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Behavior of Atmospheric ^{210}Pb Concentrations and Their Correlations with Meteorological Parameters in Abidjan

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Abstract Monitoring of radionuclides in aerosols is essential for understanding the factors influencing temporal variations in atmospheric deposition of elements such as ^{210}Pb, which is frequently used as a tracer in atmospheric studies. However, long-term monitoring remains limited. Very few studies on air radioactivity are available in Africa and particularly in Côte d'Ivoire. In this study, we measured the radioactivity of ^{210}Pb in surface aerosols collected on a weekly basis in the city of Abidjan in south–eastern Côte d'Ivoire from March 2022 to February 2023, using HPGe gamma spectrometry. The mean activity of ^{210}Pb was 0.78 ± 0.2 mBq/m^3. This activity concentration showed strong seasonal variations, with maximum concentrations recorded during the dry seasons and minimum in the rainy seasons. Statistically significant correlations were observed between ^{210}Pb concentration and temperature (r = 0.47; p-value = 0.02), as well as a negative correlation with precipitation (r= -0.48; p-value = 0.02). A statistical test comparing the data obtained in the present work with some bibliographic values of ^{210}Pb in aerosols showed that our level of activity concentration is statistically identical to the other previous studies carried out in the world. The active dependence of ^{210}Pb with temperature and precipitation was investigated through simple linear regression analysis. Model evaluation and comparison functions were also used. Overall, this study provides an understanding of the behavior, correlations and variability of observed ^{210}Pb concentrations influenced by atmospheric conditions in our study area.

Keywords: *^{210}Pb, Aerosol, Coastal city, Environmental radioactivity*

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1. Introduction

It is generally accepted that our environment, including the atmosphere, is slightly radioactive. The atmosphere is also a very efficient medium for the transfer of natural and man-made radionuclides, including some harmful substances in air, from one place to another. ^{210}Pb is a naturally occurring radionuclide produced by the decay of ^{222}Rn, which in turn is derived from the ^{238}U decay chain. Together with other descendants of ^{222}Rn ($T_{1/2}$ = 3.82 d), this radionuclide has a significant impact on the effective dose of ionizing radiation received by humans by inhalation and ingestion [1]. However, it is important to note that the presence of ^{210}Pb in atmospheric air is often associated with other factors, such as emissions from factories or human activities, in addition to natural sources. The concentration of ^{210}Pb in air can vary depending on these factors, but it is generally present at relatively low levels and can be used as an indicator for scientific research and air quality monitoring. The global inventory of measured ^{210}Pb deposition flux is estimated to be about 3.77 × 10^{16} Bq/y, i.e., 3.8 × 10^{25} atoms/y, which agrees well with the global emission flux inventory of 4.65×10^{25} atoms/y (deduced from the activity flux: 9.79 × 10^{19} Bq/y) for ^{222}Rn [2,3]. The atmospheric residence time of ^{210}Pb in the atmosphere is usually estimated using the activity ratios of its progeny ^{210}Bi and ^{210}Po [3,4]. Once ^{210}Pb is formed, it rapidly associates primarily with submicron aerosol particles that contribute significantly to the formation and growth of aerosols in accumulation mode, characterized by a diameter ranging from 0.07 to 2 µm, which represent an important reservoir of air pollutants. Then the fate of ^{210}Pb is thus closely linked to that of carrier aerosols [5]. Studies by [5,6,7] revealed that long-lived ^{210}Pb is almost entirely absorbed by aerosol particles in accumulation mode, with an average activity median aerodynamic diameter (AMAD) particle size of 0.55 µm. These aerosols containing radionuclides are then removed from the atmosphere by dry and wet deposition processes, the latter being the predominant mechanism for the removal of these radionuclides from the atmosphere. Long-term measurements of these radionuclides provide valuable information on atmospheric processes, with ^{210}Pb serving as a tracer for different aerosol species [8]. With

its full energy gamma peak of 46.5 keV, ^{210}Pb is easily detectable by non-destructive gamma spectrometry when collected in the air on an aerosol filter. Given the large regional and temporal variations of ^{210}Pb, it is important to assess these variations in different geographical contexts. This will also be very useful for specific tracer applications in regional studies, such as atmospheric mixing and transport. In Africa and especially in its tropical zone, there is insufficient data on measurements of ^{210}Pb in air at ground level. The present study was undertaken with $PM_{2.5}$ airborne as ^{210}Pb is associated with fine particles. Moreover, Huang et al. found that $PM_{2.5}$ can explain more than 50% of the variation in ^{210}Pb in aerosols [9]. The main objective of this study is providing more in-depth information on the factors responsible for temporal variations in concentrations.

More specifically, this study aims to:

- investigate the temporal variations of ^{210}Pb in the study site environment;
- investigate the correlation of ^{210}Pb concentrations with local climatic conditions;

create a model that takes into account the meteorological conditions in order to predict the behavior of ^{210}Pb concentrations.

2. Material and Methods

2.1. Study Area

Samples of aerosol particles in air were taken at a height of 15 m above the ground to minimize the number of dust particles entering the sampler. The sampling station was located at the Faculty of Sciences and Technology of the University Félix Houphouët–Boigny (located at longitude “3°58' 57"” West–“5°21'23"” North) (Figure 1) in the commune of Cocody in the city of Abidjan. Abidjan is the economic capital of Côte d’Ivoire and is located in the south-east at the edge of the Gulf of Guinea at an altitude of 18 m above sea level. Côte d'Ivoire is the transition zone between the humid equatorial climate and the dry tropical climate, which allows the country to be divided into two main zones: the south and the north. The difference between these two zones lies in the humidity level, which can sometimes be around 100 % in the south and as low as 20 % in the north during harmattan weather (cool, dry wind from the Sahara). In the south, below Yamoussoukro, the political capital of Côte d’Ivoire, the climate is equatorial and therefore very humid. This climate zone is characterized by high rainfall and temperatures around 30°C. The north is dominated by the tropical climate of the savannahs with average temperatures ranging from 28° to 37° Celsius. This climatic region also experiences a great rainy season and a great dry season [10].

In the south, there are four seasons:

- the long rainy season: from April to mid-July, with frequent rainfall and numerous storms;
- the short dry season: from mid-July to September, when the sky remains open;
- the short rainy season: from September to November, with some light rainfall;
- the long dry season: from December to March, marked by the northern trade winds (harmattan).

Meteorological data, including rainfall, air temperature, wind speed, and direction, were obtained on site by a wireless VANTAGE PRO 2 weather station.

2.2. Air Sampling and Determination of ^{210}Pb Activity Concentrations

Aerosol samples (particle size ≤ 2.5 µm) were collected by drawing atmospheric air through a glass microfiber filter (GF 10 047) with a diameter of 47 mm using a Sven Leckel low volume air sampler with a flow rate of 3 m^3/h for 168 hours. The sampling program was conducted over one year (2022–2023), with two samples per month. A total of 24 samples were taken from the measurement site with a total volume of air sampled of 504 m^3 per sample. Aerosol samples were measured with a Canberra GX3018 hyper-pure (50% relative efficiency p-type) germanium detector placed in a 7.5 cm lead-thick enclosure and covered inside with thick copper plates, 10 mm on the side walls and 1 mm on the upper and lower walls. The counting electronics is a Canberra Lynx. The obtained data were analyzed by the Genie 2000 software. The FWHM of the detector was 2.00 keV at 1.33 MeV line of ^{60}Co. The active acquisition time of the gamma spectra of the aerosol samples was 86,400 seconds per sample. The concentration activity of ^{210}Pb was calculated by spectrum deconvolution of the 46.5 keV peak. A priori measurement of blank filters for aerosol sampling showed the absence of ^{210}Pb activity in the filters. The efficiency calibration was carried out with two Bernard Damas standard filters ref C569 of 51 mm diameter, charged, one by a multi-gamma source consisting of 11 radionuclides covering an energy range from 59.6 keV (^{241}Am) to 1836 keV (^{88}Y) and the other in ^{129}I for the energy ray 29.7 keV. A yield transfer to a 47 mm filter was carried out using the Monte Carlo GESPECOR calculation code [11]. The activity concentration of ^{210}Pb is determined by the following relationship [3]:

$$A_{Pb-210} = \frac{N}{t}\left(\frac{1}{v \times \varepsilon \times BR}\right)$$

Where N: total counts under the photopeak of ^{210}Pb;

t: counting time;

V: volume of air passed through the filter paper (m^3);

ε: photopeak efficiency for the detector;

BR: branching ratio of the radionuclide of interest (4% for ^{210}Pb).

The above formula assumes that the decay during counting is negligible, taking into account that the half-life of ^{210}Pb is very large compared to the counting and sampling times. The detection limit for all filters varies from 0.11 to 0.2 Bq. In this study, the statistical analysis of the data, the description of the ^{210}Pb activity concentrations, the tests of the linear correlation coefficients and the linear regression models were carried out using the R computational environment and its graphical user interface RStudio [12]. Meteorological data (precipitations, daily temperatures, and relative humidity) to identify possible correlations were collected using the VANTAGE PRO 2 weather station at the sampling site.

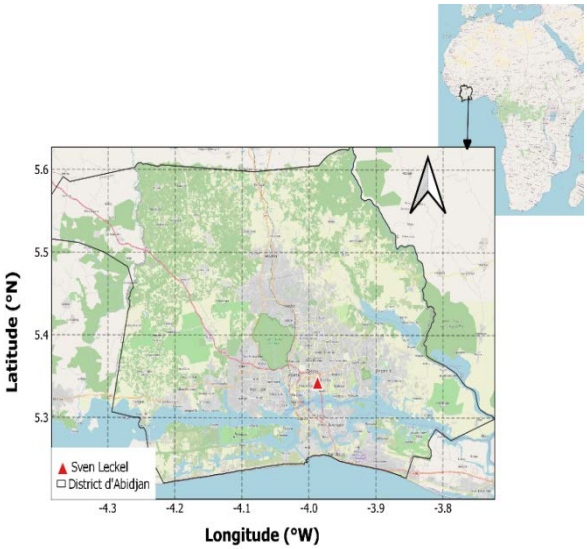

Figure 1. Sampling location in Abidjan (Cote d'Ivoire)

3. Results and Discussion

3.1. Meteorological Conditions During the Sampling Period

Average weekly temperatures ranged from 24.61°C to 29.17°C from March 2022 to February 2023. The lowest temperature was recorded in September 2022, while the highest was observed in February 2023, reaching 29.17°C. As far as precipitation is concerned, we recorded a variation ranging from 0 mm to 273 mm in June 2022, this month being characterized as the wettest of the whole year.

3.2. Temporal Variations ^{210}Pb Activity

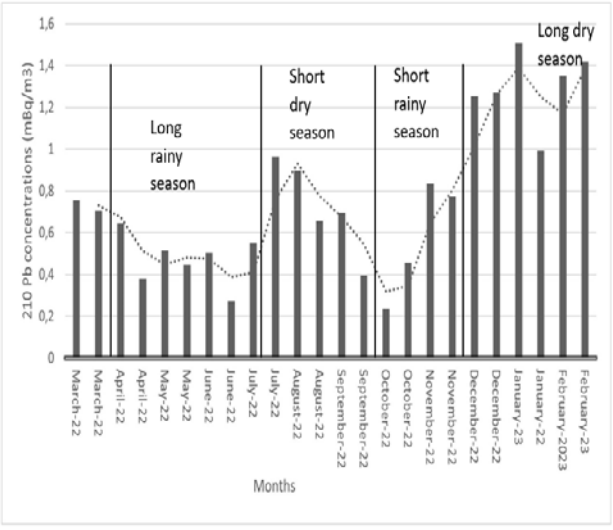

Figure 2. Monthly variation of ^{210}Pb activity (mBq/m^3) in aerosol

Activity concentrations of ^{210}Pb in aerosol filters ranged from 0.24 to 1.5 mBq/m^3, with an average of 0.78 ± 0.20 mBq/m^3. The lowest concentration was observed in October 2022, while the highest concentrations were recorded from December 2022 to January 2023 (Figure 2). This variability resulted in minimum levels of concentration during high precipitation seasons. The highest values were recorded from December to February and mid-July to September, corresponding to the dry periods of the year.

3.3. The ^{210}Pb Activity Concentration Levels in Abidjan

Our mean value (Table 1) fits harmoniously within the range of values recorded by [13], who collected data on activity concentrations of ^{210}Pb in ambient air worldwide, observing levels ranging from 0 to 1 mBq/m^3. It should be noted that the highest concentrations of ^{210}Pb were recorded in subtropical and temperate latitudes of the Northern Hemisphere, a finding supported by sources such as [9,14].

Table 1. Variation in ^{210}Pb Activity concentration (mBq/m^3) in the surface air around the word

Location	Activity range	Average activity	Data source
Xiamen (China)	0.17 – 3.31	1.26 ± 0.78	[9]
Detroit (USA)	0.30–4.22	1.2	[4]
Belgrade	0.30–3.17	1.2	[33]
Malaga (Spain)	0.28–0.92	0.54	[23]
Granada (Spain)	0.2–1.1	0.57	[30]
Milan (Italy)	0.22–1.31	0.7	[1]
Mangalore (India)	0.13–3.4	1.1 ± 0.73	[3]
Kaïga (India)	0.3–3.4	1.3 ± 0.73	[3]
Uk (Edinburgh)	0.1–0.74	0.21 ± 0.01	[21]
Japan (Tsukuba)	0.2–0.8	0.45	[32]
Lisbon	0.036–0.524	0.181 ± 0.111	[31]
Estonia	0.08–2.53	0.43	[20]
Pakistan (Islamabad)	0.056–0.76	0.284 ± 0.15	[8]
El-Minia (Egypt)	N.A.	1.2 ± 0.15	[29]
Abidjan (Côte d'Ivoire)	0.24–1.51	0.78 ± 0.2	**This study**

3.4. Study of Distribution of ^{210}Pb Activity Concentration

The overall results of the activity concentration of ^{210}Pb measured at our site were the subject of an in-depth statistical analysis. The Shapiro–Wilk normality test was used to assess the concentrations obtained from our 24 samples. This test reveals that our data set follows a normal distribution with $p = 0.14 > 0.05$. This is corroborated by the Kolmogorov–Smirnov test with $p = 0.8 > 0.05$. These results, which are of high statistical significance, with a 95% confidence interval, confirm the relevance of the mean of 0.78 ± 0.2 mBq/m^3 for our dataset. (Table 2) illustrates the statistics of distribution concentrations.

Table 2. statistics of ^{210}Pb activity concentrations

Radionuclide	Quartiles					Mean	Uncertainty
^{210}Pb	Min.	1st	Median	3rd	Max.	μ	σ
	0.24	0.49	0.70	0.97	1.51	0.78	0.20

We compared our theoretical mean value with averages obtained in other regions of the world (Table 1) using a student test with welch correction. This test with

p = 0.30 > 0.05, shows that the activity concentration level of ^{210}Pb at our study site is statistically similar to the averages of previous studies conducted worldwide. The confidence interval for the obtained mean value m is defined as follows: $m \pm \frac{t_{0.95}}{\sqrt{n-1}}$ [8] with $t_{0.95}$ = 1 and a standard deviation of 0.20 mBq/m^3. Overall, we can statistically state that the activity level in ^{210}Pb concentration is between 0.58 and 0.98 mBq/m^3. This result is consistent with the range of concentrations obtained in several previous studies.

3.5. Seasonal Variation of ^{210}Pb Activity

At the sampling site, ^{210}Pb activity concentrations demonstrated seasonal variation, this result is similar to the finding of many previous studies in other coastal regions [3,9,15]. Changes in ^{210}Pb activity depend on the rate of local radon emanation, and meteorological parameters, particularly temperature, precipitation, and moisture content of air and soil [16,17]. Our measurement site is located in the intertropical zone of confluence between two air masses, the monsoon (humid) and the boreal hemisphere (dry) [18]. ^{210}Pb concentrations are highest during the months of the great dry season (December-March) corresponding to the northern hemisphere winter and lower during the rainy season (April-July) in summer of northern hemisphere climates. These general trends have also been observed by [1,3,5,15,19,20]. During rainy periods, unstable southwesterly (monsoon) oceanic air masses cause vertical developing cloud formations that result in heavy precipitation [18]. The purifying effect of this precipitation on ambient air is responsible for the substantial decrease in aerosol concentrations and thus those of ^{210}Pb [3,21]. Studies by [22,23,24] on radioactivity in ambient air noted the same finding. The influence of precipitation on the sweep and removal of ^{210}Pb atoms from the air during dry and rainy seasons was studied by one-factor analysis of variance (ANOVA). The result of this statistical test, illustrated in (Figure 3), shows that the average value, 0.98 mBq/m^3, of the dry season is statistically different from 0.48 mBq/m^3 of the rainy season p-value = 3.20 ×10^{-5} < 0.05.

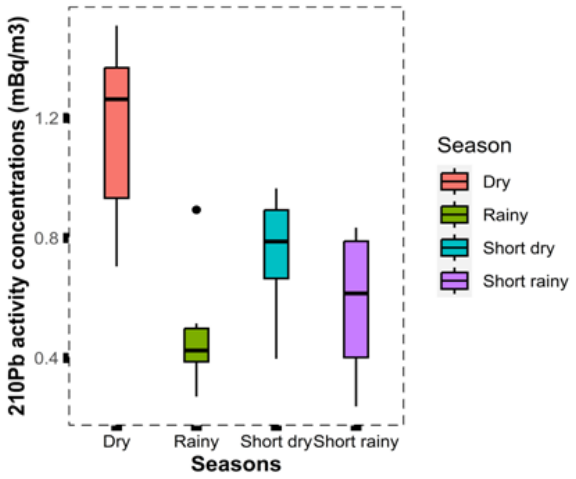

Figure 3. ^{210}Pb boxplot activity for dry and rainy season

The leaching of ^{210}Pb aerosols by rainwater is supported by the results of numerous studies that have found significant correlations between ^{210}Pb deposition fluxes in rainwater and precipitation amounts in Geneva [25] or Taiwan [26]. Dry periods of the year, characterized by the arrival of continental air masses such as the boreal hemisphere trade winds (harmattan) from the northeast, are responsible for a significant increase in ^{210}Pb concentrations in the atmosphere [18]. During these periods, tropospheric aerosol particles, carrying ^{210}Pb nuclei, from arid areas accumulate in the atmospheric layer. This seasonal behavior of ^{210}Pb could be explained by its origin, namely ^{222}Rn, which has higher concentrations during dry periods than during the rainy season [27]. In addition, temperature inversion conditions in ambient air may impede vertical mixing of ^{222}Rn, contributing to increased concentrations of ^{210}Pb atoms at the soil surface [15]. Low rainfall [2] and air mixing characteristics in the upper layers of the atmosphere promote prolonged residence time of aerosols in the atmosphere, resulting in continuous accumulation of ^{210}Pb [28]. Figure 4 illustrate the variations in ^{210}Pb concentrations as function of rainfall during the sampling period.

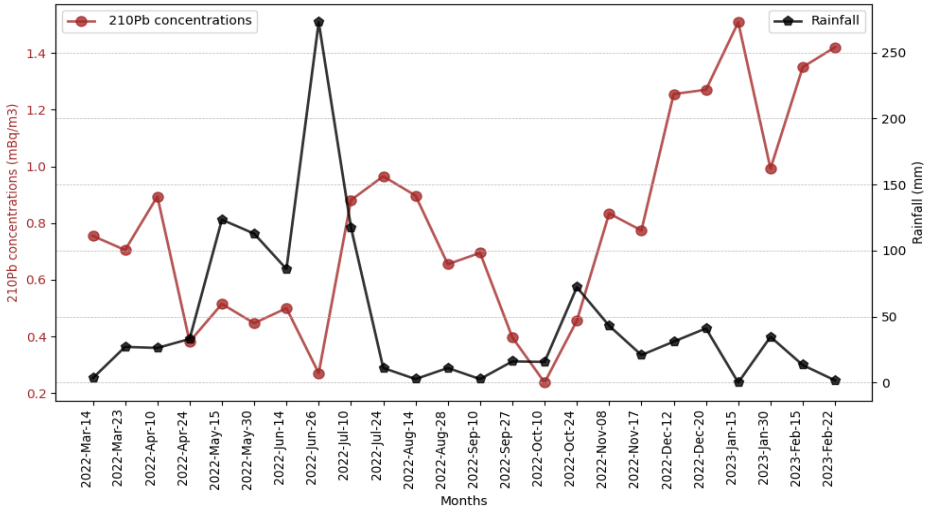

Figure 4. Evolution of ^{210}Pb as a function of rainfall

The process of leaching atmospheric aerosols by precipitation results in a decrease in the activity concentrations of ^{210}Pb in surface air. A negative correlation between these two parameters can therefore be expected as shown by the Pearson rank correlation coefficient (Table 3). Correlations between ^{210}Pb concentrations and precipitation, temperature and humidity were studied. We obtained the correlation coefficients for these parameters as -0.48, 0.47 and -0.06. Although some authors have observed opposite correlations for temperature and humidity [20], the main factor influencing the decrease remains abundant precipitation, which is negatively correlated with ^{210}Pb concentrations [3,5,9,20]. The (Figure 5) highlights a correlation between ^{210}Pb concentrations, rainfall, and temperature. More specifically, periods of rainfall between 0 and 50 mm coincide with elevated temperature, indicating simultaneous positive and negative correlation.

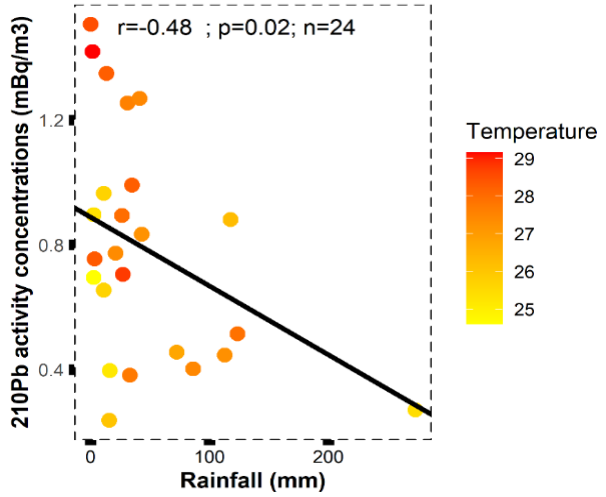

Figure 5. Relationship between ^{210}Pb concentrations, rainfall and temperature

3.6. Simple Linears Models

The linear relationship between ^{210}Pb and temperature, as predicted by Model 2, was confirmed by the p-value test ($p = 1.9 \times 10^{-10} < 0.05$), demonstrating the great significance of this relationship. In addition, the percentage variance ($r^2 = 0.83$) indicates that 83% of the temporal variability of ^{210}Pb is explained by ambient temperature. For the use of precipitation as an explanatory variable for ^{210}Pb, the coefficients of Model 1 were found to be largely significant, and this was chosen for statistical reasons ($r^2 = 0.20$; $p = 4.2 \times 10^{-10} < 0.05$) (Table 3). However, the low variance of Model 1 suggests that it would be wise to introduce other explanatory variables (multiple linear regression) into the model to improve its predictive capacity. This paves the way for further modelling of ^{210}Pb using nonlinear models with precipitation. The evaluation of the models ends with a residue analysis, which checks whether the fundamental assumptions of the linear model are violated. The Jarque and Bera non-normality test applied to the residuals of models 1 ($p = 0.74 > 0.05$) and model 2 ($p = 0.59 > 0.05$) generated p values ($p > 0.05$), meaning that there is no reason to conclude that the residues do not follow a normal distribution.

Finally, the homoscedasticity test Breusch-Pagan test generated a p-value ($p = 0.16 > 0.05$; Model 1) and ($p = 0.25 > 0.05$; Model 2). Therefore, we can conclude that the residuals have minimal variance, which confirms the statistical validity of the different models.

Table 3. Summary of the most significant linear models for ^{210}Pb predictions

Models	R^2	p-value <0.05
(1): ^{210}Pb = 0.932 – 0.002R	0.20	4.2×10^{-10}
(2): ^{210}Pb = 0.031T	0.83	1.9×10^{-10}
Meteorological parameters	^{210}Pb	
	r	p-value
Rainfall (mm)	-0.48	0.02
Temperature (°c)	0.47	0.02
Relative humidity	-0.06	0.76

4. Conclusion

Sampling was conducted twice a month over a one-year period. During this period, the concentration of ^{210}Pb ranged from 0.24 to 1.51 mBq/m^3, with an annual average of 0.78 mBq/m^3. Student's statistical test applied to the data estimated the activity level in ^{210}Pb concentration, between 0.56 and 0.98 mBq/m^3. These values are consistent with those reported in the scientific literature for this radionuclide. The ^{210}Pb activity concentration showed a strong seasonal variation. Indeed, the highest concentrations were observed during the dry seasons of the year. Conversely, rainy seasons have reduced concentrations of aerosols containing the radionuclide ^{210}Pb in ambient air. Precipitation, by removing this radionuclide from the atmosphere, plays an essential role in the temporal variation of ^{210}Pb, which exhibit a negative correlation with precipitation intensity and a positive correlation with temperature. Therefore, these two parameters play a role in the seasonal variation of ^{210}Pb. Several linear prediction models have been developed to try to explain this temporal variation. However, this variability cannot be fully explained by a simple linear equation, which instead requires the use of multiple linear regressions taking into account other meteorological parameters, or even the exploration of nonlinear models.

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