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LABELLING SEWAGE SLUDGE WITH IODINE 131.

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by

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SUMMARY

A method of labelling digested sewage sludge with iodine 131 was investigated for the South Australian Engineering and Water Supply Department, who are investigating the possibility of disposing of sewage into the ocean, and hence are interested in tracing the movement of the sludge in the sea, in order to determine whether pollution of neighbouring beaches would occur. Satisfactory labelling of both solid and liquid phases was achieved.

INTRODUCTION

The South Australian Engineering and Water Supply Department is interested in disposing of digested sewage sludge into the ocean through a 2" diameter pipeline extending 2 miles offshore, and capable of delivering a daily batch of 35,000 gallons in 2 hours. The scheme will be possible only if local councils are satisfied that beaches in the vicinity will not be polluted, and the Isotopes Section of the A.A.E.C. was approached to examine the possibility of tracing with radioactive isotopes the movement of a trial batch of sludge in the sea. It is proposed either to construct a 4" pipeline for the test, in which case 35,000 gallons of sludge would be delivered over a period of 24 hours, or to deposit this volume of sludge from a barge, at a point corresponding to the end of the proposed pipeline.

Iodine 131 was selected as the most suitable available radio-isotope. The minimum detectable concentration of Iodine 131 is of the order of 10^{-3} μc/gallon, so that one curie added to 35,000 gallons of sludge would be detectable (by scintillation counters) when diluted by a factor of more than 12,500 although sensitivity would be less on the sea bed owing to the higher background.

Tracing of the solid phase of the sludge was thought to be of primary importance, so a method of attaching the iodine firmly to the solids was sought. Previous work with Iodine 131 indicated appreciable adsorption of this isotope on muds, glass, etc., when no carrier is present, so investigations were based on this premise.

EXPERIMENTAL

Preliminary Tests

The freshly treated sludge has a density of 0.98 g/cc, and is at a slightly higher temperature than the ocean. It is composed of 15 - 20% solid matter and 80 - 85% liquid (by volume). Approximately 5% by weight of the solids are colloidal, but may be precipitated by the addition of ferric ion (3% by weight is the figure quoted by S.A. Engineering and Water Supply Dept.). The larger solids settle in less than half an hour on standing, both from undiluted and highly diluted sludge.

Complete solubility of the solids was not attained with any of the reagents tried, (e.g. concentrated nitric, hydrochloric and perchloric acids, and caustic soda), while prolonged heating produced a coagulated suspension; however cold nitric acid ($\sim$5N) gave partial solution together with a uniformly fine suspension which settled slowly. All counting was done with the solid phase in this form.

50 μc of iodine 131 (in sodium thiosulphate solution) supplied by the Radiochemical Centre, Amersham, was diluted to 1 litre with demineralised water. This stock solution was used in all subsequent experiments.

Adsorption Tests.

1. The partition coefficient of iodine 131 between the solid and liquid phases of the sludge was first investigated as follows -

50 ml of iodine 131 stock solution was added to 50 ml of sludge in a stoppered container. The mixture was shaken thoroughly, and a 20 ml sample centrifuged. The total volume of each phase was measured, then 10 ml of supernatant solution (containing the colloidal matter) removed and counted. A scintillation counter was used in all experiments except where otherwise specified. The solid phase was washed repeatedly with very dilute potassium iodide solution to prevent desorption, drained, and dissolved in nitric acid. The acid solution was made up to 50 ml, and a 10 ml aliquot was counted. The total activity in each phase was then calculated. A control experiment using demineralised water was run concurrently to check on losses due to adsorption on glass vessels, washing etc., and samples from each experiment counted daily.

Daily results were plotted on semi-log paper and decay curves for iodine 131 drawn from the initial activity in the control and in each phase of the sample. Subsequent points falling above or below these curves gave an immediate indication of increased or decreased adsorption on the solid phase. Any adsorption on the glass walls would have shown up as a drop in activity of the control.

Adsorption on the solid phase rose from an initial 9% of total activity in sample to 45% after 5 days. This result was obviously unsatisfactory, hence the experiment was repeated with the following variations:-

(a) A mechanical shaker was used to ensure thorough mixing of the samples. This increased the initial adsorption (i.e. adsorption within 1 hour), to 25% but in 8 days an increase of only 4% to 29% was obtained.

(b) The colloidal matter was precipitated by the addition of Al^{+++} ions (as the sulphate) before adding iodine 131 to the sludge. The initial adsorption of 20 - 25% remained constant over 8 days. See Fig. 1.

2. At this stage, an experiment based on work done in Japan[2] on co-precipitation of iodide ions by metallic oxides was carried out.

0.15 mg of Fe^{++} (as the sulphate) was added to 10 ml of sludge and 5 ml of iodine 131. The sample was shaken and left exposed to air for several hours, iodide ions being strongly adsorbed and carried down by the fine flocculent precipitate of ferric hydroxide formed. After further shaking, 1 ml of the suspension was evaporated to dryness on a planchet and counted, using an end-window G.M. tube in a lead castle. A further 2 ml of the suspension were centrifuged, the volume of each phase measured, and 1 ml of the supernatant liquid evaporated on a planchet and counted. The total activity in each phase was calculated, that in the solid being found by difference. This procedure was repeated daily.

An initial adsorption of 30% on the solid rose to 87% after 3 days, but it was decided that direct adsorption on the sludge would be preferable, as density differences could mean that the precipitate of ferric hydroxide with adsorbed iodine might not follow the movement of the sludge when released in the ocean.

As it was evident that a large proportion of the adsorbed activity was on the colloidal matter, it was decided to allow time for adsorption to take place before precipitating the colloids with ferric ion.

3. Two samples each containing 100 ml of sludge and 50 ml of iodine 131, (plus a control) were allowed to stand, with frequent shaking, for 2 hours and 24 hours respectively. Ferric chloride (5% Fe^{+++} by weight) was then added. After thorough mixing, 15 ml samples were centrifuged, the volume of each phase being measured. The total activity of each phase was found as in previous experiments.

An initial adsorption of 94% was achieved with both samples, and remained constant at this level for 8 days, i.e. no improvement was gained by increasing the adsorbing time beyond 2 hours. See Fig. 2.

Desorption Tests

1. To check on desorption from the solids on dilution of the sludge, samples of activated and precipitated sludge were diluted by factors of 10 and 100 with sea water, then shaken mechanically for 2 hours. The solid phase was then separated and both phases counted. No desorption occurred in either case.

2. The effect of prolonged washing was next investigated.

2.5 ml of activated and precipitated solid sludge were diluted to 250 ml with sea water, then shaken continuously over a period of 8 days. 15 ml samples were removed each day, centrifuged, and each phase counted as described above, the total activity in each phase being calculated.

The initial activity on the solid phase was 93% of the activity added to the sludge. After 8 days this had fallen to 79% of the added activity, or 85% of its original value, i.e. at a dilution of 100, only 15% desorption had taken place over 8 days. See Fig. 3.

DISCUSSION

The above results indicate that the use of iodine 131 as a tracer for the solid sludge is feasible. Certain difficulties remain to be resolved, e.g., if 5% is an accurate estimate for the weight of Fe^{+++} necessary for complete precipitation of solids, then a very large quantity, (approximately 20 tons) of ferric chloride would be required for precipitation of a 35,000 gallon batch of sludge. Further investigation may well prove this figure to be unnecessarily high; alternatively, the problem might be overcome by labelling a smaller sample of sludge, precipitating the solids, and mixing them with the main batch. The dilution factor, too, will of course be much larger in the actual test, and adsorption of iodine by marine life, etc. will need investigation although previous experience has indicated that this is not appreciable. On the other hand, desorbed iodine should follow the movement of the sludge fairly closely, at least in the very early stages of the experiment,(i.e. the first hour or two). The greatest difficulty may well prove to be adequate mixing of the sludge with the iodine and ferric chloride, especially in the event of a barge being used for the test.

The concentration of iodine 131 in laboratory tests was 50 μc/gallon, corresponding to approximately 2 curies in 35,000 gallons. The extremely low background in water, and the sensitivity of the scintillation counters used, should enable this figure to be cut to at least 1 curie if necessary to satisfy safety and economic requirements. However, the highest possible initial activity is desirable to achieve the maximum sensitivity of detection ; thus the addition of 4 curies would enable detection of the iodine 131 down to a dilution factor of 50,000.

REFERENCE

1. Sugawara, Koyama and Terada "Co-precipitation of iodide ions by some metallic oxides with special reference to iodide accumulation in bottom water layers and interstitial water of muds in some Japanese Lakes". Geneva Papers.

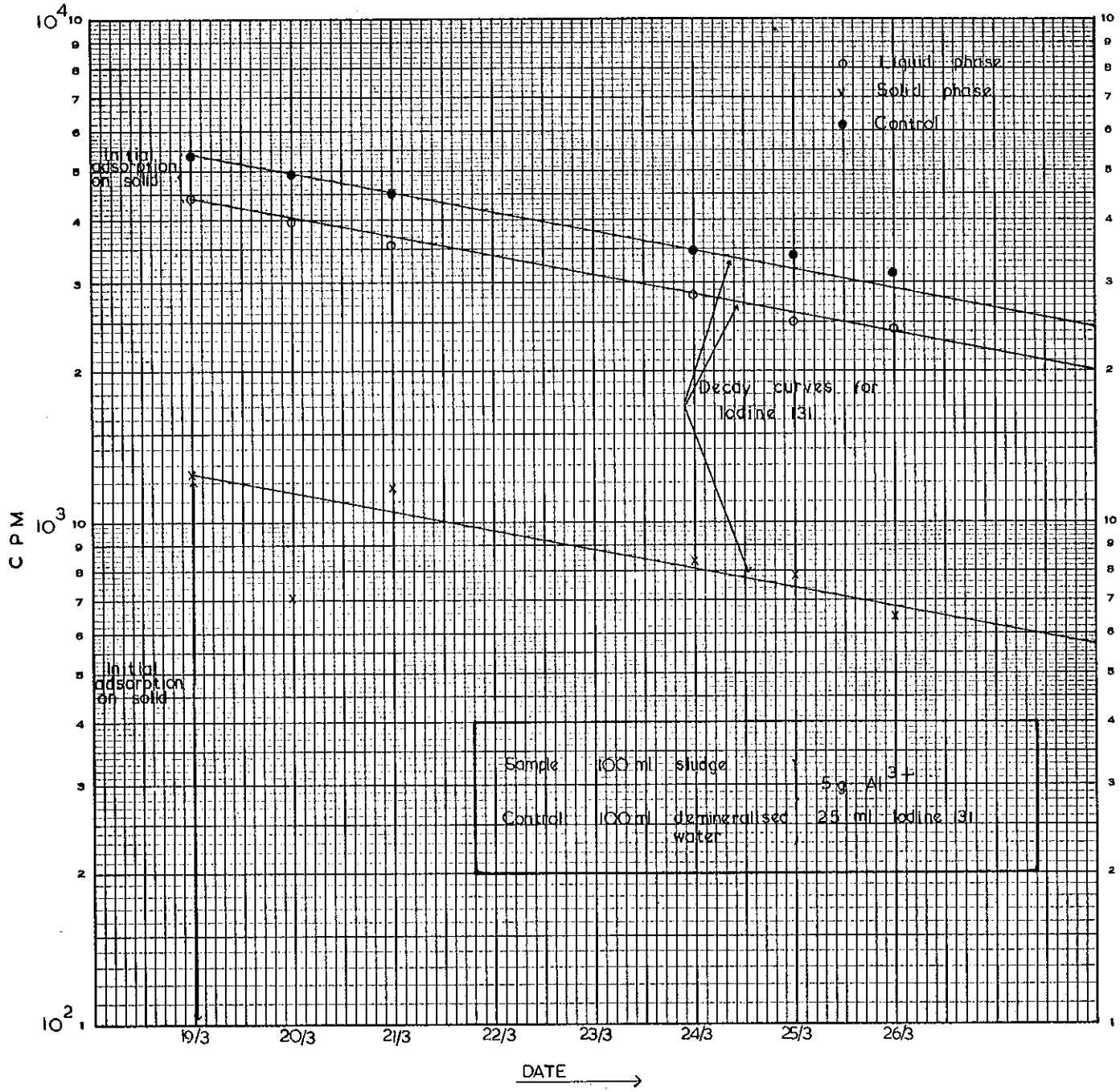

Fig. 1. Adsorption of iodine 131 on sludge

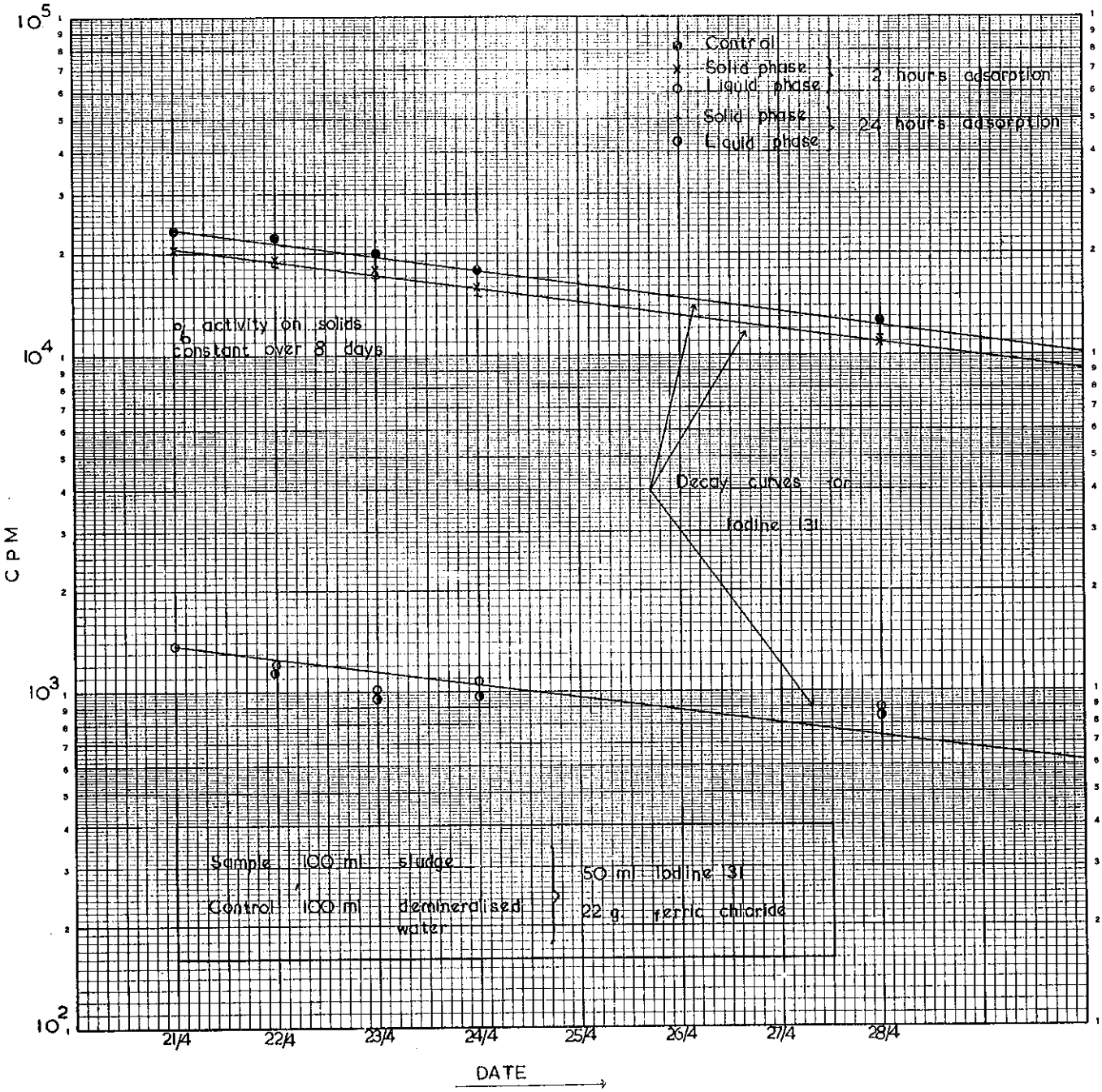

Fig. 2. Adsorption of Iodine 131 on sludge

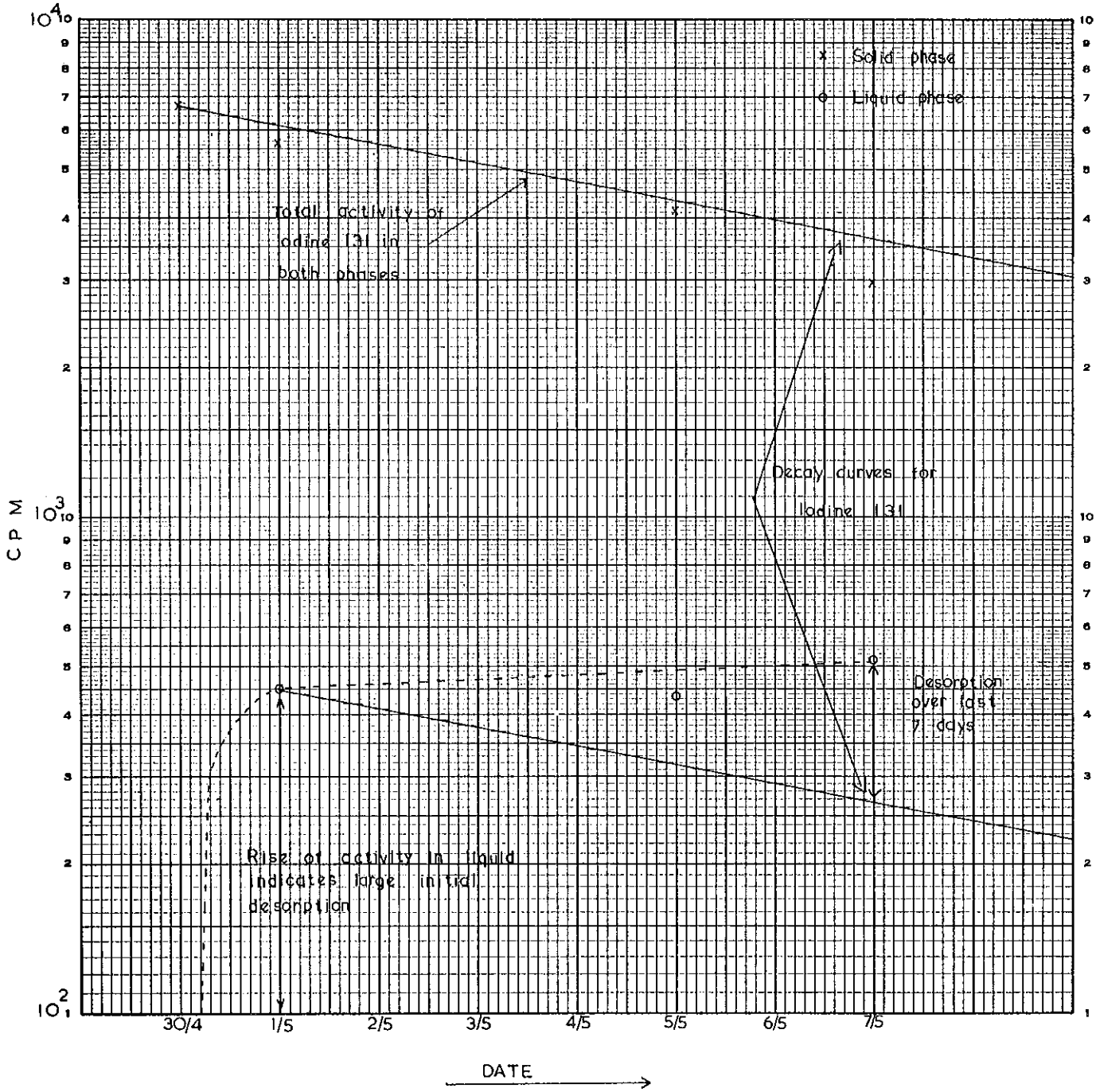

Fig. 3. Desorption of Iodine 131 from sludge