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A PIXE SET-UP FOR AIR POLLUTION STUDIES IN SOUTH AMERICA*

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ABSTRACT:

As an outgrowth of the PIXE-SP Project an experimental set-up for particle-induced X-ray emission analysis will soon be available at São Paulo. The irradiation chamber, nearly completed, will provide sequential measurements of several (up to 70) sample-targets. The major focus of the Project will be on the investigation of atmospheric aerosols using this set-up. Preliminary results on the urban São Paûlo aerosol characteristics are given for the first time both as an illustration of the analytical potential of the PIXE method and as an example of aerosol data analysis. Samples collected in São Paulo were subject to PIXE analysis at Tallahassee, using the Florida State University facilities. Only results for S and Pb (and Br) are presented.

1. INTRODUCTION

Particle-induced X-ray analysis has proven to be one of the most powerfull analytical tools for the measurement of very small samples[1,2].

In the early 1975 we initiated the PIXE-SP Project with two main objectives: (i) to introduce the PIXE method in the technology of our country, and (ii) for a more specific application, i.e., to use this method for air pollution studies through the aerosol component of the atmosphere[2].

The main activities of our group since the Project began have been concentrated in two areas: (i) the experimental set-up for PIXE analysis, including data processing, and (ii) the first SP-aerosol sampling and subsequent measurements. These parts are described in this paper and some preliminary results for S and Pb (and Br) are presented.

2. THE EXPERIMENTAL SET-UP

The irradiation chamber that is being built since mid'75 is now almost ready to be mounted on line with the Pelletron accelerator of the Physics Institute (IFUSP)[3]. The chamber will be used beyond the split-pole magnet spectrograph in room A of the Pelletron experimental area. Its main features are shown in figures 1 and 2.

The uniformity of the beam flux[1,2] will be accomplished by means of a metallic foil diffuser and proper collimation of the scattered particles. The collimator is made of an

aluminum tube (ϕ_{ext} = 3.8cm, L=5cm) with 4 carbon apertures, two of 4.0mm diam. and two of 5.0mm diam..

The chamber is basically an assembly of the following parts: (i) the long entrance tube that contains the difuser foil and the beam collimator; (ii) the cylindrical tube (ϕ_{int} =15.0cm, H =24.0cm) that houses the targets to be bombarded; (iii) six tubes, also cylindrical, connected to the previous part (central chamber). Two of the tubes are aligned and form part of the system for changing the targets, which uses a conventional slide changer mechanism, operated manually. The third tube is the entrance window for the X-ray detector and the fourth is a plexiglass window for visual inspection of the central chamber interior (e.g. target position). The fifth tube, aligned with the entrance one, has a Faraday cup for beam charge integration. Finally, the last tube-under the central chamber is coupled to the 2" diffusion pump for evacuation on the system. All these parts (tubes and flanges) were made out of stainless steel. Carbon is used for the beam defining appertures and also to line the faraday cup as a means to reduce background radiation.

A Si (Li) detector* with warranted resolution <195eV FWHM at 5.9 KeV is ready for use. Electronics and computer facilities are provided by the Pelletron Laboratory, which includes an IBM 360/44 computer on-line with the data acquisition system[3,4]. To facilitate data processing, a computer program for X-ray spectra automatic analysis was

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*Manufactured by ORTEC Inc., Oak Ridge, Tennessee.

adapted[4,5] to run off-line in the IBM computer.

For the atmospheric aerosol sampling, 5-stage cascade impactors of Batelle design are being made in the machine shop of the IFUSP. Construction of time-sequential filter samplers is also planned for the future.

It is expected that the whole set-up be fully operational by the end of this year.

3. THE FIRST SP-AEROSOL SAMPLING

With the collaboration of the group for aerosol studies of the Florida State University (FSU), Tallahassee, Fl., and the state environmental protection agency CETESB, S.Paulo, the first SP-sampling was carried out between April 29 and May 13 of this year.

FSU samplers (one cascade impactor and three time-sequential filter samplers - "streakers"[6] - were distributed among three sites in the city of São Paulo: (i) downtown (one streaker, at station PR - Praça da República*), (ii) intermediate zone (one streaker, at station MO - Moema*), and (iii) periphery (one streaker and the cascade impactor, at station IF - Instituto de Física in the campus of the University of São Paulo). Fig. 3 gives the exact location of these three sites in the city map.

In total we sampled 14 days for each of the streakers (April 29-May 13) and 9 days for the cascade impactor (April 30-May 8). The air was drawn through the cascade impactor at

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*Both stations from SP air pollution monitoring network (CETESB).

the rate of 1 ℓ/min (measured) and about (not measured) 0.8 ℓ/min through the streaker samplers.

Using the PIXE method, elemental concentration measurements of half of the streaker filters and part of the impactor polistirene slides was done between May 21 and 23 at Tallahassee**. The streaker filters were bombarded each 2mm, corresponding to a period of 2hrs. sampling. A total of about 40 hrs. of accelerator plus 20 hrs. of computer time were needed in these measurements. In the computer processing of the obtained 315 X-ray spectra we used the REX program[7].

4. RESULTS AND DISCUSSION

The results from the PIXE analysis of part of the first SP-aerosol sampling are still under study; however some preliminary data on S and Pb (and Br) are presented here to illustrate the application of the method for aerosol studies.

Some relevant data on the city of São Paulo, important in the present study are the following: altitude - 750m, population ≃ 7.5 million, ~1.1 million automobiles, and >30.000 industries (in the Greater São Paulo area). The combination of automobile plus industrial sources in the contamination of the city atmosphere, and also its relatively close proximity (~50 Km) to the Atlantic ocean (source of marine aerosols), makes the situation in São Paulo

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** One of the authors (LCB) participated in the irradiation of the samples and the data reduction at Tallahassee.

a very complex one to be investigated.

Fig. 4 shows the time variation of sulfur in the three sites for the first week of sampling. This figure also includes the filter pictures (in the sama time scale) that show marked correlations in their structure, for the three sites. The darkness in the filters, related supposedly to the carbon loading in the particulate matter, does not appear to be strongly correlated to sulfur.

Fig. 5 shows similar results for the time variations of lead and bromine at the three sites. They are shown together because lead and bromine from automotive sources should be correlated. However fig, 5 shows that there are large peaks in lead not present in bromine. As to the source of these peaks we need further investigation.

Fig. 6 gives the size distributions for sulfur and lead measured at the IF-station. Also for comparison we give in this figure representative data[8] from two regions in the northern hemisphere: the city of St Louis, Mi and the north of Florida (remote location), USA.

From figures 4, 5 and 6 we see that the last days of the measured period show the onset of an air pollution episode were distinctly higher values contrast with the rather clean weekend values (May 1-2). The whole episode can be seen in the next figure (fig.7) where all six streaker records are presented in a photographic composition. The whole episode lasted for about 5 days. During this episode the concentrations for both sulfur and lead reach very high values even in the city periphery (IF-station). The shape of the size distributions

are characteristic of combustion processes of aerosol production, just as should be expected for sulfur and lead in a urban atmosphere.

In the future we plan to complete the streaker samples measurements using our set-up at São Paulo.

5. CONCLUSIONS

The availability of the PIXE set-up in South America should open new horizons for the application of physics in the investigation and solution of local environmental problems. International cooperation in these matters is also becoming more and more an essential ingredient for the success of any research project. As we know pollution does not respect national borders.

The first SP-aerosol sampling has proven to be very important in the stablishment of new guidelines for future experiments. Although in the present communication we only report the results for few elements (S, Pb and Br) a more complete analysis of all the results is presently underway. Detailed determination of the sample correlation coefficients including the meteorogical data is being done. This may give more information about the aerosol sources and the physico-chemical process in the São Paulo Atmosphere.

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FIGURE CAPTIONS

Fig. 1 - Schematic layout of the apparatus and cut-away view (lower left) of the target chamber: (1) diffuser, (2) collimator, (3) target chamber, (4) Faraday cup, (5) target changer, (6) ion gauge, (7) cold trap, (8) diffusion pump, (9) chamber window, (10) target loader and unloader, (11) target frame, (12) X-ray detector, (13) X-ray collimator and absorber (behind), (14) magnetic suppressor.

Fig. 2 - Irradiation chamber photograph.

Fig. 3 - Localization of the sampling sites (IF, PR and MO) in the city of São Paulo. The continuous line is the city limit and the downtown area is shown by the dashed line.

Fig. 4 - Time variation of sulfur amounts (in nanograms) for the three sites. In the upper part, in the same time scale, the filters photographs are included (irradiation spots are in some cases visible).

Fig. 5 - Time variation of lead and bromine amounts (in nanograms) for the three sites.

Fig. 6 - Size distributions for sulfur and lead concentrations at IF-site. Impactor stage: 1(>4μ), 2(2-4μ), 3(1-2μ),

4(0.5-1μ), 5(0.25-0.5μ). Data from St. Louis (STL) and North of Florida (NFL), U.S.A., are included for comparison.

Fig. 7 - Photographic composition of the filter samples for the whole sampling period.

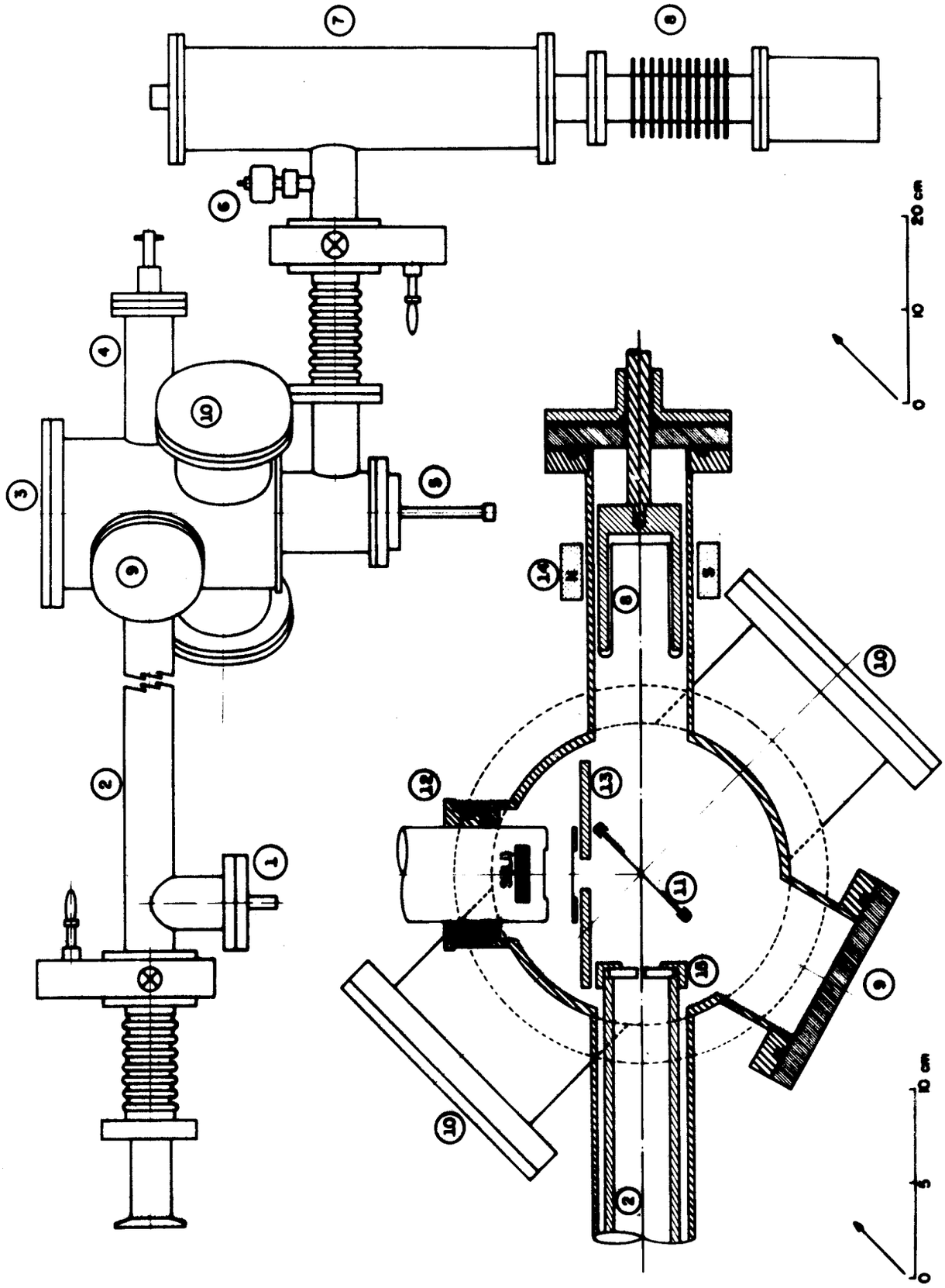
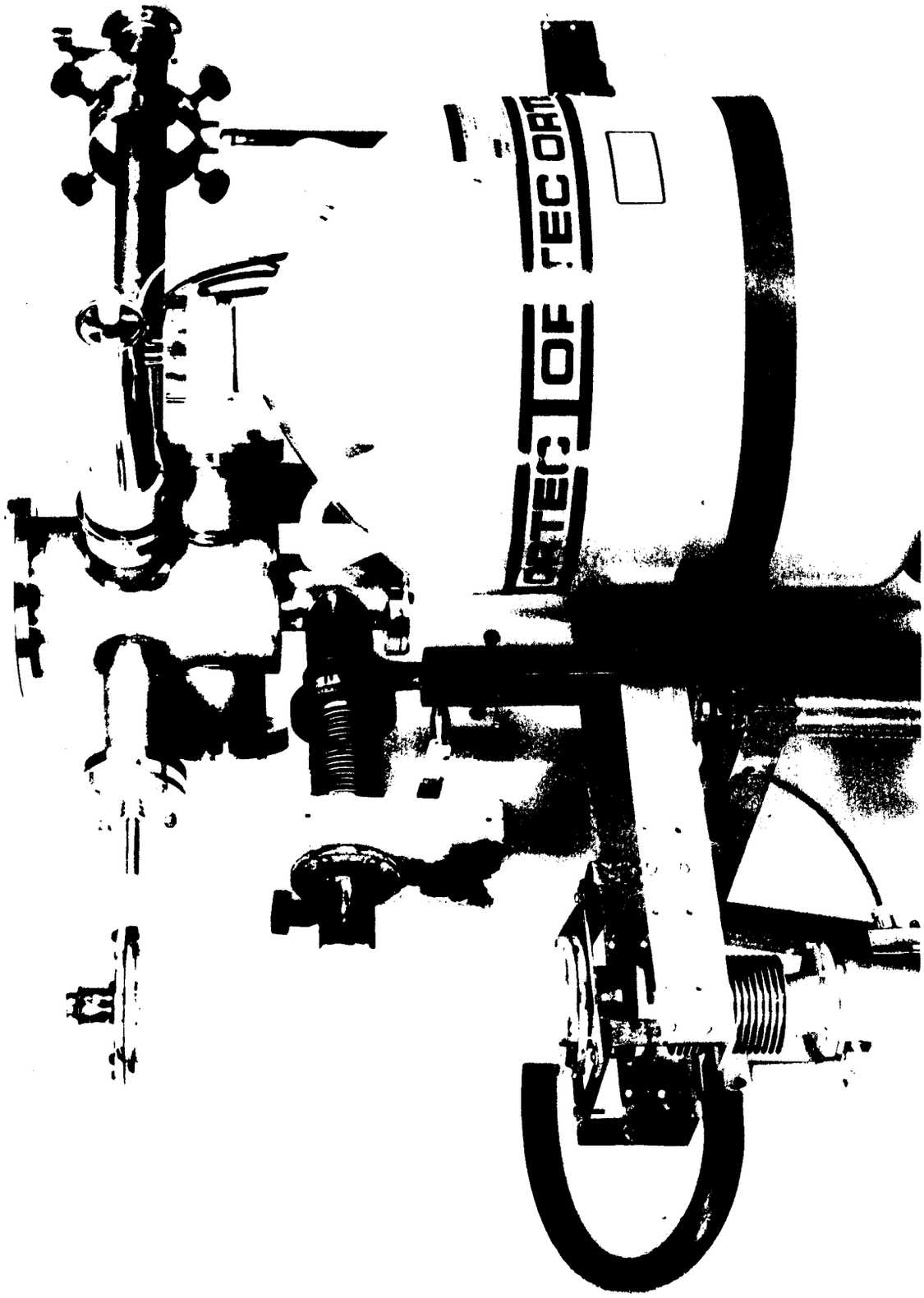


FIG. 1



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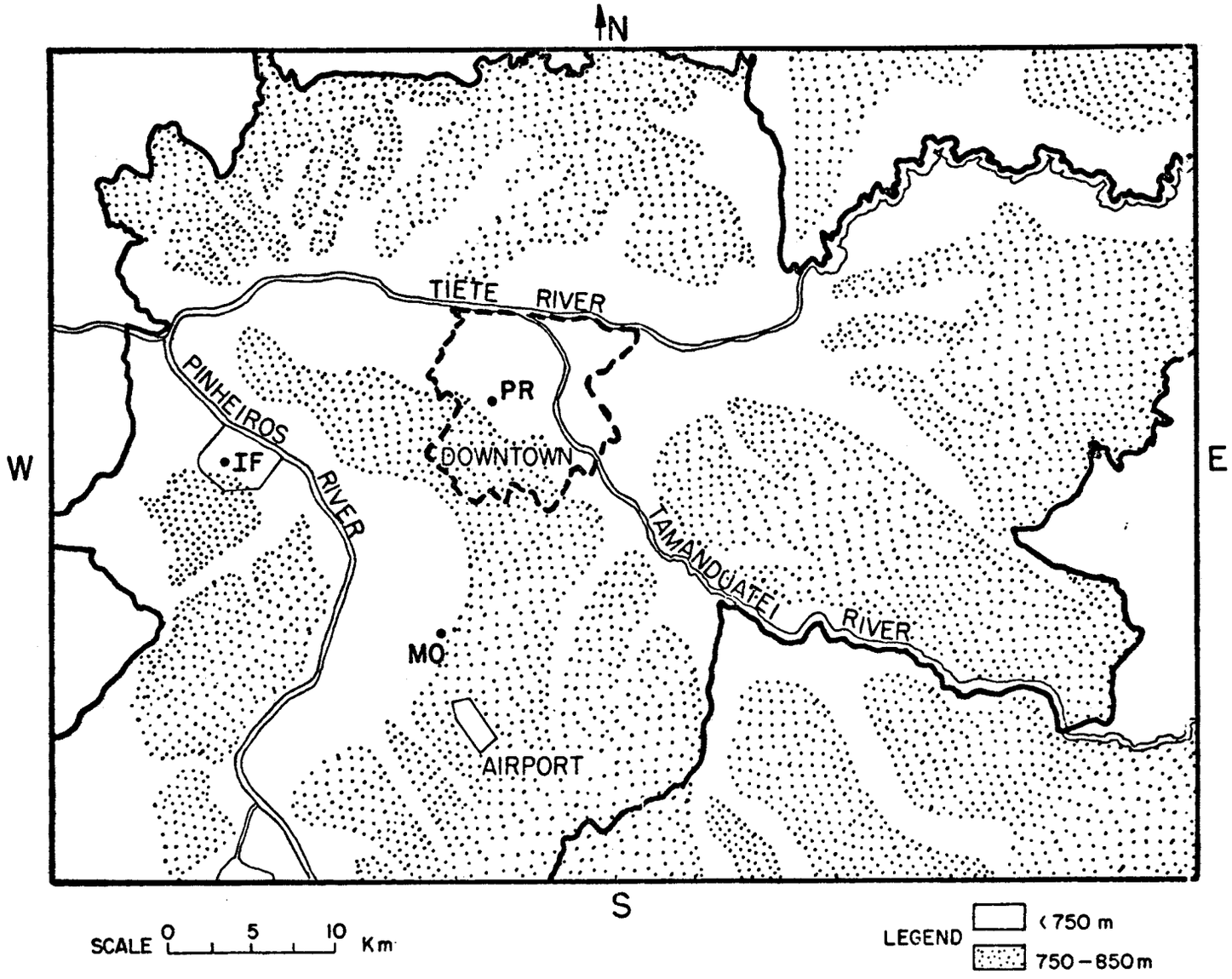
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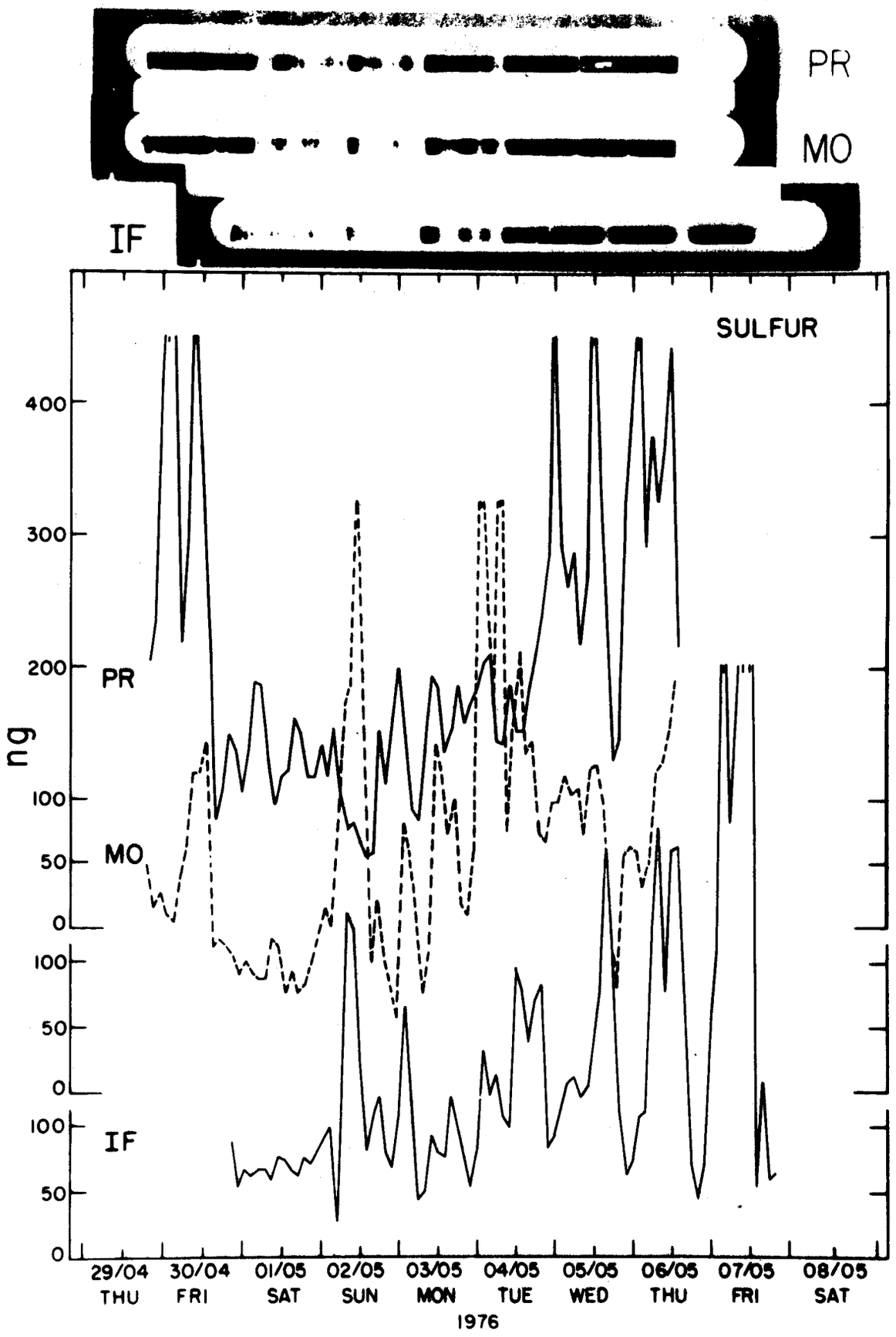
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FIG. 4

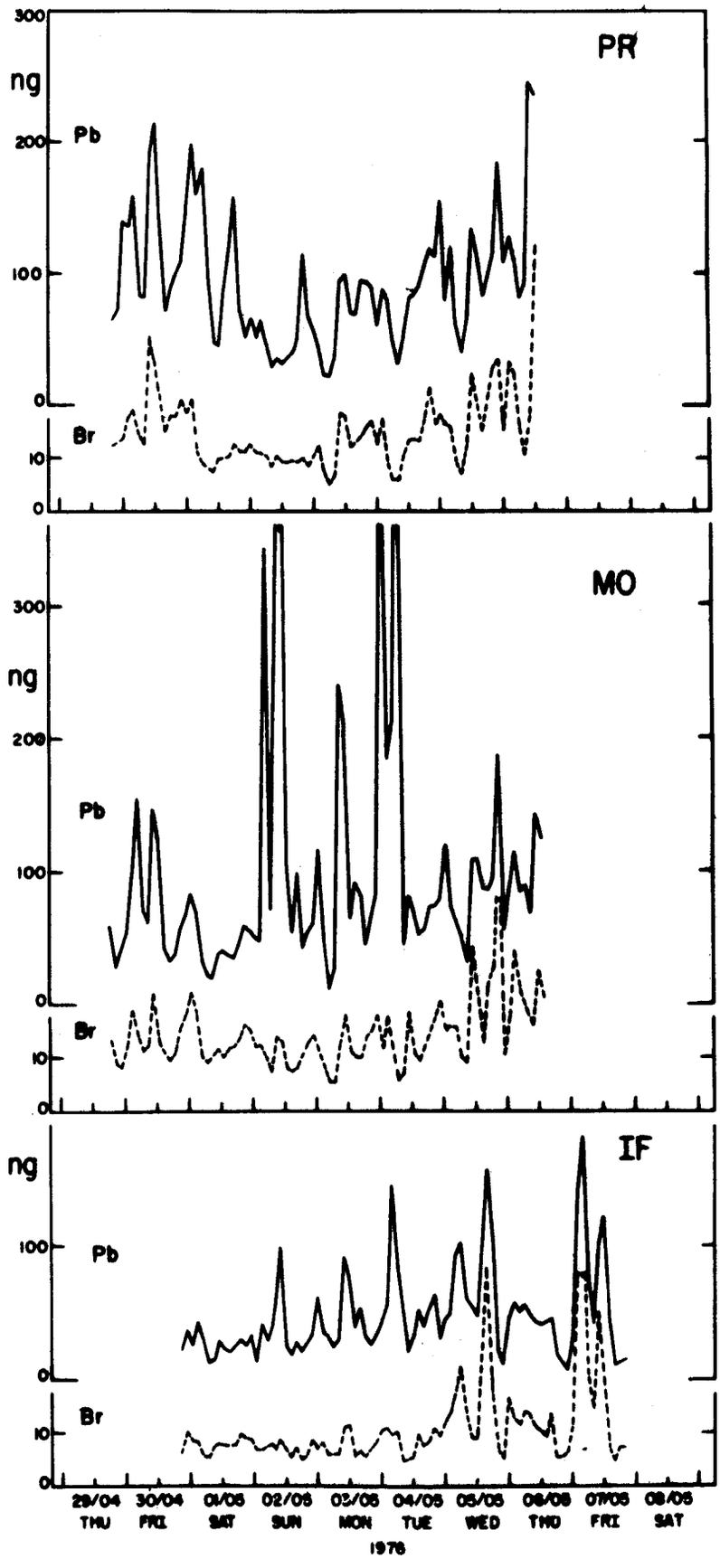
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FIG. 5

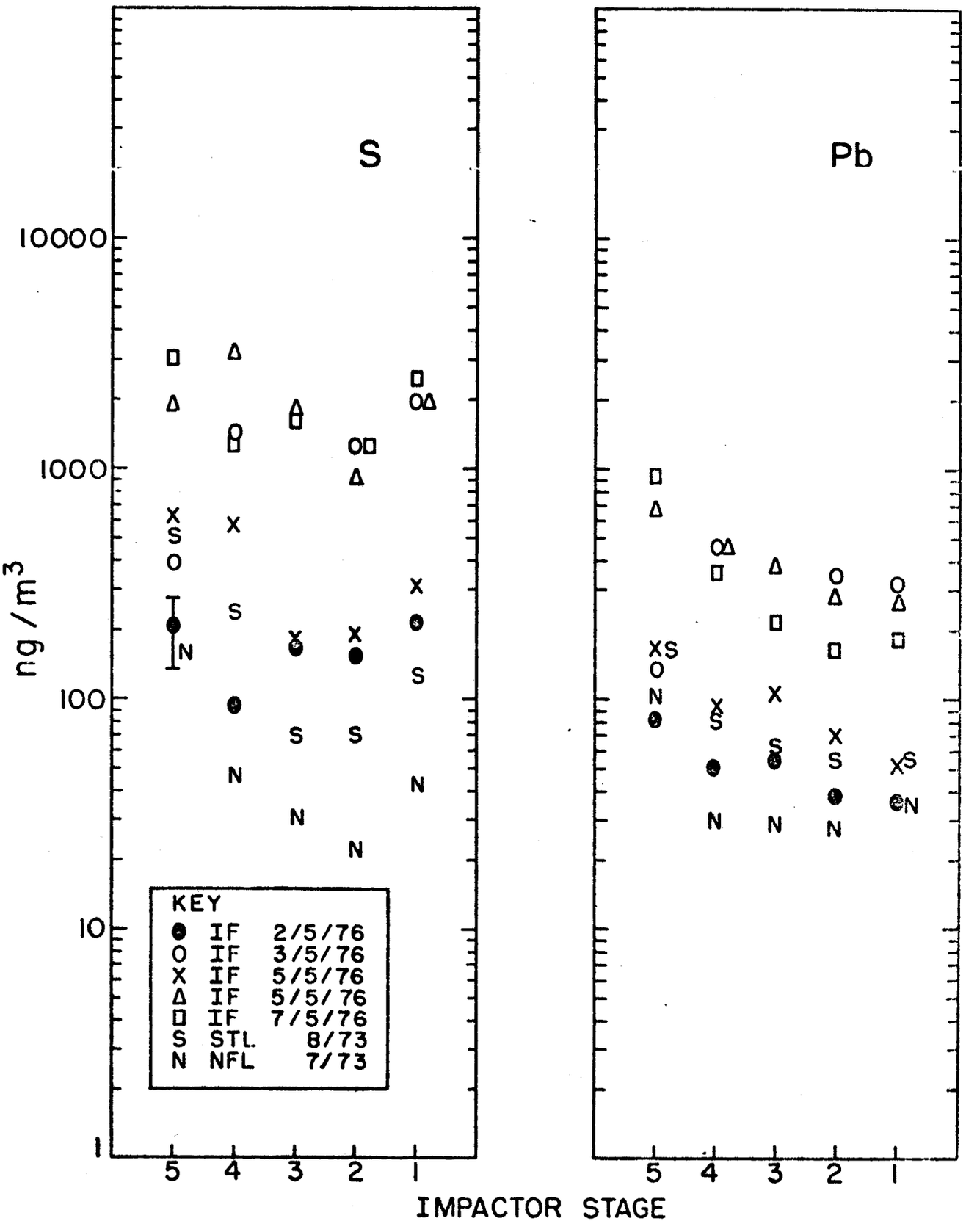
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FIG. 6

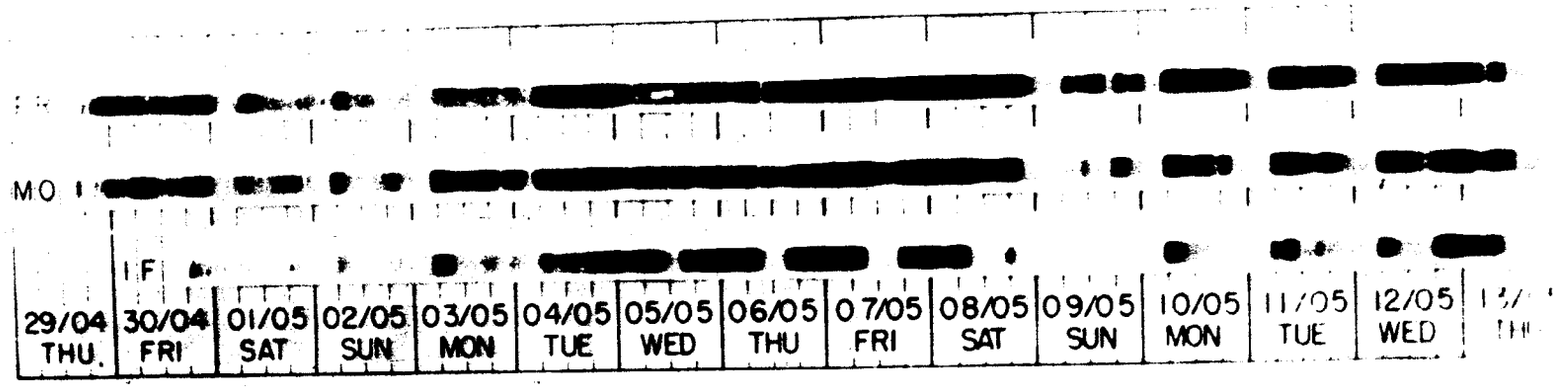
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FIG.7