Analysis of gross alpha, gross beta activities and ^{7}Be concentrations in surface air and statistical prediction model

F. Arkian[1] [a], **M. Salahinejad** [b], **J. Amidi** [a]

[a] National Radiation Protection Department, Tehran, Iran
[b] Nuclear Research Center, Tehran, Iran
P. O. Box: ٤٤٩٤-١٤١٥٥ Tehran, Iran.
Email:farkian@aeoi.org.ir

Abstract

Measurement of gross α, gross β activities and cosmogenic beryllium-٧ (^7Be) concentrations were made both daily and weekly during ٢٠٠١, ٢٠٠٢, ٢٠٠٣ and ٢٠٠٤ from samples of atmospheric aerosols filtered from the air at Tehran Nuclear Research Center (٣٥ ^{o} ٤١' N) and Zahedan (٢٨ ^{o} ٢٩' N). The data are sufficiently numerous to allow us to examine variations in time and through these measurements we have established atmosphere. The air concentrations of gross β and gross α activities and ^7Be concentration displayed a lognormal distribution during the study period. Both of β activities and ^7Be have maximum concentrations during warm mid-year months. In this work, the data on concentration and meteorological data have been made use of in order to determine a model for gross α and gross β and ^7Be, respectively. The model can be used to estimate that part of the trend in gross β and in ^7Be levels that can be accounted for by trends in local meteorology. We didn't find any relation between gross α and meteorology parameters. A satisfactory agreement between the results of the model and the measurements was highlighted.

Key words: ^7Be concentrations, gross β activity, gross α activity, meteorology parameters

١.Introduction

Radioactivity in the atmosphere originates from natural radioactive decay, cosmogenic production and from nuclear weapon testing and nuclear accidents. Inhalation is one of pathways by which radioactive nuclides are incorporated into the human body. In our laboratory, we are continuously measuring the radioactivity in the surface air. It is not purpose of this paper to predict the impact of specific incremental radiation exposure arising from human activities of population but with information base provided, national and international bodies may be able to select appropriate criteria for the radiological protection of natural population communities and ecosystem.

Result of measured gamma emitting nuclides show that cosmogenic ^7Be is the only nuclide found in the air of Iran. ^7Be ($t_{1/2} = 53\,d$) is one of radioactive products of the bombardment of the atmosphere by cosmic rays. About ٧٥٪ of ^7Be is produced in the stratosphere and ٢٥٪ in upper troposphere (Johnson and Viezee, ١٩٨١). Once ٧ Be is formed in the troposphere it rapidly associates primarily with sub micron sized aerosol particles. ٧Be in these aerosols may subsequently enter the marine as well as terrestrial and vegetation environment via wet or dry deposition events (Papastefenou, ١٩٩٤).

In this paper we report four years of continuous measurements of gross α and gross β activities and concentration of ٧Be in surface air. Using these data, the present research was undertaken with the following principal goals:

a) To perceive the variations of gross α and gross β and ٧Be concentration in Tehran station and compare to Zahedan station.
b) To identify the main meteorological parameters which are responsible for the variation of those concentrations.
c) To model the data obtained from our sampling site as a function of meteorological parameters such as temperature, precipitation, wind speed relative humidity, etc. With the regression model we believe that we can estimate the ground level concentration associated with changes in meteorological conditions.
d)

[1] Corresponding author

٢. Experimental procedures

Airborne dust samples were collected daily in fiber glass filters, ٤٧ mm diameter (collection efficiency ٩٩,٩٩٪ for ٠,٨ pore size) with an air sampler at a flow rate ٣٩ m^3 day^{-1} and weekly aerosol samples have been collected with high-volume air sampler in cellulose filters (Petrianov). The air sampler was lodged ١ m above the ground, on Tehran and Zahedan stations. The fiber glass and cellulose filters use to count the gross α, gross β and ٧Be, respectively. The instrumentation to count the gross α and gross β activities was automatic beta analyzer (Nuclear Enterprises) and SZn (Ag) counter, respectivly. Long-lived beta activity is total beta activity four days after the end of sampling, when short-lived ٢٣٢Rn progeny had decay into ^{210}Pb. Since the levels of radioactivity encountered in environmental samples are low, long counting times were necessary, on the order of ٣٠٠٠٠ second. The background of detector was determined before and after use by ٣٠٠٠٠ second and was subtracted from sample counts. All the calculations have been made using the appropriate density thickness corrections for efficiency to correct gross beta (based on ٩٠Sr) measurement to specific activity in Bqm-٣ with estimated error at ± ١ sigma.

Weekly aerosol samples have been collected with high-volume air sampler in cellulose filters (Petrianov) and the ٧Be contents of samples were determined with semiconductor gamma spectrometry. Measurement of ٧ Be in each sample was carried out by non–destructive gamma-ray spectrometry by means of its ٤٧٧,٦ keV gamma-ray using a hyper–pure Germanium (HPGe) detector made by CANBERRA (relative efficiency about ٢٠٪ to the efficiency of ٣ x٣ NaI at ٢٥cm distance; resolution ٢keV for ١٣٣٢ keV gamma-ray of ٦٠Co) connected to a ٤٠٩٦ channel pulsed–heigh analyzer. The counting efficiencies of the HPGe detector were measured by using a standard sample containing known amount of radioisotopes such as ١٣٣Ba, ١٣٧ Cs and ٦٠Co. Counting time for each sample was ٦٥k seconds leading to the detection limit about ٠,٨ Bq sample-١.

The ٧Be concentration was calculated using a ٥٣ day half-life, gamma counting efficiencies of ١,٢٪ and branching ratios of ٠,١١. The concentrations were corrected for decay to the mid-collection period.

٣. Results and discussion

The gross α, gross β activities and ٧Be data used in the analysis are daily and weekly values of concentration in surface air. Aerosol sampling for atmospheric radionuclides commenced in January ٢٠٠١ and terminated in July ٢٠٠٤.

The result from individual measurements of the gross α and gross β activities and ٧Be concentration were analyzed to derive the statistical estimate characterizing the dissipation. Table١ provide arithmetic mean (AM) and related statistical information such as geometric mean (GM), standard deviation (S.D.), maximum and minimum values. These data are given (.١٠ $^{-3}$) Bqm $^{-3}$.

Plots of the frequency distribution show highly skewed (flat on the right) histograms for beta and semi symmetric and symmetric one for α and ^7Be, respectively (Figs ١-٣). A range of values of ١٨,٧×١٠ $^{-3}$ Bqm $^{-3}$ and geometric mean of ٦,٨×١٠ $^{-3}$ Bqm $^{-3}$ were found for gross α activity and a range of values of ١٢١,٨×١٠ $^{-3}$ Bqm $^{-3}$ and geometric mean of ٥٢,٠×١٠ $^{-3}$ Bqm $^{-3}$ were found for gross β activity. A range of ٦,١×١٠ $^{-3}$ Bqm $^{-3}$ and the mean value of ٤,٤×١٠ $^{-3}$ Bqm $^{-3}$ at ground level air were measured for ^7Be activity. This letter value consistent with concentration of ^7Be in ground level air that have been reported by others. For ٢٨ sampling sites, Feely at al. (١٩٨٩) listed the mean ^7Be surface air concentration for each month, averaged over all sampling years. Duenas at al. (١٩٩٩) listed the mean gross α, gross β activities and ٧Be concentration in surface air during the years ١٩٩٢-١٩٩٥, a range of values of ٧,٥×١٠ $^{-3}$ Bqm $^{-3}$ and geometric mean of ٦,٨×١٠ $^{-3}$ Bqm $^{-3}$ were found for gross α activity and a range of values of ٥٦,٨×١٠ $^{-3}$ Bqm $^{-3}$ and geometric mean of ٥٢,٠×١٠ $^{-3}$ Bqm $^{-3}$ were found for gross β activity. A range of ٤,٤×١٠ $^{-3}$ Bqm $^{-3}$ and the mean value of ٤,١×١٠ $^{-3}$ Bqm $^{-3}$ at ground level air were measured for ^7Be activity.

١. Temporal Variations of gross α, β and ٧ Be concentration

Table ٢ lists the mean concentration (AM concentration) for gross α , gross β and ^{٧}Be and their standard deviation, for each month, averaged over all sampling years for all samples collected at site. The number of samples in each mean (n) is also given.
On examining this table, it can be said:

a) The highest values of gross beta and gross α activities are registered in November, while those of ^{7}Be concentration came from July.
b) The lowest values of gross beta and gross α activities are registered in February, while those of ^{7}Be concentration came from January.
c) The data of our sampling site shows seasonal variations in the concentration of ^{7}Be . Seasonal variations show minimum averages in fall and winter months and maximum averages during the summer months, probably reflecting the seasonal variations in the transport rate of air from the stratosphere to the troposphere.

The ^{7}Be data for two sites (Tehran and Zahedan) at a different latitude in northern hemisphere show pronounced seasonal variation in surface air concentrations of this radionuclide. Such seasonal variations have long been known to occur (Duenas et al., ١٩٩٩; Cruikshanket et al., ١٩٥٦; Parker, ١٩٦٢; Peirson, ١٩٦٣). Observed seasonal variations in the concentration of ^{7}Be in the surface air have often been attributed to the influence of variations in the exchange rate of air between the stratosphere and the troposphere. Although this influence is clearly real, other factors are also is importance (Feely and et al., ١٩٨٨). In the both sites, variation in the concentration of also result in part from seasonal variations in the rate of vertical mixing within the troposphere with the highest concentration being found during the warmer months (Viezee and Singh,١٩٨٠, Dutkiewicz and Husain ,١٩٨٥). Monthly concentrations of gross beta and gross alpha activities and ^{7}Be in the surface air during the study period ٢٠٠٢ are presented in Figures ٤-٧. The data shows seasonal variations in the concentrations of gross beta activities and ^{7}Be . The concentrations of both are highest during the warm mid-year months. Such seasonal variation has long been known to occur (Duenas et al.,١٩٩٤). Despite of different origins, the concentration of both ^{7}Be and gross beta are partially correlated. Both have maximum concentrations during the warm mid-year months. An important factor in producing peak concentrations of ^{7}Be during the warmer month is the increased rates of vertical transport of air between the stratosphere and the troposphere that occurs during those warm seasons. During the warm months, the solar heating of the surface of the Earth leads to the heating of the air in contact with the surface. Cooler air sinks, displacing the warm, less dense air and forcing it upward. This new air is heated in turn and is forced upward. A convective circulation is produced, carrying surface air upward and bringing downward air from higher levels. This vertical transport carries down to the surface layer the ^{7}Be that has been produced within the upper troposphere, as well as that which has entered the troposphere from the stratosphere.

٣,٢. Analyses of some meteorological factors affecting variation in concentration

The study of meteorological fluctuations has reached, in the last few years, an increasing importance to environmental pollution researchers because its knowledge permits the elaboration of empirical models able to predict periods of potential radioactive pollution. As meteorology plays an important role in the dispersion and transport of pollutants. We have performed a study to identify which meteorological parameters are strongly associated with the fluctuations of daily concentration. During the period of this study, meteorological data (wind direction, temperature, pressure, relative humidity, and precipitation and sunshine hours) were supplied by geophysics institute weather station in Tehran.
Three factors play an effective role in the climate of Tehran: Alborz Mountains, the western humid currents, and the latitude. As a matter of fact, Alborz Mountains make the weather in Tehran moderate. In northern Tehran, the weather is moderate and mountainous, and in plains it is semi-arid. A major part of precipitation takes place in winter. Average annual rainfall stands at ٢٠٠ millimeters. The cold season usually begins in December, but in the mountainous regions, it begins earlier. The cold season lasts ٣ or ٤ months. In mid-March, the weather grows warm. In late April, the weather begins to grow warm at a faster pace, so that in mid-May, it is rather hot.
We studied the variability of the gross β , gross α and ^{7}Be concentration at Tehran station as resulted from local meteorological condition. First, we performed a simple regression of the gross β , gross α and ^{7}Be concentration and some meteorological factors and then we carried out a multiple regression in order to determine the extent to which the variation in concentration might be attributed to the combination of these meteorological parameters. In our analysis we use weekly average of the daily maximum temperature (T_{max}), daily precipitation (RR), daily average relative humidity (H), daily hours of sunshine (SS), daily average pressure (P) and daily average wind speed (V).
In table ٣, the correlation coefficient between concentration and those meteorological factors are summarized. These correlation coefficients are in ٩٩,٩ confidence level.

In order to find the meteorological factors that influence the gross β , gross α and ^{7}Be concentration and evaluate them in order of importance, a multiple regression analysis was carried out with the SPSS ١٢,٠ software. The meteorological variables were obtained chosen by decreasing order of the linear coefficient.
In the validity analysis of each one of the regression equations were taken into account: the relative error of coefficient of each independent variable, the standard error of each independent variable, the standard error of the estimate and the R-squared value. Using these criteria, the following equations were chosen for the gross β and ^{7}Be , respectively, from the results of the forward regression method:

$$A_{\beta}(.١٠^{-3}) = -(٠,٤٧ \pm ٠,٧٤١)+(٠,٠٠٢ \pm ٠,٠٠١)T_{Max} +(٠,٠٠١ \pm ٠٠٠)P \quad (١)$$

$$A_{Be-7}(.١٠^{-3}) =(٢,٢٣١\pm ٠,٨٨٤)+(٠,٠٧٧\pm ٠,٠٤٢)T_{Max} +(٠,٠٠١\pm ٠٠)RR+(٠,٠٩٢\pm ٠,١٠٩)SS \quad (٢)$$

These equation exhibit the number of parameters that most interfere in the fluctuation of the gross β and ^{7}Be . Judging from these equations it can be said that the meteorological variables most influencing the daily gross β activity are: T (temperature) and P (pressure). For the weekly ^{7}Be concentration are: temperature (T), precipitation of that week (RR) and sunshine (SS).
For the gross α activity standard error estimate is too large. Therefore we didn't consider any relation between gross α and meteorology parameters.
A significant feature of the present is that, although it seems to be masked by that to temperature in the regression process, this phenomenon can be qualitatively appreciated in Fig ٨ which represents weekly ^{7}Be concentration and weekly rainfall data during ٢٠٠٢.

٣,٢,١. Model as a function of meteorological parameters

To test validity of equation in estimation of gross α and gross β activities and ^{7}Be concentration in Tehran, we performed a study applying Eqs. (١) and (٢) to weekly data which were not used in the stepwise regression method.
In Figs ٩ and ١٠ we plotted the theoretical line (dotted line), witch showed a concordance between the calculated values and those experimentally observed. The solid lines correspond to lineal least-squares fit to the data. The regression equation obtained are given as

$$\beta_{observed}\ (\text{mBq m}^{-3}) = ١,٨٦٥ \times \beta_{calculated} + ٠,٢١٨٧ \quad (٣)$$

$$^{7}Be_{observed}\ (\text{mBq m}^{-3}) = ٠,٢٩٤١ \times {}^{7}Be_{calculated} + ٦,٨٣٨ \quad (٤)$$

Our study revealed the validity of the relationship for gross β and ^{7}Be and meteorological variables implied in the similarity in the graph slops in Figs ٨ and ٩.
A significant feature of the present results is that, although it seems to be masked by that of temperature in the regression process, this phenomenon can be quantify appreciated in Fig١٠ which represent weekly ^{7}Be concentration and weekly rainfall data during ٢٠٠٢. Rainfall being incidentally very strong or very light may result in rather extreme activity values. Although this affect is clearly noticeable in individual cases (our case) it does not play an important role in the overall evaluation of ^{7}Be (Duenas et al, ١٩٩٩).

References

Cruishank A.J., Cowper G., Grummite W.E.,١٩٥٦, Production of in the ^{7}Be atmosphere, *Canadian Journal of Chemistry*, ٣٤:٢١٥-٢١٩.

Duenas C., Fernandez M.C., Liger E. , Carretero J., ١٩٩٩, Gross Alpha, gross beta activities and ^{7}Be concentration in surface air: analysis their variation and prediction model, *Atmospheric environment,* ٣٣: ٣٧٠٥-٣٧١٥.

Duenas C., Perez M., Fernandez M.C., Carretero J., ١٩٩٤, Disequilibrium of Radon an its short-lived daughters near the ground with atmospheric stability. *Journal of Geophysical Research* ٩٩, (D٦):١٢٨٦٥-١٢٨٧٢.

Dutkiewicz V.A., Husain L., ١٩٧٩, Determination of stratospheric ozone at ground level using ^{7}Be/ozone ratios, Geophysical Research Letter, ٦(٣):١٧١-١٧٤.

Dutkiewicz V.A., Husain L., ۱۹۸۵, Stratospheric and tropospheric component of ^{7}Be in surface air, Geophysical Research, ۹۰:۵۷۸۳-۵۷۸۸.

Feely H. W., Larson R. J. and Sanderson (۱۹۸۸), Factors that cause seasonal variation in ^{7}Be concentrations in surface air, *J. Environ. Radioactivity.* ۹: $223-249$.

Johnson W. B., and Viezee W. (۱۹۸۱), Stratospheric ozone in the lower troposphere – I. Presentation and interpretation of aircraft measurements, *Atmos.Environ.* ۱۵: $1309-1323$.

Papastefanou C., Ioannidou A., ۱۹۹٤, Aerodynamic size of ^{7}Be in ambient aerosols. *Journal of Environmental Radioactivity* , ۲٦:۲۷۳-۲۸۱.

Parker R. p., ۱۹٦۲, ^{7}Be and fission products in surface air, *Nature,* ۱۹۳, ۹٦۷-۹٦۸.

Peirson D.H., ۱۹٦۳, ^{7}Be in air and rain. *Journal of Geophysical Research*, ٦۸(۱۳):۳۸۳۱- ۳۸۳۲.

Reitz G. (۱۹۹۳), Radiation environment in the stratosphere, *Radiat Prot Dosim.*, ٤۸: ٥-۲۰.

Viezee W., Singh H.B., ۱۹۸۰, The distribution of ^{7}Be in troposphere :implication on stratospheric/tropospheric air exchange. *Geophysical Research Letters,* ۷(۱۰):۸۰٥-۸۰۸.

Table۱. Statistical parameters of the different measurements

	Gross β ($\times$۱۰$^{-3}$ Bqm^{-3})	Gross α ($\times$۱۰$^{-3}$ Bqm^{-3})	^{7}Be($\times$۱۰$^{-3}$ Bqm^{-3})
Data	٦۹۰	٦۹۰	۲۱۸
AM	۲۲۲,٦۰	۸٦,۱٦	۷,۲٤
GM	۲۱۷,۱۲	۸۳,٤۷	٦,۸٦
S.D.	٥۸,٤۱	۲۱,۷۷	۲,٥۲
maximum	٥۲۳,۰۸	۱۹٥,۱٦	۲٥,٤۹
minimum	۸,۰۲	۱,٤	۱,۳٦

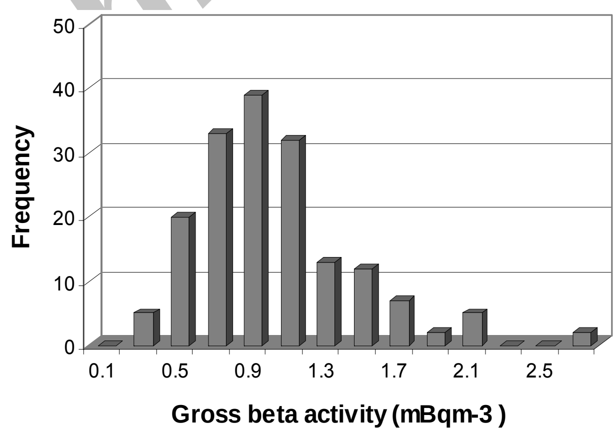

Figure۱. Frequency histogram of gross β activity in Tehran station.

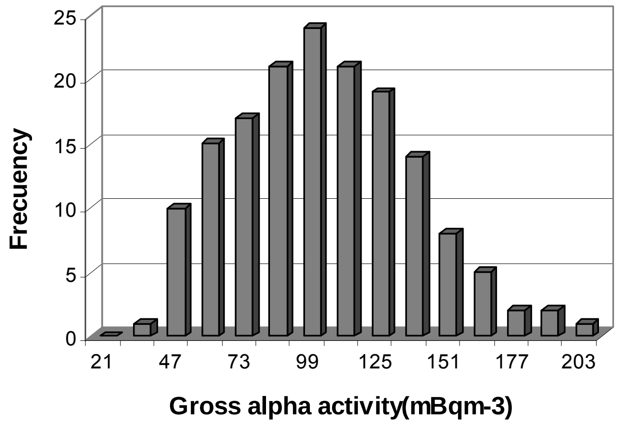

Figure۲. Frequency histogram of gross α activity in Tehran station.

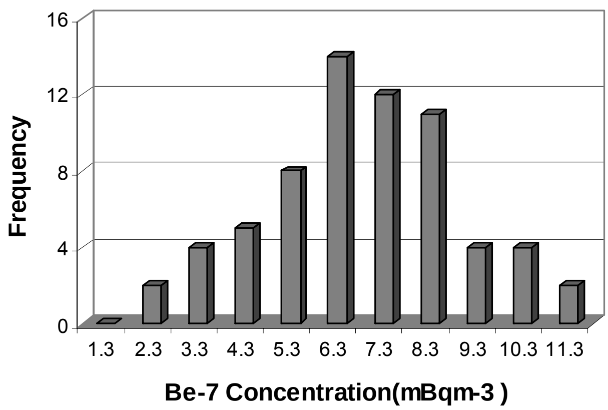

Figure۳. Frequency histogram of ۷Be concentration in Tehran station.

Table۲. Mean monthly measured concentration in surface air averaged over all sampling years.

Month	Gross α (×١٠$^{-3}$ Bqm^{-3})			Gross β (×١٠$^{-3}$ Bqm^{-3})			^{7}Be(×١٠$^{-3}$ Bqm^{-3})		
	AM	***S.D***	***n***	***AM***	***S.D***	***n***	***AM***	***S.D***	***n***
January	٧٤,٥٨	٢٧,٦١	٦٤	١٩٦,٤٧	٥٦,٩١	٦٤	٥,٦٢	٢,١٠	١٩
February	٧١,٠٤	٢٥,١٢	٥٤	١٧٤,٩١	٦٦,٠٤	٥٤	٦,٢٥	٢,٢٠	١٧
March	٩٤,٥٧	٢٩,٨١	٣٠	٢٤٣,٣٨	٨٩,٣٨	٣٠	٥,٨٨	٣,٦٩	٩
April	٧٣,٢٥	١٥,٢١	٤٧	١٩٦,٣٧	٤٤,٤٤	٤٧	٦,٨٧	٢,٤٦	١٨
May	٧٨,٣٠	٢٨,٣٧	٥٩	٢٠٩,٢٤	٦٦,٠٩	٥٩	٨,١٥	١,٦٧	١٦
June	٧٩,٢١	٣٧,١٦	٦١	٢٠٦,٣٢	٨٢,٢٥	٦١	٩,٠٣	١,٨٥	٢١
July	٧٧,٩٠	٢٠,٥٣	٦٩	١٩٤,٢٢	٦٨,٨٦	٦٩	٩,٦١	٢,٩٢	٢٢
August	٧٩,٩١	١٧,٢٩	٦٦	٢٠٩,١٦	٥٤,٥١	٦٦	٨,٠٣	٠,٥٨	٢٠
September	٩٣,٧٦	١٢,٢٦	٦١	٢٤٧,٢٢	٣٧,٤٧	٦١	٧,٤١	٢,٨٦	١٩
October	١١٠,٠١	٨,٦٩	٥٨	٢٧٧,٢٨	٢٦,١٨	٥٨	٧,٤٩	٢,٧٩	٢١
November	١٢١,٢٠	٢٠,٨١	٥٤	٣٠٥,٤٦	٦٣,٨٠	٥٤	٥,٦٥	٢,٧١	١٧
December	٨٠,١٧	١٨,٣٩	٦٧	٢١١,١٩	٤٥,٠٣	٦٧	٦,٨٧	٤,٤٤	١٩

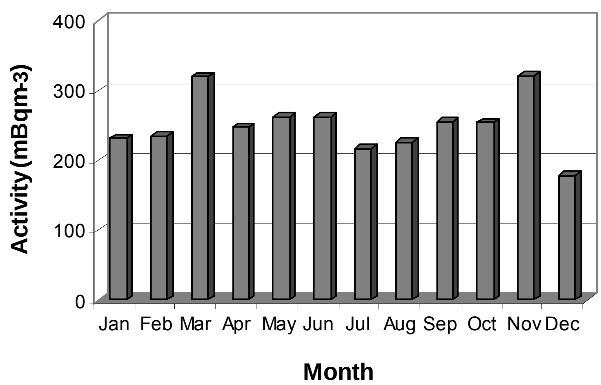

Figure ٤. Monthly concentrations of long-lived beta activity in the surface air in Tehran station during ٢٠٠٢.

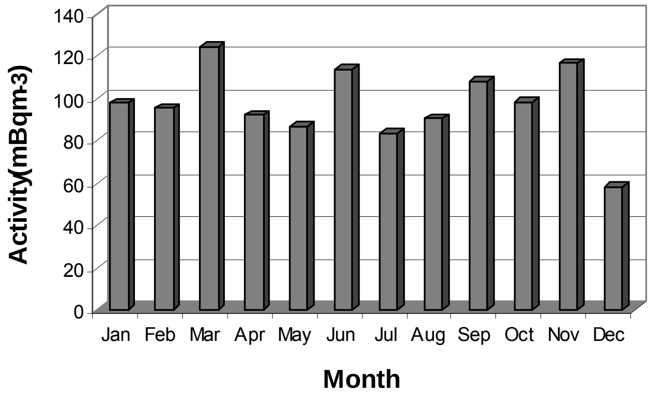

Figure ٥. Monthly concentrations of gross α activity in the surface air in Tehran station during ٢٠٠٢.

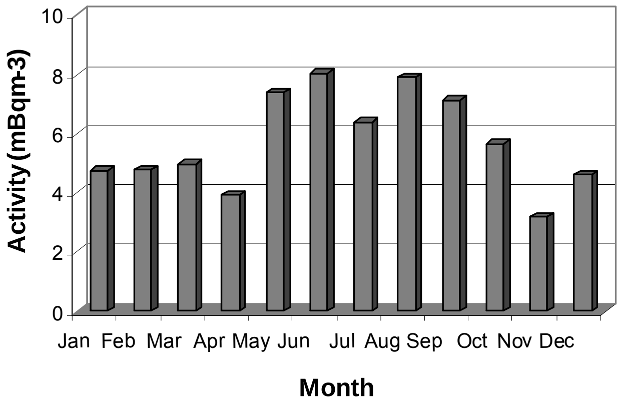

Figure٦. Monthly variations in the concentrations of ^{7}Be in the surface air in Tehran station during ٢٠٠٢.

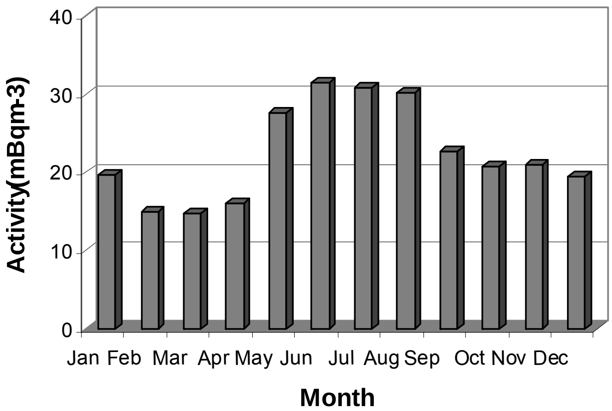

Figure٧. Monthly variations in the concentrations of ^{7}Be in the surface air in Zahedan station during ٢٠٠٢.

Table٣. Linear correlation coefficient between surface air concentrations and some meteorological factors

Activity	T_{max}	H	P	RR	WF	SS
Gross α	٠,٠٨٨	-٠,٢٢٩	٠,١٢٦	-٠,١٦٩	٠,٠١٧	٠,٢٠٣
Gross β	٠,٠٩٤	-٠,٢٣٤	٠,١٣٢	-٠,١٨٩	٠,٠١٢	٠,٢١٨
^{7}Be	٠,٥٥٩	-٠,٤٥٥	٠,٣٣٨	-٠,٢٣	٠,٣٣٨	٠,٤٤

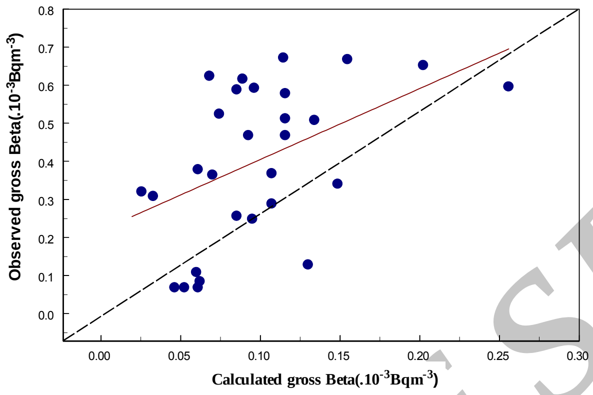

Figure 8.Observed gross beta activity versus the calculated.

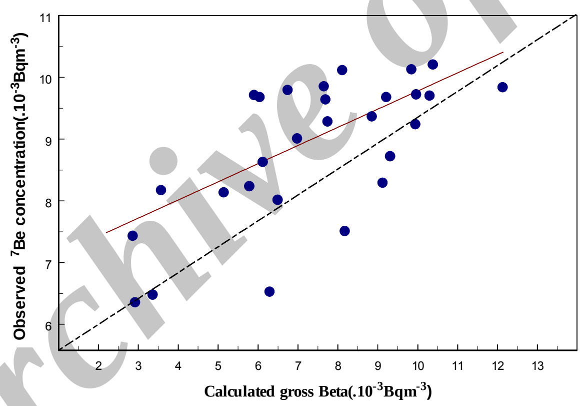

Figure 9.Observed ^{7}Be concentration versus the calculated

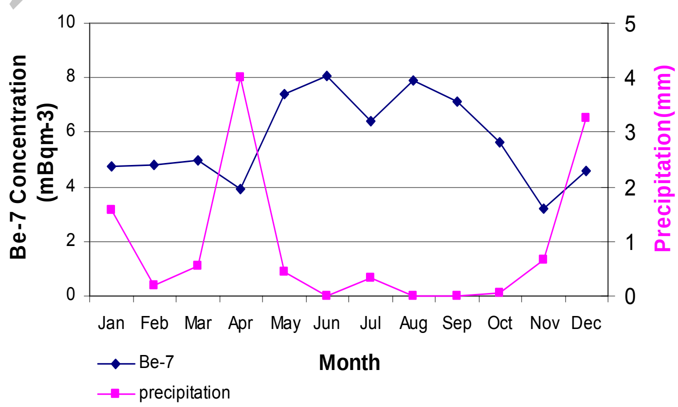

Figure١٠. ^{7}Be concentration and precipitation during ٢٠٠٢.