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RESUSPENSION IN THE PALOMARES AREA OF SPAIN

A SUMMARY OF EXPERIMENTAL STUDIES

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Abstract

This paper presents some experimental data concerning resuspension in the area of Palomares, Almeria, Spain. The spacial and temporal variations of the resuspension paramemeters are presented together with correlations of the airborne plutonium concentrations with wind speed and frequency of wind speed.

Key words: resuspension, Palomares, plutonium, resuspension factor, wind speed, mass loading, dust loading.

1. INTRODUCTION .

Resuspension has been recognised as a long-term potential pathway for human exposure to contaminants in the soil. Radioactive material deposited onto the ground can be resuspended into the air by wind or by other disturbances. In addition, resuspension and subsequent re-deposition in agricultural areas may lead to contamination of crops and foods derived from grazing animals.

In southwest Spain (Palomares), an area of 226 Ha has been contaminated by Pu and Am, following the accidental release of four nuclear warheads and subsequent spread of fissile material. Inhalation is the most important potential contamination pathway due to the climatology of the area (a typical agricultural Mediterranean area with little precipitation) which favours resuspension. A better undestanding of resuspension·processes will help to validate model predictions. This paper presents some experimental data obtained in the area of Palomares.

2. DESCRIPTION OF THE STUDY AREA

The area of contamination around Palomares covers some 226 Ha of cultivated and uncultivated land. The cause of the contamination was the collision of two military aircraft during a mid-air refuelling operation in June 1966. The two aircraft were destroyed and four thermonuclear bombs fell onto the area, three of them on land and one into the Mediterranean sea. Two bombs failed to deploy their parachutes and, on impact, a conventional explosion was followed by the release and ignition of part of the fissile material. Although some countermeasures were taken (Iranzo et al., 1988), some residual contamination remained. Operations of removal of the contaminated soil began inmediatly after the accident (January 16, 1966) and ended in January 1966.

Farming procedures in the area are typical of Mediterranean agricultural areas with little precipitation, of the order of 200 mm, and artificial means of irrigation are required. Until recent years, a system of irrigation by flooding with water pumped from wells in the area was used, whereas, at present, the drop to drop system is employed. This system consist in a plastic pipe passing closely along the plants. The pipe is disposed with a hole, close to the plant, to allow the exit of water. The main crops in the area are tomatoes, barley and alfalfa. The production of water melons, melons and peppers has greatly increased in recent years. Other products represent a small percentage of the total crop production.

3. EXPERIMENTAL PROGRAMME

After the accident, the level and the amount of spread of surface α-contamination was measured. The highest levels of contamination were found in non-cultivated areas located between small hills 1500 m southeast of the town. Most of the area (120 Ha) showed an activity lower than 12 kBq m^{-2}, 87 Ha showed an activity between 12 and 120 kBq m^{-2}, 17 Ha showed an activity between 120 and 1200 kBq m^{-2} and 2.2 Ha showed an activity greater than 1200 kBq m^{-2} (Iranzo et al., 1986; see figure 1).

When the environmental surveillance programme began, six study plots of 50 m x 50 m were established within the areas of high, medium and low surface α-activity . Values of soil plutonium concentration are shown in table 3. Two control plots were also established. Periodic soil sampling has been conducted in all of these plots since 1966 from nine points along the diagonals, equidistant from each other. Each soil sample was 3 cm diameter and 45 cm deep and divided into five sections (Iranzo et al., 1990). Surface samples have also been taken. A metallic parallelogram (25 cm x 25 cm x 5 cm) was used to collect the top 5 cm surface soil samples. The activity in these soils shows a heterogeneous distribution.

Samples of radioactive aerosol have been collected continuosly using high-volume samplers with a flow rate of 1 m^3 min^{-1} placed at various locations in the area. Two of these have been located in the area around the impact point of bomb number 2 (stations 2-1 and 2-2 in Area 2), one in the village of Palomares (station P) and another (statation 3-2) in the area around the impact point of bomb number 3, Area 3 (Iranzo et al., 1970). These locations are shown in figure 1. Air sampling started in the third week of June 1966, at stations 2-2, 3-2 and P. Continuous air sampling was made at stations 2-1, 2-2, 3-2 and P since the fegenning of July 1966 until 1969. In 1970 sampling was carried out at stations 2-2 and P until 1991. Sampling at station 2-1 started in 1984. At present sampling is been made continuously at stations 2-1, 2-2 and P. Since July 1987 two of the samplers, one from Area 2 (station 2-2) and the one located in the village (station P) were equiped with a PM-10 size-selective inlet, which exclude particulate material of diameter larger than 10 μm). These samplers operate at a flow rate of 1.7 m^3 min^{-1}. The inlet is designed in such a way so as to allow the passage of particles smaller than 10 μm, whereas larger particles impact and are retained on the collection shim placed inside the inlet.

In 1992, two high-volume cascade impactors (flow rate 1.13 m^3 min^{-1}) were located in Area 2 at differents heights, 1.80 m and 3.0 m. The impactors fractionate the airborne particles into six fractions. The particle upper cutoff diameters for the first five stages were 7.2, 3.0, 1.5, 0.95 and 0.49 μm respectively.

4. SUMMARY OF RESUSPENSION RESULTS

A review of the data obtained during the experimental radiological surveillance programme carried out in the area of Palomares since the time of the accident has allowed the following resuspension parameters to be measured.

4.1 RESUSPENSION FACTORS

The resuspension factor is defined as the ratio between the concentration in the air at some reference height and the quantity of the contaminant per unit area of ground surface. Resuspension factors have been calculated based on annual average Pu air concentrations obtained over the years and the surface contamination of Pu in the top 2 cm of soil (Iranzo and Salvador, 1970; Iranzo et al., 1987; Iranzo et al., 1988). The data obtained indicate that the resuspension factor decreases progressively with time, from an initial average value of the order of 10^{-7} m^{-1} to values of the order of 10^{-9} m^{-1} some months later, and in the order of 10^{-9} - 10^{-10} m^{-1} after several years.

Resuspension factor around Palomares, after several years, varied from 2.8 x 10^{-10} to 2.7 x 10^{-9} m^{-1}, depending on the initial surface contamination. As has been observed by other workers the resuspesion factor appears to be higher in those areas with the lowest initial deposits (Garland et al.,1990). The interannual variability is about 40 %.

The reduction with time of the resuspension factor, K, calculated from experimental data obtained at station 2-2 (see figure 1) can be related to time t (in months) by the following exponential.

$$K\ (m^{-1}) = 1.29 \times 10^{-9} \exp [\ -\ 0.00976\ t\ ]$$

Fig 2 shows the variation of the resuspension factor with time after the release of contamination.

4.2 DUST LOADING FACTORS

The dust loading factor, S_e, is defined as the ratio between the activity concentration in the air, C_a, and concentration in soil, C_s.

$$S_e\ (kg\ m^{-3}) = \frac{C_a\ (Bq\ m^{-3})}{C_s\ (Bq\ kg^{-1})}$$

Dust loading factors have been calculated based on weekly air concentration measurements taken during two consecutive years at two different stations located in the Palomares area (one sited in a cultivated zone and the other one in the urban area) and the average soil concenttration in the study plots. These data show an average dust loading factor of around 100 µg m^{-3} with a large standard deviation. The airborne particle concentrations measured in these two consecutive years show an average of 93 $\pm$ 39 $\mu g\ m^{-3}$ at the station located in a cultivated area and 100 $\pm$ 32 $\mu g\ m^{-3}$ at the station in the urban area.

Figures 3a and 3b show the dust loading factor in the urban area, in 1983 and 1984 respectively, while figures 4a and 4b show the dust loading factor during the same years at the station in the cultivated area (Iranzo et al., 1987). There is a very clear dependence on the time of year. The figures show peaks in summer time, decreasing rapidly to rather low levels with the onset of winter.

Dust loadings of up to 12 $mg\ m^{-3}$ have been found at a distance of about 100 m from where a very large pool for irrigation purposes was being constructed using heavy earth moving equipment. It is most likely that even higher levels would have been found on the construction site.

During the years of the experimental period, crop samples were taken from a number of contaminated areas. It was found that, in general, dust concentrations on plant surfaces, which is defined as the quotient between the activity per unit of mass of the fresh vegetable (Bq/g) and the surface soil activity concentration (Bq/g), were dependant on type of crop, the area from which it was harvested and the part of the crop which was analysed. In general, the highest dust concentration values correspond to the parts of the crop with characteristics most favourable for interception and retention. Similar observation was made in studies on soil-to-plant concentration ratios (Iranzo et al., 1988).

Table 1.a and 1.b shows data obtained from olive plants collected at the beginning of September 1983 (Iranzo et al., 1989) and the amount of rain in August 1983, respectively.

It is apparent from the table that the value obtained for the olive fruit is about fifty times lower than that obtained for the leaves. These differences can be explained by the larger surface area of leaves in relation to fruit per unit mass. Given the height of the olive tree, it is unlikely that soil splash would have made any significant contribution to the surface contamination (Aragòn et al., 1992).

Table 2 shows data obtained from barley samples taken during May 1983.

The results are similar to those obtained for the olive plant in that the grain exhibits a retention capacity approaching an order of magnitude lower than the espicule. In this case there was no rain in the area since mid-February so that field loss in run-off can be considered neglegible (Iranzo et al., 1988). A similar explanation can be proferred to that for the olive plant (i.e. the grain and straw have lower surface areas per unit mass than the espicule. In addition, the espicule will offer a better interception surface for atmospheric material

4.4 CORRELATION OF AIR CONCENTRATION WITH WIND SPEED

Studies involving the correlation of wind speed with air concentrations have been carried out in order to assess the importance of wind speed on resuspension.

Wind speeds measured in 1988 (Aceña, 1990a; Aceña, 1990b; Aceña, 1991) were classified into four categories: 0 to 3 m s^{-1}, 3 to 5 m s^{-1}, 5 to 7 m s^{-1}, and higher than 7 m s^{-1}. Average air concentrations were correlated with average wind speed during each sampling period. The highest correlation was found at the site in the urban area. A linear correlation coefficient of 0.79 was found for a number of 14 pairs of data (see fig 5). No significant correlations in the other sampling stations were observed.

It will be noted that among the several factors which influence the amount of resuspended material, the duration of each wind class should be considered. Consequently, the variation of resuspended air concentrations according to frequency of wind speed has been studied. Only significant correlations were observed in the urban area. A linear correlation (r= 0.73 for 14 data pairs) was found between the frequency of winds higher than 7 m s^{-1} and the plutonium activity concentration in air (see figure 6). Also, a slightly better correlation (see figure 7) was found between plutonium concentration in air and the frequency of windspeed greater than 5 m s^{-1} (r=0.76 for 14 data pairs).

CONCLUSIONS

1) The resuspension factor decreases exponentially with time, from an initial average value of the order of 10^{-7} m^{-1} to values of the order of 10^{-9} m^{-1}, some months later, and of the order of 10^{-9} - 10^{-10} m^{-1} several years later.

2) The dust concentration due to resuspension averaged 93 $\pm$ 39 μg m^{-3} (cultivated area) and 100 $\pm$ 32 μg m^{-3} (urban area). Calculated dust loading factors and average particle concentrations also show a very good agreement with airborne dust concentrations at these sites.

3) In general, the highest values of dust concentration on plant surfaces correspond to those parts of plants which exhibit the most favourable characteristics for the interception and retention of particles. Thus, demonstrating the importance of the deposition of resuspended material.

4) The best correlation between wind speed and airborne plutonium concentration was found at the site located in the urban area of Palomares village. For this location, a correlation cofficient of 0.76 (14 pairs of data) was observed between the frequency of wind speed greater than 5 m s^{-1} and plutonium concentration in air.

5) Resuspension of radioactive material following a contamination episode is the most important process in dose estimates, espcially because agricultural crops may become contaminated. This is espcially true in areas influenced by a Mediterranean climate and subject to little rainfall.

TABLE 1.a. Dust concentration on different parts of an olive plant

Olive (fruit) ...... 0.03 mg g^{-1}
Olive (leaves) ...... 1.40 mg g^{-1}

TABLE 1.b. Amount of rain in August 1983

August 23 .... 42 mm
August 27 .... 16 mm

TABLE 2. Dust concentration on different parts of a barley plant

Barley grain	1.9	mg g^{-1}
Barley straw	1.7	mg g^{-1}
Espicule (grain hask + hair)	15.0	mg g^{-1}

TABLE 3. Plutonium concentration in soils of the selected plots

plot	Pu-239 + Pu-240 (Bq/g)		
	average	maximum	minimum
2-1	0.44	1.60	0.03
2-2	2.06	3.31	0.80
3-1	1.10	2.00	0.05
3-2	1.79	5.70	0.23
5-1	0.13	0.30	0.02
5-2	0.29	0.99	0.01

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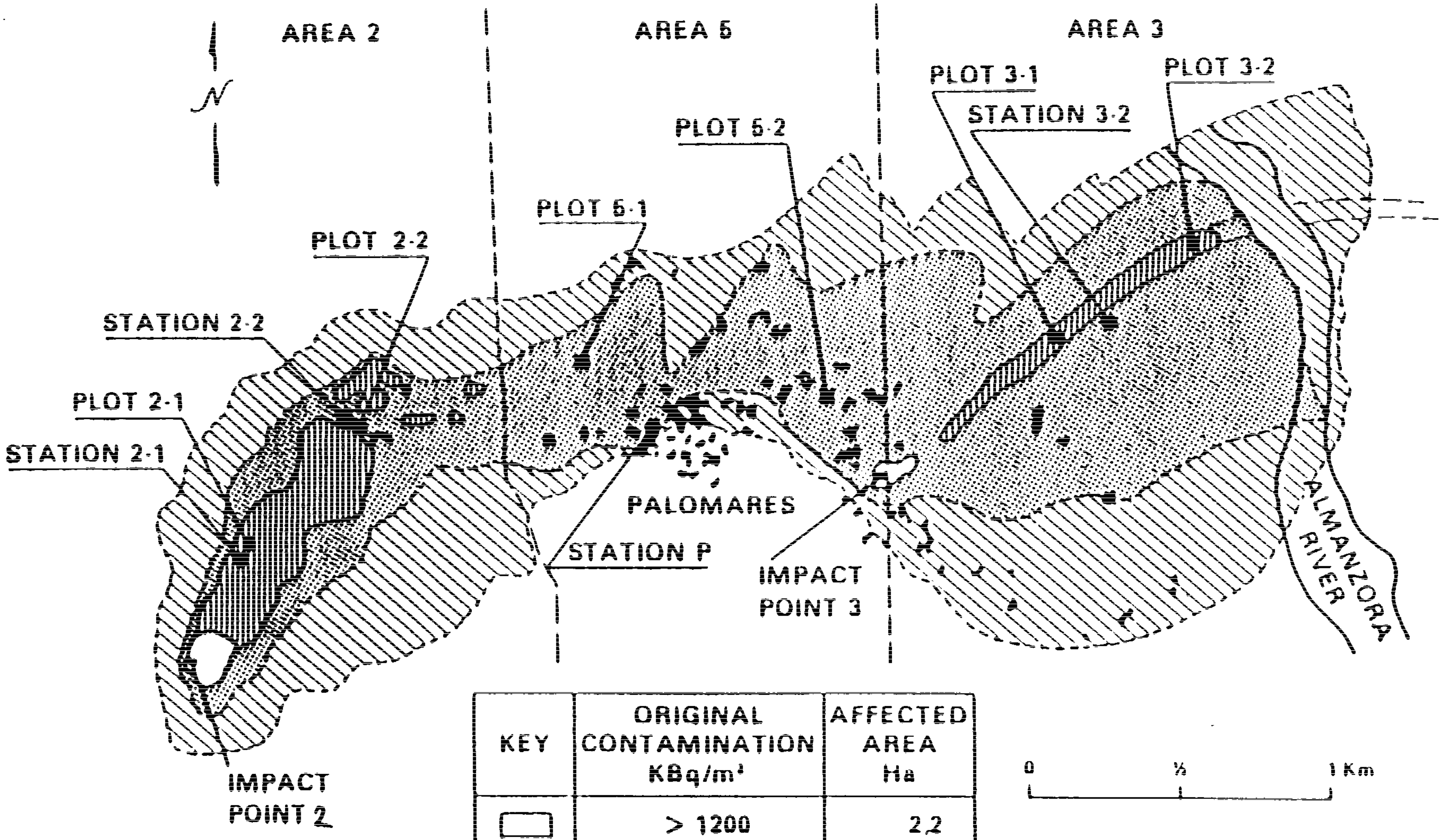
FIGURE 1. PALOMARES AREA: ORIGINAL CONTAMINATION LEVELS AND LOCATION OF SAMPLING STATIONS

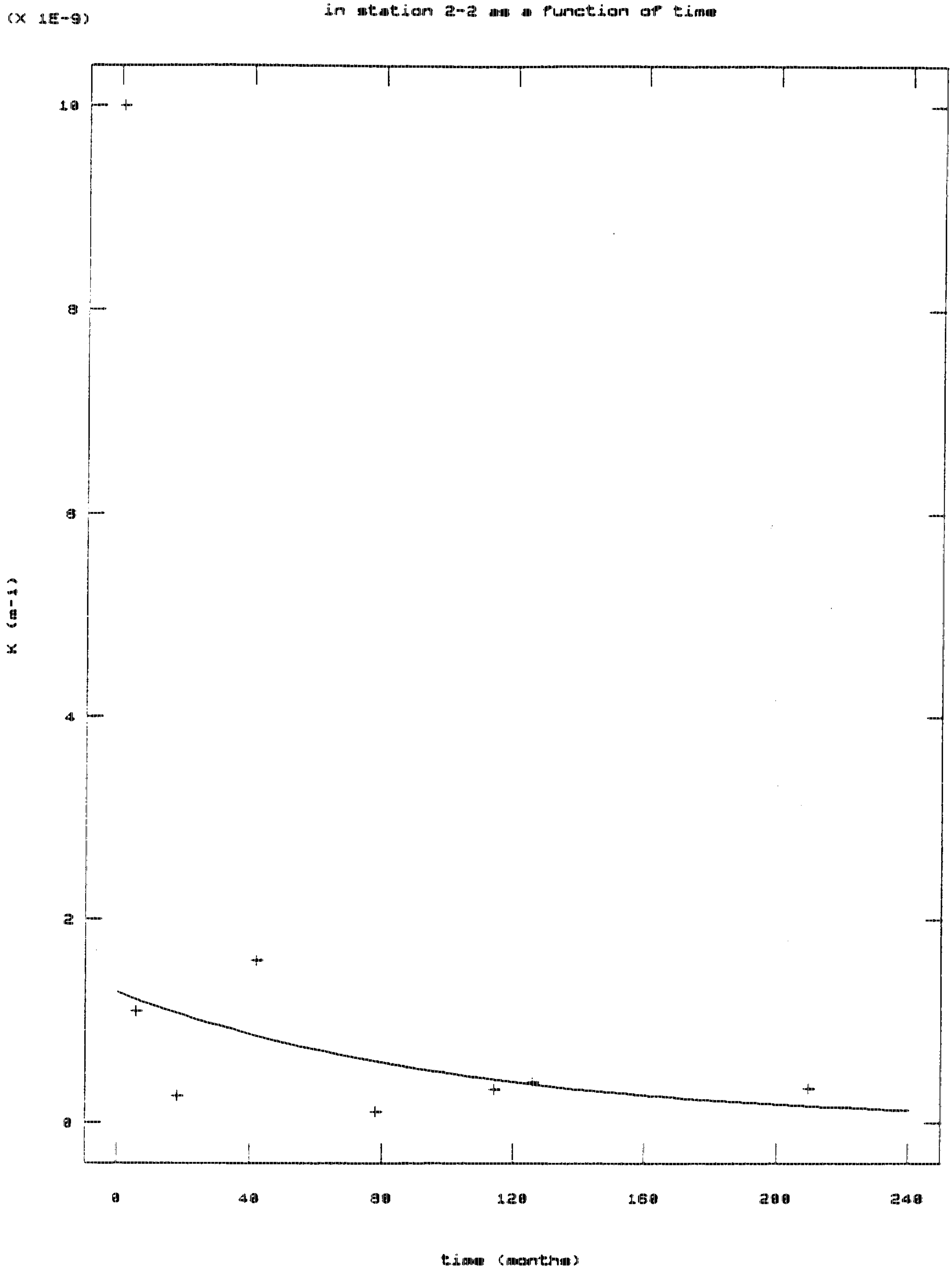


KEY	ORIGINAL CONTAMINATION KBq/m²	AFFECTED AREA Ha
(blank)	> 1200	2,2
(vertical lines)	120-1200	17
(dotted)	12-120	87
(diagonal lines)	< 12	120

● STATION
■ PLOT

Fig.2. Variation of resuspension factor

in station 2-2 as a function of time



Dust loading factors

Urban area

1983

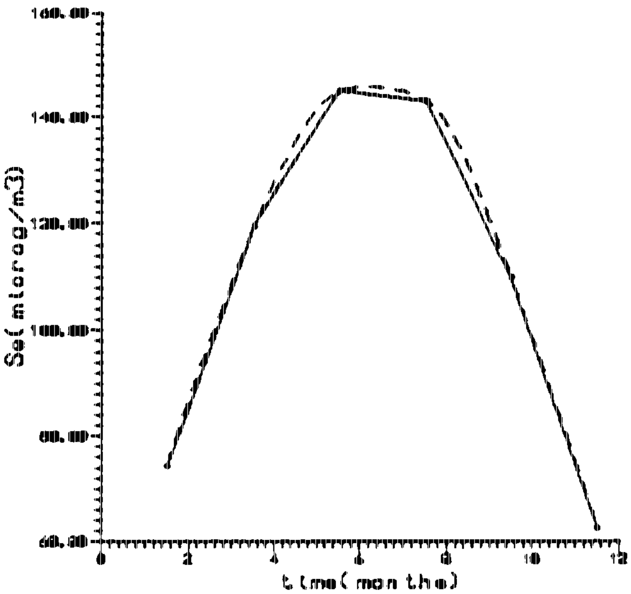


Figure 3a

1984

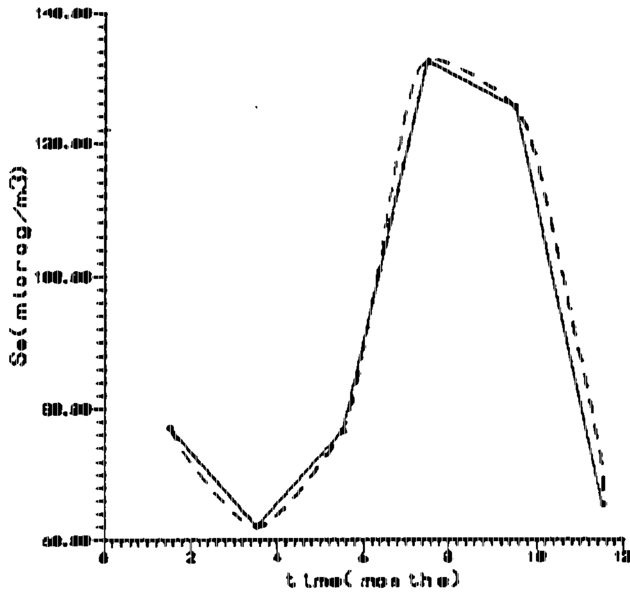


Figure 3b

Cultivated area

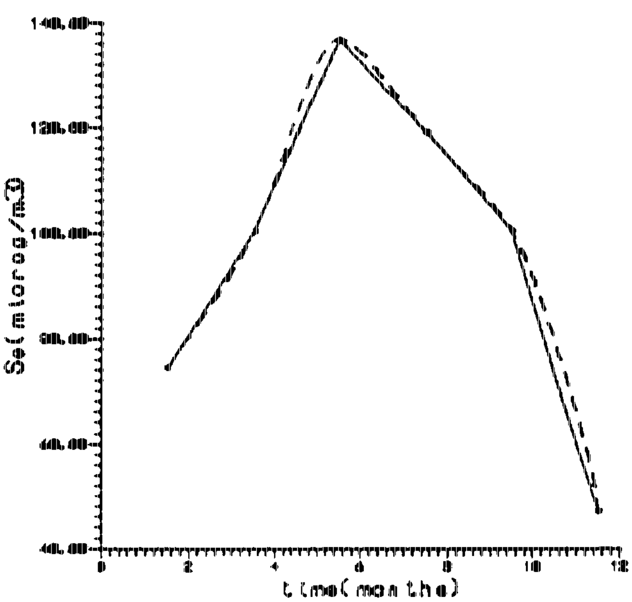


Figure4a

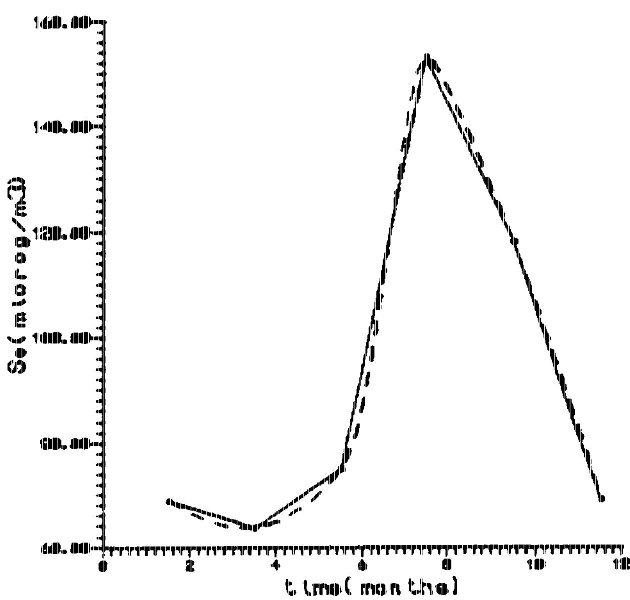


Figure 4b

Figure 5. Relationship between Plutonium

airborne concentration and wind speed

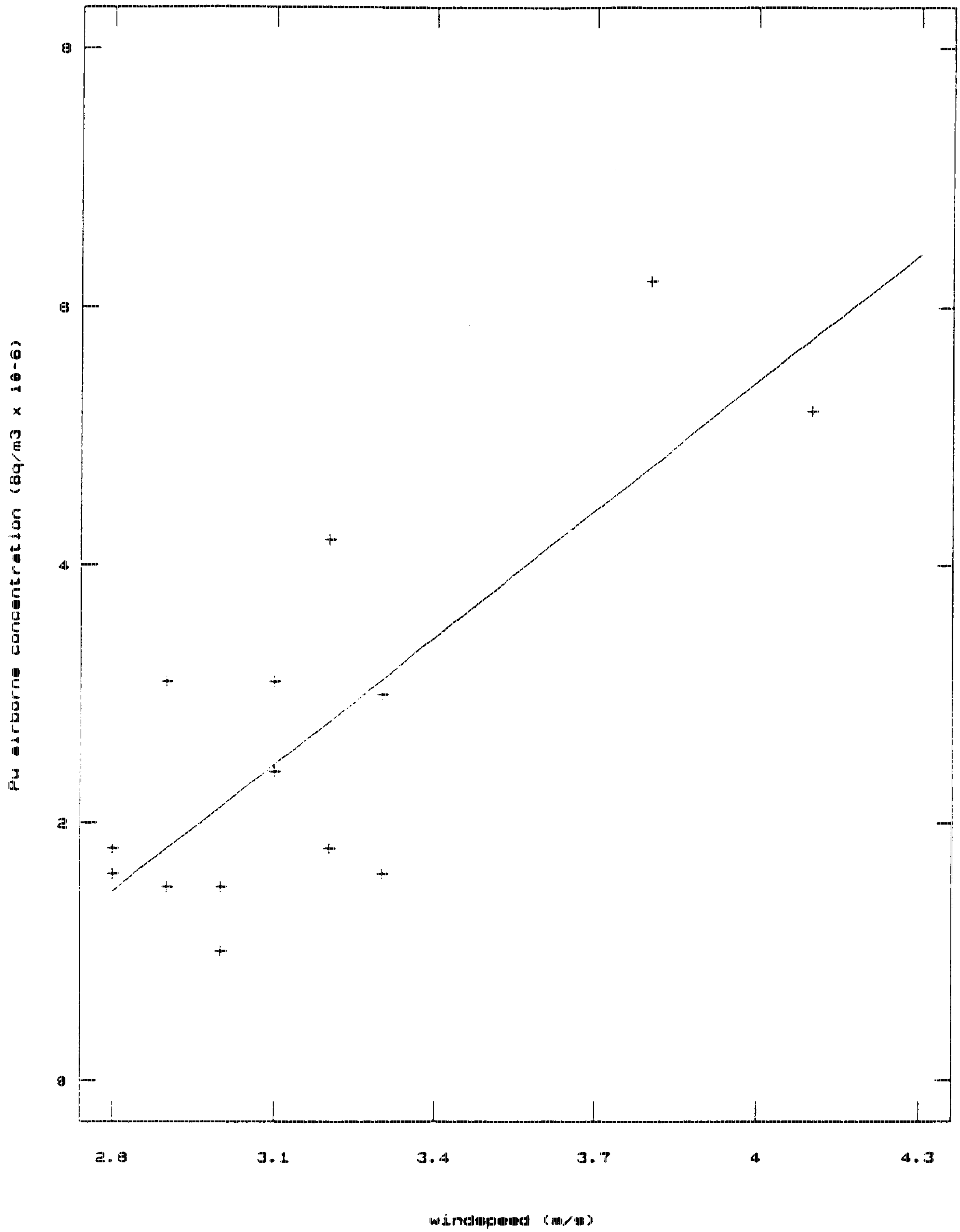


Fig.6. Relationship between Pu airborne

concentration and windspeed frequency

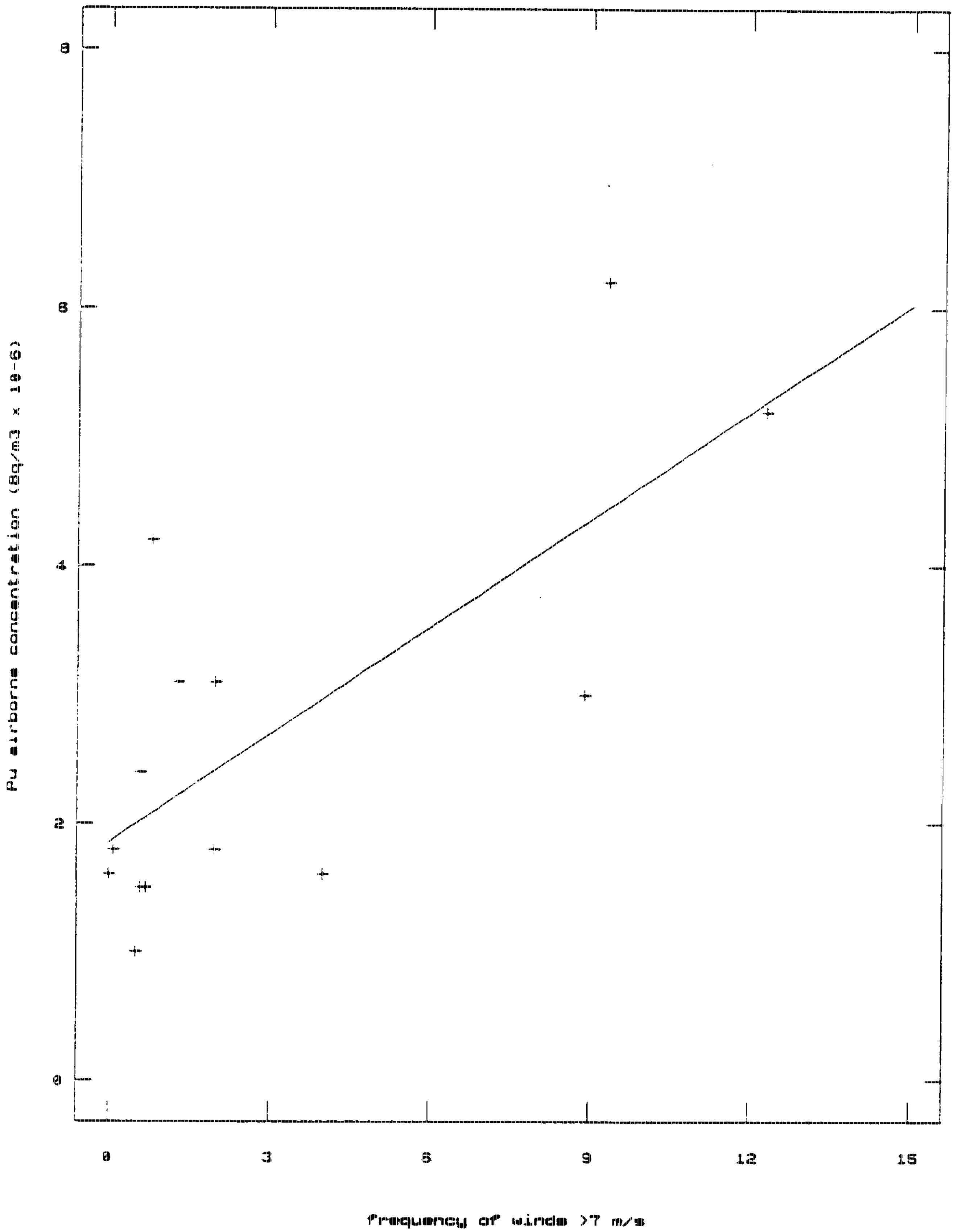


Fig. 7. Relationship between Pu airborne

concentration and windspeed frequency