Ind site, followed by the Dow site, and with concentrations observed which were well below these values at the rural sites (Table 3). Related to this, the sum of concentrations of PAHs bound to $\text{PM}_{2.5}$ and PM_{10} at the rural sites (ranging from 2.1 to

2.8 ng m^{-3}) was approximately 3 times less than the sum of PAH concentrations at the Dow site (between 7.3 and 9.6 ng m^{-3}) and approximately 6 times less than the sum of PAHs concentrations at the industrial site (ranging from 15 to 20.8 ng m^{-3}). This might demonstrate the greater contributions of vehicular traffic to the concentrations of PAHs in the atmosphere, adding to the contributions from the industrial activities present at the Ind site.

Figure 4 shows the relative composition of the PAHs bound to $\text{PM}_{2.5}$ and PM_{10} for 3 rings (Acp, Flu, Phe, Ant), 4 rings (Fl, Pyr, BaA, Chr), 5 rings (BbF, BkF, BaP, DBA), and 6 rings (IPY, BghiP). According to Hu et al. (2016), the distribution of the different numbers of rings at a site may be related to the sources of emissions present in the area. Our results showed for the different fractions ($\text{PM}_{2.5}$ and PM_{10}) that PAHs with 5 and 6 rings predominated at the Ind and Dow sites, which according to previous studies are markers of vehicular emissions (Tobiszewski and Namieśnik, 2012; Simcik et al., 1999; Park et al., 2002), while the presence of a high percentage of PAHs with 4 rings generally originates from coal combustion (Tobiszewski and Namieśnik, 2012; Lee et al., 2018), as observed in the results obtained for the two rural sites (R1 and R2). Lighter PAHs are generally considered to be produced at low to moderate temperatures, such as in wood and coal

Table 3 Mean concentrations (\pm standard deviation) of PAHs (ng m^{-3}) bound to $\text{PM}_{2.5}$ and PM_{10} at the different sampling sites in the region of Arequipa, Peru, during the year 2018. ANOVA between sites, different letters indicate statistical difference

PAHs	Sites								ANOVA
	Ind		Dow		R1		R2		
(ng m^{-3}) in $\text{PM}_{2.5}$	Mean	SD	Mean	SD	Mean	SD	Mean	SD	
Acp	0.20	0.08	0.16	0.05	0.15	0.06	0.16	0.06	ns
Flu	0.02	0.01	0.02	0.01	0.01	0.01	0.02	0.01	ns
Pha	0.50 a	0.20	0.39 ab	0.12	0.35 ab	0.16	0.38 b	0.15	$p = 0.0224$
Ant	2.5 a	4.5	1.14 ab	0.57	0.59 b	0.26	0.44 b	0.20	$p = 0.0159$
Fl	0.06 a	0.07	0.04 b	0.02	0.03 b	0.02	0.05 b	0.02	$p = 0.0382$
Pyr	0.25 a	0.20	0.19 a	0.07	0.08 b	0.03	0.09 b	0.04	$p < 0.0001$
BaA + Chr	0.57 a	0.77	0.27 b	0.10	0.09 b	0.04	0.08 b	0.03	$p < 0.0001$
BbF	1.8 a	1.7	1.07 b	0.46	0.34 c	0.29	0.21 c	0.05	$p < 0.0001$
BkF	1.0 a	1.1	0.52 b	0.22	0.14 c	0.06	0.09 c	0.02	$p < 0.0001$
BaP	1.9 a	1.7	0.85 b	0.30	0.22 c	0.10	0.15 c	0.03	$p < 0.0001$
DBA	0.26 a	0.28	0.10 b	0.04	0.04 c	0.02	0.02 c	0.01	$p < 0.0001$
IPY	2.3 a	2.0	0.96 b	0.37	0.33 c	0.15	0.17 c	0.05	$p < 0.0001$
BghiP	3.7 a	2.7	1.7 b	0.6	0.44 c	0.19	0.29 c	0.06	$p < 0.0001$
Total	15.0 a	13.9	7.4 b	2.3	2.8 c	1.1	2.1 c	0.4	$p < 0.0001$
in PM_{10}									
Acp	0.23 a	0.16	0.20 b	0.17	0.19 b	0.17	0.14 b	0.10	ns
Flu	0.15	0.23	0.03	0.04	0.02	0.01	0.01	0.01	$p = 0.0002$
Pha	0.49 a	0.23	0.47 ab	0.15	0.36 bc	0.12	0.32 c	0.09	$p = 0.0004$
Ant	5.6 a	7.4	1.8 b	1.6	0.55 b	0.32	0.61 b	0.44	$p < 0.0001$
Fl	0.11 a	0.18	0.07 ab	0.05	0.02 b	0.02	0.05 ab	0.05	$p = 0.0241$
Pyr	0.45 a	0.42	0.31 a	0.19	0.08 b	0.04	0.10 b	0.08	$p < 0.0001$
BaA + Chr	0.82 a	0.92	0.44 b	0.31	0.07 c	0.05	0.09 c	0.08	$p < 0.0001$
BbF	2.5 a	2.0	1.7 a	1.3	0.29 b	0.22	0.24 b	0.11	$p < 0.0001$
BkF	1.3 a	1.2	0.75 b	0.47	0.12 c	0.07	0.11 c	0.09	$p < 0.0001$
BaP	2.3 a	1.9	0.91 b	0.38	0.19 c	0.11	0.16 c	0.10	$p < 0.0001$
DBA	0.52 a	0.75	0.10 b	0.04	0.04 b	0.02	0.04 b	0.07	$p < 0.0001$
IPY	2.7 a	2.0	1.10 b	0.42	0.30 c	0.16	0.18 c	0.08	$p < 0.0001$
BghiP	3.8 a	3.2	1.78 b	0.91	0.38 c	0.16	0.33 c	0.17	$p < 0.0001$
Total	20.8 a	17.1	9.6 b	3.9	2.6 c	1.0	2.3 c	1.2	$p < 0.0001$

combustion (Wang et al., 2011), while PAHs with 5 or 6 rings are generated at higher temperatures, for example, when fuel is burned in engines at high temperatures (Mastral and Callen, 2000). The differences between the rural sites (R1 and R2) and the sites with emission sources (Dow and Ind) in the Arequipa region may explain, on the one hand, the differences between the concentrations of PAHs found in the different sites and, on the other hand, the differences between the composition of the mixture of PAHs according to the number of rings.

Lifetime lung cancer risk (LLCR)

Table 4 shows the values obtained from BaP_{eq} at the different sampling sites and in the fractions studied. The

lowest annual mean values were found at the rural sites, which were between 0.22 and 0.35 ng m^{-3} in the $\text{PM}_{2.5}$ fraction and ranged from 0.26 to 0.31 ng m^{-3} in the PM_{10} fraction. At the Dow site, these values of BaP_{eq} had an annual mean of 1.2 ng m^{-3} in $\text{PM}_{2.5}$ and 1.4 ng m^{-3} in PM_{10} , but the highest BaP_{eq} values were found at the Ind site, which revealed an annual mean of 2.8 ng m^{-3} in the $\text{PM}_{2.5}$ fraction and of 3.6 ng m^{-3} in the PM_{10} fraction, which attained maximum values of 9.4 ng m^{-3} and 11.3 ng m^{-3} , respectively.

These above values are lower than those found in certain cities of China, such as in Xi'an, where a BaP_{eq} range of 2 to 64 ng m^{-3} was reported (Bandowe et al., 2014), and in Beijing city, where

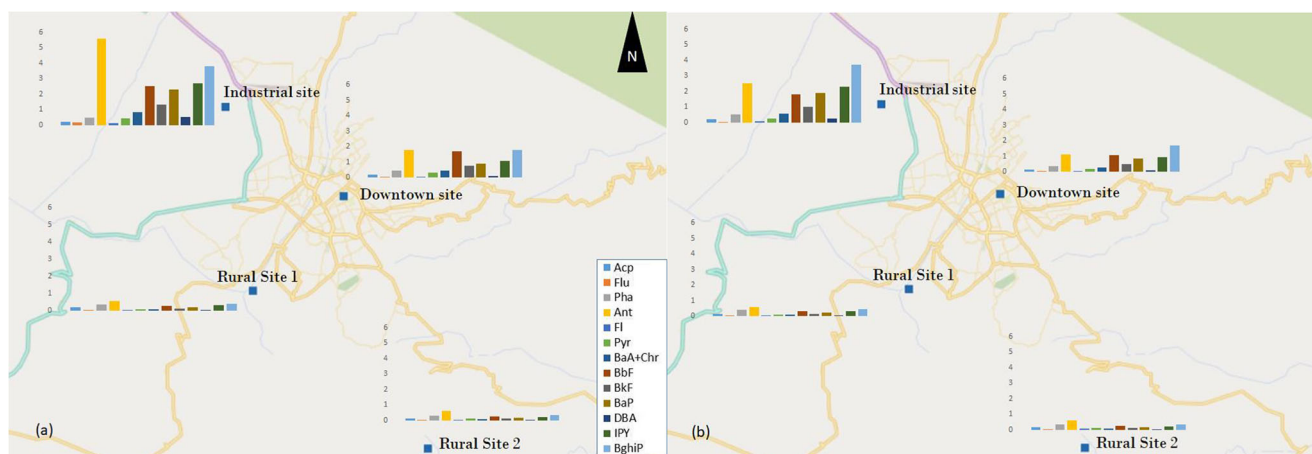


Fig. 3 Mean concentrations of PAHs (ng m^{-3}) bound to PM_{10} (a) and $\text{PM}_{2.5}$ (b) at the different sampling sites in the region of Arequipa, Peru, during the year 2018

values between 3 and 26 ng m^{-3} (Jai et al., 2011) were found. In addition, in Middle Eastern cities such as Tehran, Iran (Taghvaei et al., 2018), and the city of Zonguldak, Turkey (Akyüs and Cabuk, 2008), values were reported of $29.1 \pm 5.7 \text{ ng m}^{-3}$ and $22.5 \pm$

4.2 ng m^{-3} , respectively. The LLCR calculations obtained in Arequipa for the two fractions studied (Fig. 5) showed a well-differentiated risk between the Ind and Dow sites, where the risk of lung cancer by inhalation is moderate, and on the other hand at the rural sites (R1 and R2), where the risk of lung cancer by inhalation of particles with PAHs can be considered low. Moreover, the LLCR values at the two sites with greater sources of contamination (Ind and Dow) were particularly higher in the autumn season (Table S4).

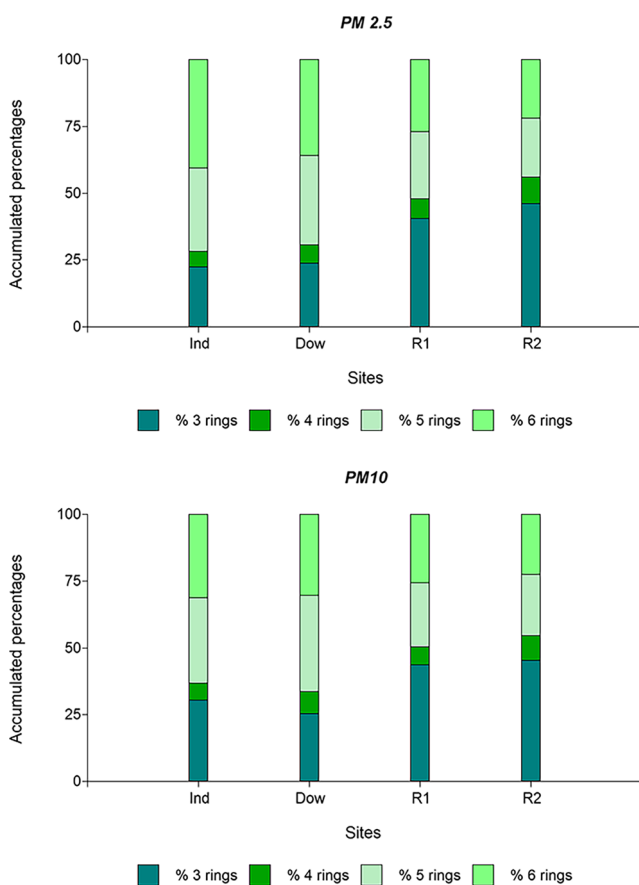


Fig. 4 Accumulated percentages of the concentrations of PAHs in $\text{PM}_{2.5}$ and PM_{10} according to the number of rings at the different study sites in the region of Arequipa, Peru

Conclusions

In the present study, the concentrations of $\text{PM}_{2.5}$ and PM_{10} particles and the associated content of PAHs were determined at four sites in the city of Arequipa, Peru. The concentration of particulate matter in the two fractions studied was the highest at the industrial site, followed by the downtown site, with the lowest being at the rural sites. With respect to seasonality, in the cold season (winter), the concentration of particles was higher compared to the other seasons.

The concentrations of PAHs found were significantly higher at industrial sites and those having high traffic, with the average annual concentration of BaP at the industrial site exceeding 1 ng m^{-3} for the two fractions studied. Moreover, it was observed that at the Ind and Dow sites, the heaviest PAHs predominated, with 5 or 6 benzene rings, in contrast with the rural sites where the percentage of lighter PAHs was higher.

With respect to the lifetime lung cancer risk values, at rural sites, the risk of lung cancer by inhalation was found to be low ($10^{-6} \leq \text{LLCR} < 10^{-4}$), whereas at the Ind and Dow sites, the values obtained indicated a moderate risk of cancer ($10^{-4} \leq \text{LLCR} < 10^{-3}$).

Table 4 Mean concentrations (\pm standard deviation) and range (minimum–maximum) of BaP_{eq} in PM_{2.5} and PM₁₀ measured at the different sampling sites in the region of Arequipa, Peru, during the year 2018

Site	Summer	Autumn	Winter	Spring	Mean (ng m ⁻³)	Range (min–max)
BaP_{eq} in PM_{2.5} (ng m⁻³)						
- Ind	2.7 \pm 2.3	4.7 \pm 4.3	1.7 \pm 1.0	---	2.8 \pm 2.5	0.4–9.4
- Dow	1.1 \pm 0.6	1.6 \pm 0.4	---	1.2 \pm 0.3	1.2 \pm 0.4	0.5–2.4
- R1	0.32 \pm 0.14	0.29 \pm 0.10	0.53 \pm 0.33	0.38 \pm 0.16	0.35 \pm 0.16	0.15–0.76
- R2	0.24 \pm 0.03	0.20 \pm 0.06	0.22 \pm 0.04	0.23 \pm 0.03	0.22 \pm 0.04	0.13–0.29
BaP_{eq} in PM₁₀ (ng m⁻³)						
- Ind	3.7 \pm 2.8	6.1 \pm 4.8	1.7 \pm 0.9	---	3.6 \pm 3.1	0.4–11.2
- Dow	0.9 \pm 0.3	1.5 \pm 0.5	2.0 \pm 0.4	1.4 \pm 0.4	1.4 \pm 0.6	0.6–2.2
- R1	0.23 \pm 0.10	0.31 \pm 0.13	0.21 \pm 0.07	0.42 \pm 0.21	0.31 \pm 0.17	0.09–0.90
- R2	0.30 \pm 0.32	0.21 \pm 0.06	0.20 \pm 0.01	0.33 \pm 0.04	0.26 \pm 0.19	0.10–1.00

The results obtained in the present study showed the influence of the sources of emissions at the industrial sites and at those sites with high traffic with respect to rural sites, not only in terms of the amount of particles

but also in the greater risk of cancer produced in the population living at these sites.

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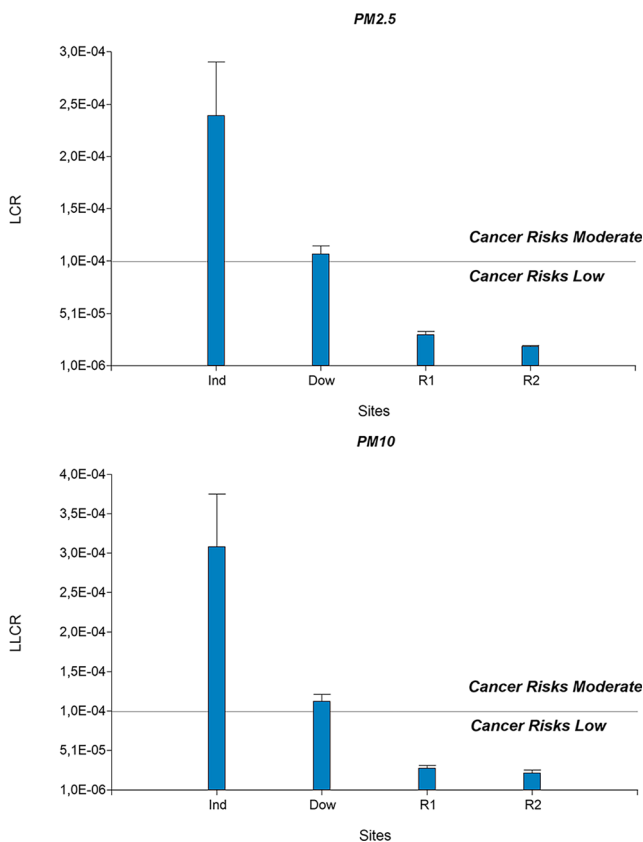


Fig. 5 Lifetime lung cancer risk estimation (LLCR) in fractions PM_{2.5} and PM₁₀ at the different study sites in the region of Arequipa, Perú

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