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RADIOACTIVITY VARIATION IN THE WATERS OF SOME SPRINGS IN THE STATE OF SAO PAULO, BRAZIL

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SUMMARY

This paper contains a study of the variations in radioactivity in the waters of eight springs in the State of Sao Paulo, Brazil. The method consisted of, first, measurement of radioactivity levels in the waters of these springs over a period of twelve months, and then the processing of these field data using computer programs based on statistical methods.

The following conclusions were drawn from the results obtained: the variation in the radioactivity level is related to the dry and wet seasons and it was found to be dependent on the extent of dilution caused by rain. Dilution in this way was observed immediately in waters circulating in shallow aquifers (sandstones) during the wet season. In deeper aquifers (volcanic rocks), on the other hand, dilution was found to occur later on in the dry season.

Although this paper contains a study of natural radioactivity, the field and statistical procedures implemented herein can also be applied in exactly the same way to monitor radioactivity levels in surface and ground waters caused (accidentally or otherwise) by nuclear installations. In view of the fact that several less developed countries now have nuclear installations, clearly the importance of monitoring of this type in those countries cannot be over-emphasized.

1. INTRODUCTION

The purpose of this work was to study the temporary radioactivity of some spring waters in the state of Sao Paulo, considered to be the most radioactive of Brazilian waters, in accordance with the Brazilian

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Code of Mineral Waters. It was also considered important to observe possible variations in the radioactivity levels, and to correlate these levels with possible parameters. For this purpose water samples were collected, and their radioactivity analysed, for a period of twelve months. Spring water samples of 200 ml were collected monthly during the year of May 1978 to April 1979. The total atmospheric precipitation during the same period was also collected.

The determination of temporary radioactivity was carried out at the "Instituto de Pesquisas Energeticas e Nucleares" (IPEN), Sao Paulo, Brazil, where the concentration of ^{222}Rn was analysed using liquid scintillation detectors. Results of radioactivity determinations, together with the chemical and physico-chemical results, were processed by the computer programs CORELE and ACP at the Department of Laboratories, Service of Geochemistry of the *Bureau de Recherches Geologiques et Minieres* (B.R.G.M.), Orleans, France.

Although this paper contains a study of natural radioactivity, the field and statistical procedures implemented in this work can also be applied in exactly the same way to monitor radioactivity levels in surface and ground waters caused (accidentally or otherwise) by nuclear installations. Considering that nowadays a number of less developed countries (e.g. India, Pakistan, etc.) possess nuclear installations, it is clear that radioactivity monitoring is becoming increasingly important in these countries for ensuring that no serious injury is inflicted upon the environment by these installations. Furthermore, since the overall expertise in running nuclear installations *may* be less in the developing countries than in the developed countries, monitoring of the type in the less developed countries clearly takes on an additional dimension of importance.

2. GEOLOGY

The region of *Aguas da Prata* occupies the western part of the alkaline massif of *Pocos de Caldas*, which extends over an area of 800 sq. kms, and is considered one of the largest nephelinic volcanic complexes. This massif is emplaced between granites and gneisses, and is composed principally of nephelinic rocks such as tinguaites, foyaites and phonolites. In the interior of the massif, rocks older than the nephelinic intrusion are found, such as sandstones, siltstones, shales and volcanic rocks (e.g. tuffs, breccia, agglomerates and ankaratritic lavas). The town of Aguas da Prata is limited by a dyke of tinguaite on the west, and by clastic rocks on the east[1].

The principal springs of this region belong to this set of igneous and clastic rocks (Figure 1). The *Platina* spring issues from phonolitic rocks; the *Paiol* from volcanic tuffs and breccia; the *Prata-Antiga* and *Prata-Nova* from diabases; *Villela, do Boi* and *Prata-Radioativa* from sandstones, and *Vitoria* from phonolites and diabases covered by sandy alluvium[2].

3. RESULTS AND DISCUSSION

The concentration values of radioactivity are shown in Figure 2. In this figure a separation of spring waters into three groups of different radioactivity can be observed. This separation is due mainly to the different lithologies from which these spring waters issue, and secondly, to the depth of some of the springs. For example, the *Paiol* spring is a well 133 m deep (volcanic tuffs and breccia) and has the lowest radioactivity. The *Platina, Prata-Antiga* and *Prata-Nova* springs, issuing from phonolites and diabases, and the *Prata-Radioativa* from sandstones covered by siltstones, show a higher radioactivity. Waters with the highest radioactivity are those issuing from sandstones (*do Boi* and *Villela* springs) and phonolites, with diabases covered by sandy alluvium (*Vitoria* spring).

The origin of radioactivity in the waters of the springs considered is related to the radioactive minerals found in this region: zirconium, badelleyte, thorium minerals, monzaite, apatite and eudialite. The fractures of the sandstones are filled with a yellow material,[3] generally containing 0.1–0.2 percent of U_3O_8.

For the study of correlation and variation of radioactivity, all results were processed by the CORELE and ACP computer programs. The results from spring waters issuing from sandstones, and those issuing from volcanic rocks and diabases, were treated separately.

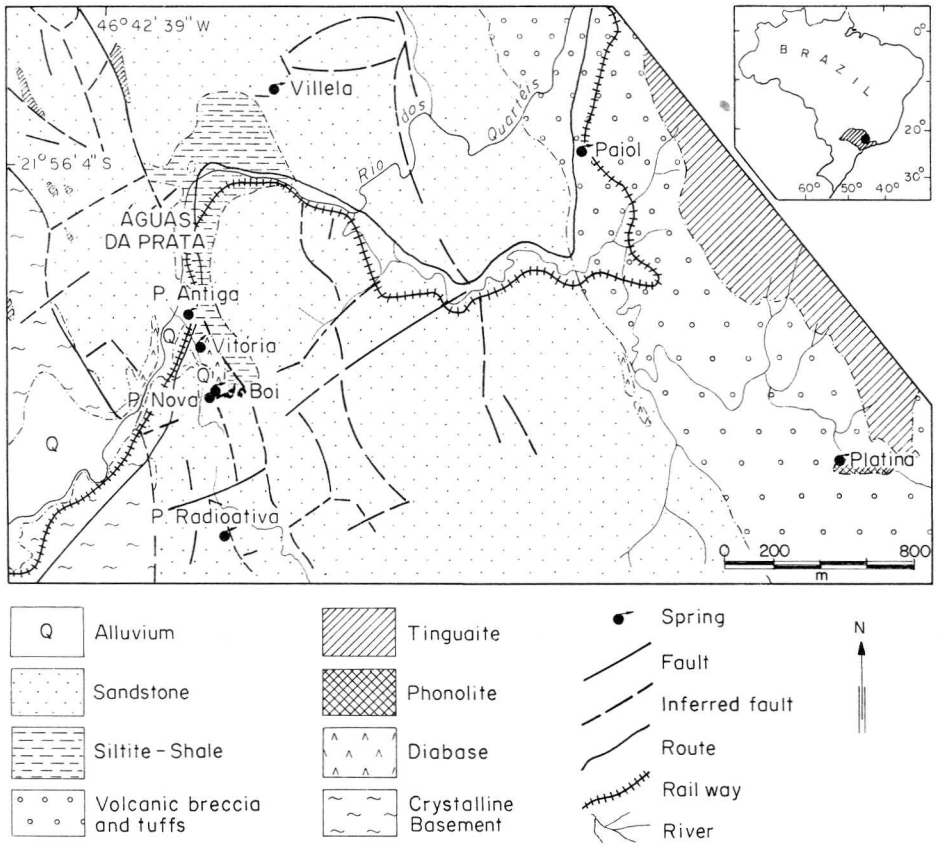

Figure 1. Geologic map of Aguas da Prata showing springs

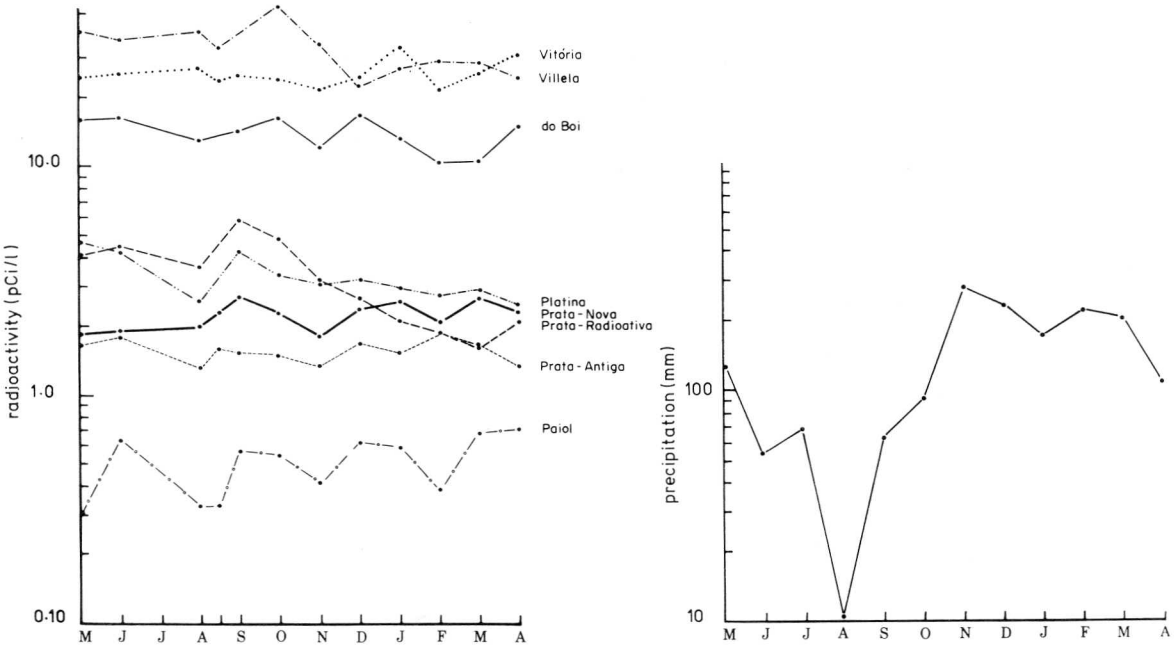

Figure 2. Variation of radioactivity during 12 months (May 1978–April 1979) and atmospheric precipitation

Table 1. Correlations found from the CORELE program

Correlation	Waters from sandstones	Waters from volcanic rocks and diabases
Above 0·8	Conductivity—HCO_3, Mg	I–Zn Conductivity—HCO_3, Na, SO_4 Ca–Mg
Between 0·8 and 0·5	pH–Zn Conductivity—Ca, PO_4 Radioactivity—Diss.O_2, K Ca–HCO_3, Mg Cl–NO_3, PO_4	Temperature—F pH–HCO_3, F I–SO_4, Conductivity, Cr, HCO_3 SiO_2–NO_2
Inverse Correlation around −0·5	Radioactivity—HCO_3, Mg	Temperature—Ca, Mg Radioactivity—F, HCO_3 Zn, Cr–Br

The CORELE program provided the correlation between the physical, physico-chemical and chemical parameters. Table 1 shows the correlations found using this program.

The values obtained from the CORELE program were then processed, using the ACP program which calculates the coordinates of the variables with respect to the factorial axes, giving a visual representation of the results upon a bidimensional plan. F1 is the preferential axis of the dispersion of the measurements, while F2 shows seasonal variations.

In Figures 3 and 5 the bidimensional diagrams represent the correlations between all variables. The circle in each diagram represents the maximum correlation of unity, and the correlation linearly decreases to zero at the centre. The ACP program plots F1 and F2 on the horizontal and vertical axes, respectively. However, in Figures 3 and 4, the axes F1 and F2 are rotated clockwise to emphasize the radioactivity correlation.

Results in Figures 3 and 5 are plotted on the factorial axes F1 and F2. Figure 3 shows that in these waters radioactivity is directly related to the dissolved oxygen values, and inversely related to the concentrations of Mg and HCO_3; and, to a lesser degree to those of Ca and SiO_2, and conductivity. The most radioactive waters are further related to NO_2, Cl, K and Na. The dissolved oxygen values are higher (4–19 mg/l) in waters of shallow aquifers (sandstones) than in the deeper ones (volcanic rocks,

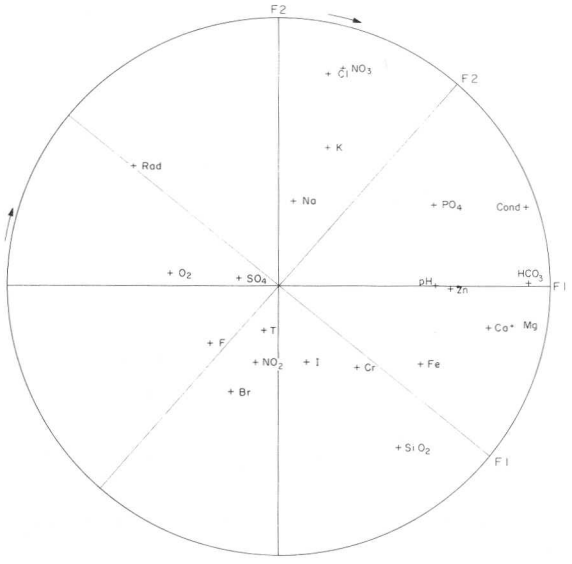

Figure 3. Bidimensional representation of correlation (*Villela, Prata-Radioativa* and *do Boi* springs)

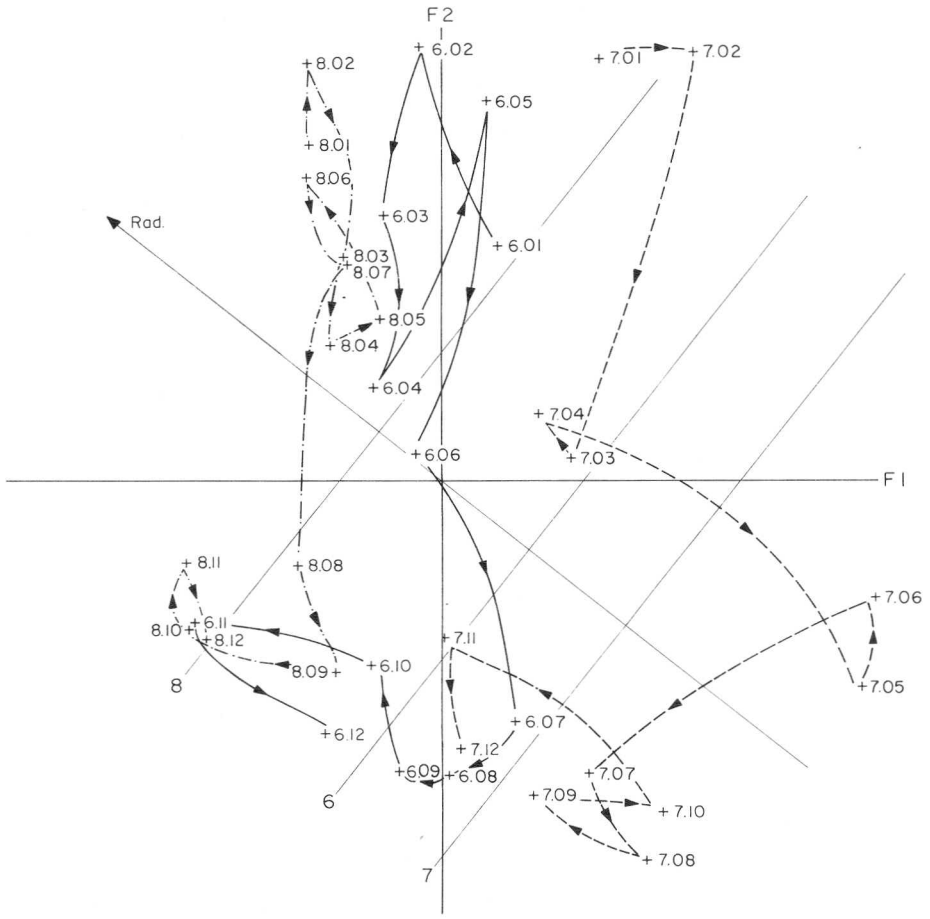

Figure 4. Bidimensional representation of monthly values of radioactivity (*Villela*–8, *Prata-Radioativa*–7, *do Boi*–6). First number of spring, second number of analysis (for example: 8·20 means *Villela* spring, analysis of June)

2–8 mg/l). As mentioned before, the sandstones contain uranium-bearing material; therefore, the direct correlation shown between dissolved oxygen and radioactivity is, in reality, a reflection of the lithology as much as the presence of NO_3, Ca and Na in the soil.

The arrangement of results in Figure 4 indicates that the *Villela* (No. 8) and the *do Boi* (No. 6) springs are the most radioactive, and the *Prata-Radioativa* spring (No. 7) the least radioactive.

Figure 5 shows that the radioactivity in these waters is directly related to the concentration of K, and inversely related to the concentrations of F, HCO_3, Na, SO_4, and also to temperature and conductivity. Waters rich in F are less radioactive, and greater radioactivity is directly related to Zn and I concentrations.

It may be seen from Figure 6 that the *Vitoria* spring (No. 5) is the most radioactive, and *Paiol* spring (No. 4) the least radioactive; the latter is also the richest in F.

F1 in Figure 4 represents the axis of radioactivity (displaced in this case), and axis F2 depicts seasonal variations. Clearly, in this figure, radioactivity values are grouped together, above and below the F2 axis, thus indicating that there is a separation of values by seasons. The same is observed in Figure 6, although in this case the axis F2 shows the values of different radioactivities, and axis F1 the seasonal variations.

Discharge measurements were also made for three springs (*do Boi, Platina* and *Vitoria*) issuing from different lithologies, every month for a year. These measurements were correlated with rainfall. A direct correlation was found to exist between rain and discharge of waters from sandstones (*do Boi*); a

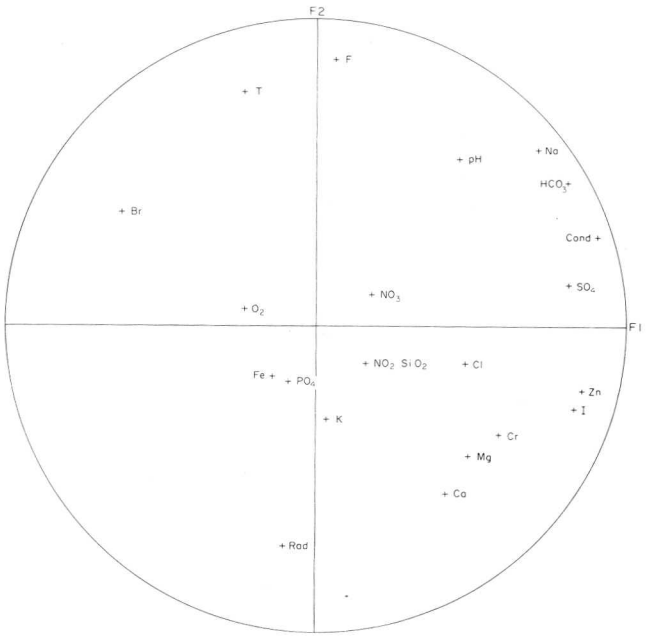

Figure 5. Bidimensional representation of correlation (*Platina, Prata-Antiga, Prata-Nova, Paiol* and *Vitoria* springs)

direct, but delayed (by a month) relationship was found for diabase and phonolites covered by sandstones (*Vitoria*), and no correlation at all (i.e. uniform discharge during the whole year) for the deep phonolite aquifers (*Platina*)[3].

Therefore, radioactivity variation during the year is related to the season, and particularly to the rain. In more radioactive waters (those issuing mainly from sandstones), the radioactivity values were higher during autumn-winter-spring, and lower during the summer when the rains are most abundant, leading to the dilution of water and thus reducing its radioactivity level (Figure 2).

Waters issuing from volcanic rocks and diabases, and those with lower radioactivity values, on the other hand, were found to have higher values in spring–summer–autumn, and lower values in autumn–winter. The reason for this could be that the dilution of spring water during the wet season (summer) is immediate in the case of waters issuing from shallow aquifers (sandstones); therefore, the radioactivity of these waters is lower in the summer. In waters issuing from deeper aquifers (volcanic rocks and diabases), on the other hand, rain dilution is delayed, and its effect is noticed only in autumn-winter (Figure 2).

A correlation was also found to exist between radioactivity values and the depth of springs. This can be explained as follows: the concentration values of Radon gas are related to the transit time of water as it escapes upwards from the issuing source. Thus, depending on the depth of the spring, and therefore the transit time, radioactivity will be reduced by radioactive decay ($T_{1/2} = {}^{222}Rn = 3.82$ days).

4. CLASSIFICATION OF WATERS

The waters are classified with respect to their radioactivity in accordance with the Brazilian Code of Mineral waters, as follows: waters of *Prata-Antiga* spring with a mean of 1,607 pCi/l, and *Paiol* spring with 530 pCi/l, are considered not to be radioactive; the spring waters of *Prata-Radioativa* with a mean of 3,500 pCi/l, *Platina* with 3,400 pC/l, and *Prata-Nova* with 2,304 pCi/l to be feebly radioactive; waters of the *do Boi* spring with a mean of 14,342 pCi/l as radioactive, and waters of the *Villela* spring with a mean of 34,337 pCi/l and the *Vitoria* spring with a mean of 26,197 pCi/l, to be strongly radioactive.

Figure 6. Bidimensional representation of monthly values of radioactivity (*Platina*–1, *Prata-Antiga*–2, *Prata-Nova*–3, *Paiol*–4 and *Vitoria*–5). First number—spring, second number—month of analysis. (For example: 5·70 means *Vitoria* spring, analysis of October)

5. CONCLUSIONS

The results of this study show that, despite their temporary radioactivity, all spring waters considered may still be regarded as mineral waters, with the exception of those of the *Prata-Antiga* and the *Paiol* springs. Waters from the *Prata-Radioativa, Platina* and the *Prata-Nova* springs are feebly radioactive, those from *do Boi* radioactive, and those from *Villela* and *Vitoria* strongly radioactive.

The waters are separated into three groups, based on their degrees of radioactivity. The most radioactive waters are those of the *Villela, Vitoria* and the *do Boi* springs, in which the radioactivity is related to the radioactive materials disseminated in the fractures of the sandstone (*Villela* and *do Boi*), and phonolites with diabases covered by sandy alluvium (*Vitoria*).

In another group of feebly radioactive waters (*Platina, Prata Nova* and *Prata-Radioativa* springs), radioactivity is related to zirconium and uranium-bearing minerals, found in nephelinic syenitic rocks (*Platina* and *Prata-Nova*), and to sandstones with siltstones (*Prata-Radioativa*). The waters of the *Paiol* and the *Prata-Antiga* springs belong to a third group, considered as not radioactive.

A decrease in radioactivity was observed with increasing depth of springs. Also, the radioactivity

variations showed a separation into two distinct groups which corresponded to the two seasons, wet and dry. Thus, the radioactivity variation is due to the dilution of spring waters by rain, leading to decreased radioactivity. This dilution was observed immediately during the rainy season (summer) in waters circulating in shallow aquifers (sandstones), and was found to be delayed in waters circulating in deeper aquifers (volcanic rocks and diabases) in which dilution was observed during the dry season (autumn-winter) only.

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