Analysis of gross alpha, gross beta activities and beryllium-7 concentrations in surface air: their variation and statistical prediction model

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INTRODUCTION

Radioactivity in the atmosphere originates from natural radioactive decay, cosmogenic production, nuclear weapons testing and nuclear accidents.

Results of the measurements have shown that cosmogenic ⁷Be is the only gamma emitting nuclide found in the atmosphere of Iran (¹).

³Be (t¹/₂ = 53d) is one of the radioactive products of the bombardment of the atmosphere by cosmic rays. About 75% of ⁷Be is produced in the stratosphere, and 25% in upper troposphere (²). Once ⁷Be is formed in the troposphere, it rapidly associates primarily with sub-micron-sized aerosol particles. ⁷Be in these aerosols may subsequently enter the marine and terrestrial environment via wet or dry deposition (³).

In this paper we report four years of continuous measurements of gross α and gross β activities, and concentrations of ⁷Be in surface air. The present research was undertaken with the following principal goals: a) To perceive the variations of gross α, gross β and ⁷Be concentration in Tehran as compared to Zahedan; b) To identify the main meteorological parameters which are responsible for the variation of those concentrations and c) To model the data obtained from our sampling sites as a function of meteorological parameters such as temperature, precipitation, wind speed, relative humidity, etc. With the regression model, the ground level concentration associated with the changes in meteorological conditions could be estimated.

Experimental procedures

Airborne dust samples were daily collected on fiber glass filters, 47-mm diameter (collection efficiency 99.99% for 0.8 pore size) with an air sampler at a flow rate of 39 m³/day⁻¹. Aerosol samples have been weekly collected with high-volume air sampler on

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cellulose filters (Petrianov). Both types of air samplers were 1 m above the ground, in Tehran and Zahedan stations.

The fiber glass and cellulose filters were used to count the gross $\alpha$, gross $\beta$ and $^7\text{Be}$, respectively. The measurements of the gross $\beta$ and gross $\alpha$ activities were performed by an automatic beta analyzer and a ZnS (Ag) counter. Long-lived beta activity was defined as total beta activity four days after the end of sampling, when short-lived $^{222}\text{Rn}$ progeny have been decayed into $^{210}\text{Pb}$. The counting time was 30000 seconds and the background was also collected for the same period of time and subtracted from the samples count.

Aerosol samples were collected on weekly basis with the high-volume air samplers and cellulose filters were used to determine the $^7\text{Be}$ contents of samples, using a semiconductor gamma spectrometer. Determination of $^7\text{Be}$ concentration was made by means of its 477.6 keV gamma-ray using a CANBERRA Hyper-Pure Germanium (HPGe) detector (relative efficiency about 20% to the efficiency of 3×3 NaI at 25cm distance, resolution 2 keV for 1332 keV gamma-ray of $^{60}\text{Co}$) connected to a 4096 channel pulsed-height analyzer. The counting efficiencies of the HPGe detector was determined by a standard sample containing a known amount of radioisotopes such as $^{133}\text{Ba}$, $^{137}\text{Cs}$ and $^{60}\text{Co}$. Counting time for each sample was 60000 seconds, leading to the detection limit of about 0.8 Bq per sample.

The $^7\text{Be}$ concentration was calculated using a 53-day half-life, gamma counting efficiencies of 1.2% and branching ratios of 0.11. The concentrations were corrected for decay to the mid-collection period.

RESULTS AND DISCUSSION

The gross $\alpha$, gross $\beta$ activities and $^7\text{Be}$ data used in the analysis were daily and weekly values of concentration in surface air. Aerosol sampling for atmospheric radionuclides commenced in January 2001 and terminated in July 2004.

Temporal Variations of gross $\alpha$, $\beta$ and $^7\text{Be}$ concentration

Table 1 lists the arithmetic mean concentration (AM) for gross $\alpha$, gross $\beta$ and $^7\text{Be}$ and their standard deviation for each month, averaged over all sampling years for all samples collected the sites. The number of samples for each mean (n) is also given in the table. On examining this table, it can be said:

1) The highest values of gross $\beta$ and gross $\alpha$ activities were registered in November, while those of $^7\text{Be}$ concentration were obtained in July.

2) The lowest values of gross $\beta$ and gross $\alpha$ activities were registered in February, while those of $^7\text{Be}$ concentration were obtained in January.

3) The data of our sampling site have shown seasonal variations in the concentrations of $^7\text{Be}$. Seasonal variations presented 2 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 $^7\text{Be}$ data for two sites (Tehran and Zahedan), which are at a different latitudes, show pronounced seasonal variation in surface air concentrations of this radionuclide (1). Such seasonal variations have long been known to occur (4, 5). Observed seasonal variations in the concentration of $^7\text{Be}$ 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 important (6). In the both sites, variation in the concentration was also resulted in part from seasonal variations in the rate of vertical mixing within the troposphere with the highest concentration being found during the warmer months (7).

Both sites have maximum concentrations during the warm, mid-year months. An important factor in the production of peak concentrations of $^7\text{Be}$ during the warmer month has been increased rates of vertical transport of air between the stratosphere and
the troposphere which occurred during those warm seasons. During the warm months, the solar heating of the surface of the earth led to the heating of the air in contact with the surface. Cooler air sinks, displaced the warm, less dense air and forced it upward. The new air has been heated in turn and forced upward. A convective circulation has been produced, carrying surface air upward and bringing downward the air from higher levels. This vertical transport carried down the $^7$Be to the surface layer which has been produced within the upper troposphere, as well as that which has entered the troposphere from the stratosphere.

The data showed seasonal variations in the concentrations of gross beta activities. The concentrations were highest during the warm mid-year months. Duenas et al. measured the mean $^7$Be concentration in surface air about $4.1 \times 10^3$ Bq m$^{-3}$ during the years 1992-1995. The concentration of $^7$Be in their study was higher than the above mentioned value, since the stability of the tropospheric air was highest toward the cold polar region and decreased the rate of vertical mixing within troposphere.

### Analyses of some meteorological factors affecting variation in concentration

Meteorology plays an important role in dispersion and transport of pollutants. A study has been performed to identify which meteorological parameters have been strongly associated with the fluctuations of daily concentration. During the period while this study was being conducted, 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: the Alborz Mountains, the western humid currents, and the latitude. As a matter of fact, the Alborz Mountains moderate the weather in Tehran. In northern Tehran, the weather is moderate and mountainous, and in the plains it is semi-arid. A major part of precipitation takes place in winter. Average annual rainfall is about 200 mm. The cold season usually begins in December, but in the mountainous regions, it begins earlier. The cold season lasts 3 to 4 months. In mid-March, the weather grows warm.

### Table 1. Mean monthly measured concentration in surface air averaged over all sampling years.

<table>
<thead>
<tr>
<th>Month</th>
<th>Gross $\alpha$ (mBq m$^{-3}$)</th>
<th>Gross $\beta$ (mBq m$^{-3}$)</th>
<th>$^7$Be (mBq m$^{-3}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>AM</td>
<td>S.D</td>
<td>n</td>
</tr>
<tr>
<td>January</td>
<td>74.58</td>
<td>27.61</td>
<td>64</td>
</tr>
<tr>
<td>February</td>
<td>71.04</td>
<td>25.12</td>
<td>54</td>
</tr>
<tr>
<td>March</td>
<td>94.57</td>
<td>29.81</td>
<td>30</td>
</tr>
<tr>
<td>April</td>
<td>73.25</td>
<td>15.21</td>
<td>47</td>
</tr>
<tr>
<td>May</td>
<td>78.30</td>
<td>28.37</td>
<td>59</td>
</tr>
<tr>
<td>June</td>
<td>79.21</td>
<td>37.16</td>
<td>61</td>
</tr>
<tr>
<td>July</td>
<td>77.90</td>
<td>20.53</td>
<td>69</td>
</tr>
<tr>
<td>August</td>
<td>79.91</td>
<td>17.29</td>
<td>66</td>
</tr>
<tr>
<td>September</td>
<td>93.76</td>
<td>12.26</td>
<td>61</td>
</tr>
<tr>
<td>October</td>
<td>110.01</td>
<td>8.69</td>
<td>58</td>
</tr>
<tr>
<td>November</td>
<td>121.20</td>
<td>20.81</td>
<td>54</td>
</tr>
<tr>
<td>December</td>
<td>80.17</td>
<td>18.39</td>
<td>67</td>
</tr>
</tbody>
</table>
In this study, First, a simple regression of the gross $\beta$, gross $\alpha$ and $^7\text{Be}$ concentrations against some meteorological factors was performed, and then the procedure has been carried out a stepwise 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 the analysis, weekly average of the daily maximum temperature ($T_{\text{max}}$), daily precipitation (PR), daily average relative humidity (H), daily hours of sunshine (SS), daily average pressure (P) and daily average wind speed (V) were used.

In table 2, the correlation coefficients between concentrations and those meteorological factors are summarized. These correlation coefficients are in 99.9 confidence level.

To evaluate the regression equations, 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 were taken into account. Using these criteria, the following equations were chosen for the gross $\beta$ and $^7\text{Be}$, respectively, from the results of the forward regression method:

$$A_{\beta}(\times10^{-3}) = -(0.47 \pm 0.741) + (0.002 \pm 0.001) \times T_{\text{max}} + (0.001 \pm 000) \times P$$  \hspace{2cm} (1)

$$A_{\beta-7}(\times10^{-3}) = (2.231 \pm 0.884) + (0.077 \pm 0.042) \times T_{\text{max}} + (0.001 \pm 00) \times PR$$
$$+ (0.092 \pm 0.109) \times SS$$ \hspace{2cm} (2)

These equations show the number of parameters that apparently modify the fluctuations of the gross $\beta$ and $^7\text{Be}$. Analyzing these equations, it can be said that the meteorological variables which influence the daily gross $\beta$ activity the most are: $T$ (temperature) and $P$ (pressure). For the weekly $^7\text{Be}$ concentration, temperature ($T$), precipitation of that week (PR) and sunshine (SS) are the most important ones. For the gross $\alpha$ activity, standard error of estimate for the equations has been too large. Therefore, no relation between gross $\alpha$ and meteorology parameters has been considered.

Model as a function of meteorological parameters

To test validity of the equations for estimation of gross $\alpha$ and gross $\beta$ activities and $^7\text{Be}$ concentration in Tehran, Eqs. (1) and (2) to weekly data which were not used in the stepwise regression method was applied.

In figures 1 and 2, the theoretical line (dotted line) between the calculated values and those experimentally observed were plotted. The solid lines correspond to linear least-squares fit to the data. The regression equations obtained are given as:

$$\beta_{\text{observed}} (\text{mBq m}^{-3}) = 1.865 \times \beta_{\text{calculated}} + 0.2187$$  \hspace{2cm} (3)

$$^7\text{Be}_{\text{observed}} (\text{mBq m}^{-3}) = 0.2941 \times ^7\text{Be}_{\text{calculated}} + 6.838$$ \hspace{2cm} (4)

Our study revealed the validity of the relationship between gross $\beta$ and $^7\text{Be}$ and meteorological variables implied, and the similarity between the graphs slops in figures 1 and 2.

A significant feature of the present results is that it seems to be masked by the temperature in the regression process. This phenomenon can be quantified in figure 3 which represents weekly $^7\text{Be}$ concentration and weekly rainfall data during 2002. The incidental very strong or very light may result in rather extreme activity values. Although this effect is clearly noticeable in special cases (our case), it does not play an important role in the overall evaluation of $^7\text{Be}$ (5).
Gross α, gross β activities and 7Be concentrations in air

Figure 1. Observed gross beta activity versus the calculated values.

Figure 2. Observed 7Be concentration versus the calculated values.

Figure 3. 7Be concentration and precipitation during the year 2002.

REFERENCES