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# MICROBIAL DEGRADATION OF PARAQUAT RESIDUE IN SOIL

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*Keywords:* Degradation, Paraquat residue, Serdang series, Renggam series.

**RINGKASAN**

Suatu kajian berhubung dengan penguraian paraquat di dalam dua jenis tanah Siri Serdang dan Siri Renggam telah dijalankan. Hasil kajian menunjukkan molekul paraquat diserap kuat oleh tanah Siri Renggam. Bagaimanapun proses penguraian lebih cepat berlaku dalam tanah Siri Serdang dibandingkan dengan Siri Renggam, di mana ini ditentukan dengan meneliti pembebasan $^{14}CO_2$ semasa pengeraman. Sisa $^{14}C$ yang tinggi menunjukkan proses penguraian yang perlahan di dalam tanah Siri Renggam.

## INTRODUCTION

Information concerning the microbial decomposition of pesticide residues in soil is very important. These compounds should be decomposed by biological or non-biological agents, and they should not accumulate and cause problems to crops. The bipyridylium compound, 1, 1' – dimethyl – 4, 4' – bipyridylium (paraquat), a highly water soluble organic cation, is tightly and strongly adsorbed to particles with cation-exchange properties. Because of its exchange with clay minerals, it suffers virtually immediate inactivation when in contact with most soils. The adsorption of herbicidal compounds by clay mineral is dependent upon the type of clay mineral involved. The adsorption of the bipyridylium herbicides by several clay minerals may reduce their availability to decomposing microbes. However, several workers have shown that the bipyridylium herbicides can be microbiologically degraded (FUNDERBURK and BOZARTH., 1967; BALDWIN *et al.*, 1968; TU and BOLLEN, 1968).

The most promising technique available at present for the routine assessment of herbicide degradation in soil is the measurement of the release of $^{14}CO_2$ from $^{14}C$–labelled substance. This technique allows continous measurement to be made of decomposition of the herbicide without periodically removing it from the soil. This method have been successfully used by several workers (WEBER and COBLE, 1968; BYAST and HANCE, 1975).

The work described in this paper demonstrates the process of adsorption and the breakdown of paraquat in two types of soil: Serdang Series and Renggam Series. Paraquat is widely used in this country as a herbicide under the commercial name of Gramoxone.

## MATERIALS AND METHODS

### Soils

Two different soil types, which are important agriculturally, were selected for study. Renggam Series and Serdang Series samples were taken 2 cm below the surface. All soil samples were ground to pass through a 3 $mm^3$ mesh sieve. The characteristics of the soils are shown in *Table 1*.

### Chemicals

Methyl $^{14}C$–paraquat chloride was obtained from the Radio Chemical Centre, Amersham. The commercial formulation of paraquat, 'Gramoxone' (I.C.I. Ltd.) was used in this experiment.

### Adsorption to soil

Five ml $^{14}C$–labelled paraquat (0.34 $\mu$Ci/ml) were transferred into test tubes containing either 0.1 or 1.0 g soil samples. The samples were shaken for 45 min. One ml supernatant was transferred into a scintillation vial containing 9 ml of scintillator (BRAY, 1960). The activity in the supernatant was counted by using a liquid scintillation Mark II Counter.

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TABLE 1: THE CHARACTERISTICS OF THE SOILS

| | Clay (%) | Water content (%) | Organic carbon (%) | pH |
|---|---|---|---|---|
| Renggam Series | 7.62 | 58.32 | 7.31 | 4.5 |
| Serdang Series | 15.55 | 72.34 | 4.65 | 5.8 |

**Incubation**

The flasks were connected to the gas-flow system as illustrated in *Figure 1*. Each soil sample (120 g) was transferred to a 500 ml incubation flask. Three different concentrations of paraquat containing 0.32 $\mu$Ci $^{14}$C-labelled paraquat were used in this experiment. One ml methyl $^{14}$C-paraquat was pipetted into the soil and mixed thoroughly. The air-flow rate to each flask was controlled with the screw clip to give a rate of about one bubble/sec. through the 4 mm (internal diameter) tube entering the adsorption solution. The adsorption solution, methanol: ethanolamine (2:1) was changed every five days. The flasks were incubated at 28°C. Incubation period for Renggam Series was 40 days and for Serdang Series 55 days.

Ten ml of adsorption solution was added to 10 ml liquid scintillation containing 4 g PPO, 0.1 g POPOP in 1,000 ml toluene (BAGGIOLINI and BICKEL, 1966). Samples were counted on a liquid scintillation Mark II Counter using an external standard.

**Estimation of residual $^{14}$C**

At the end of the incubation periods, 2 g of soil sample were air-dried, reweighed and transferred to a 50-ml flask. The soil was oxidized by a method similar to that of ALLISON (1960), modified in that the evolved $^{14}CO_2$ was trapped in 30 ml of methanol: ethanolamine solution. The adsorption solution was transferred into three different vials containing scintillation liquid. Aliquots (10 ml) of the adsorption solution were counted as before. All data used were in triplicate.

**RESULTS AND DISCUSSION**

Renggam Series soil has a higher capacity to adsorb paraquat as shown in *Table 2*.

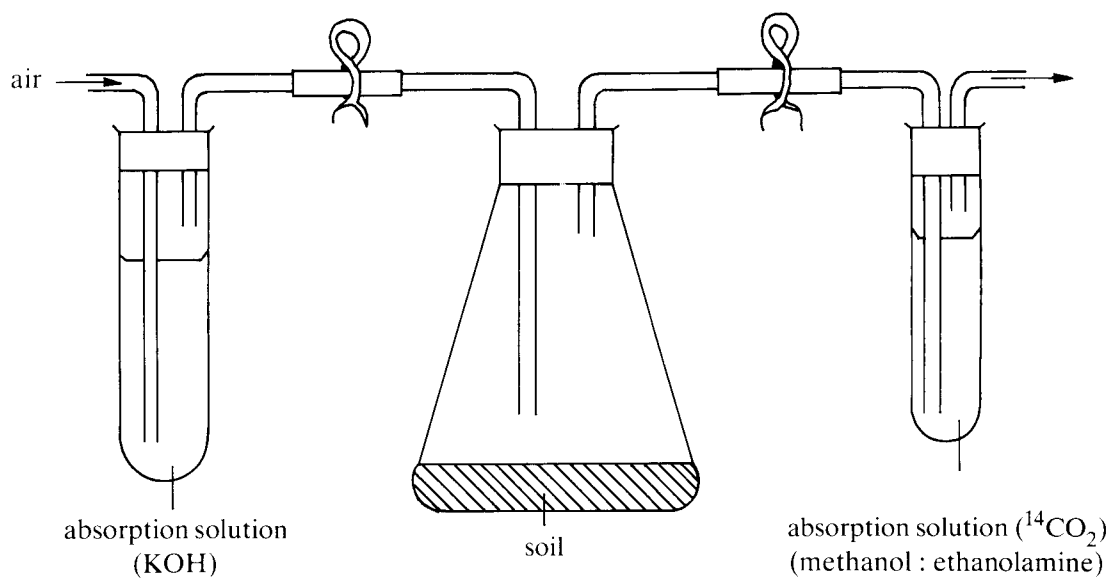


*Figure 1. Gas-flow incubation apparatus*

TABLE 2: ADSORPTION OF PARAQUAT BY SOIL

| Soil (g) | Percentage of $^{14}$C-paraquat adsorbed ± S.E. | | | | | |
|---|---|---|---|---|---|---|
| | Renggam Series | | | Serdang Series | | |
| | 100 ppm | 140 ppm | 200 ppm | 100 ppm | 140 ppm | 200 ppm |
| 0.1 | 67.45 ± 3.64 | 64.44 ± 4.92 | 27.01 ± 4.12 | 58.92 ± 6.09 | 52.06 ± 4.84 | 11.06 ± 2.19 |
| 1.0 | 97.65 ± 6.14 | 93.52 ± 4.26 | 91.82 ± 4.71 | 92.73 ± 5.25 | 87.51 ± 4.32 | 80.84 ± 6.08 |

This is due to low clay content and high organic component. Organic material in soil can react with paraquat through a cation-exchange reaction (BOON, 1964). Adsorption of paraquat by soil depends upon the organic content of the soil, where higher capacity of adsorption occurs when the soil contains more organic matter (KNIGHT and TOMLINSON, 1967). The adsorption of dipyridylium by soils results in its unavailability to weeds and it becomes deactivated (WEBER and SCOTT, 1966).

Since paraquat is strongly held by Renggam Series soil, only small amounts are available to soil microbes for degradation. This results in lower $^{14}CO_2$ evolved from Renggam Series during the incubation as shown in *Figures 2* and *3*. Incubation period for Renggam Series was 40 days and for

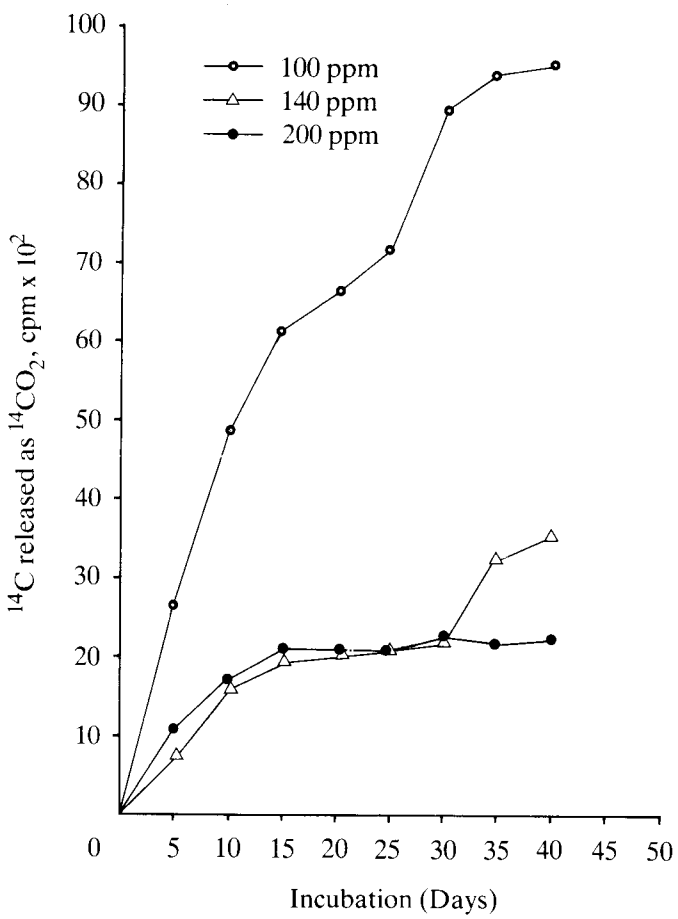


*Figure 2. Cumulative evolution of $^{14}CO_2$ from Renggam soil series.*

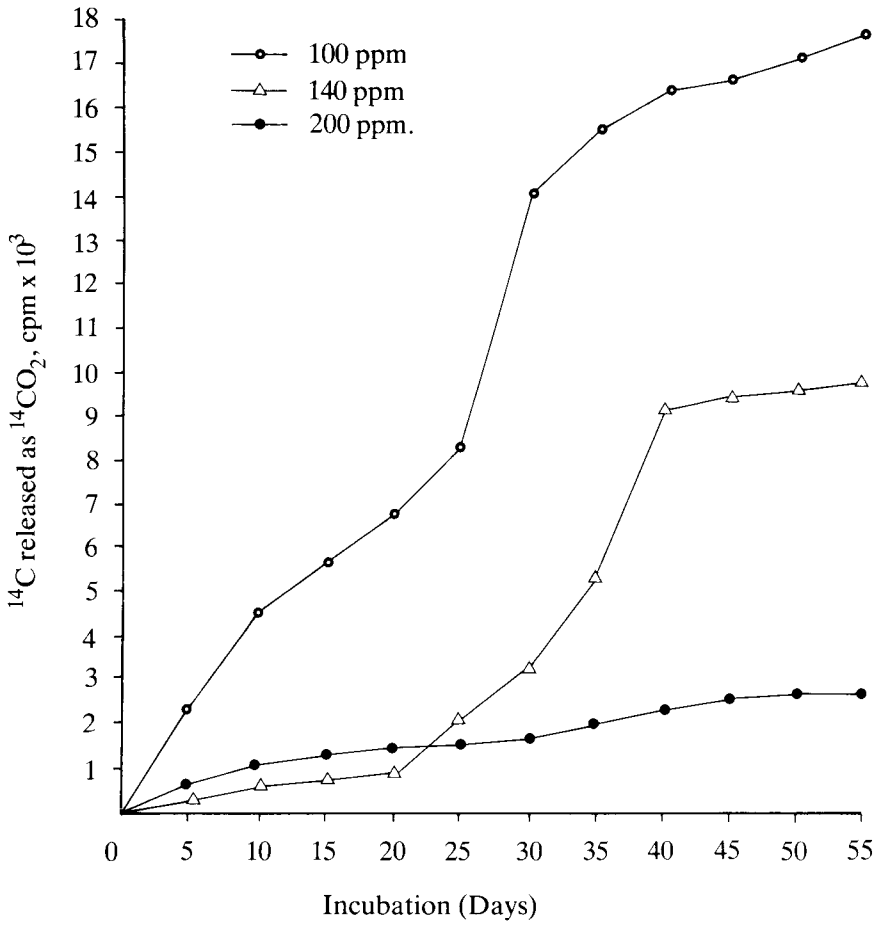


*Figure 3. Cumulative evolution of $^{14}CO_2$ from Serdang soil series.*

Serdang Series 55 days. At the end of the incubation period no more $^{14}CO_2$ could be detected.

At higher paraquat concentration (140 ppm and 200 ppm) the amount of $^{14}CO_2$ evolved was less than at 100 ppm concentration *(Figures 2* and *3)*. The reduction was as much as 80% in Renggam Series and 90% in Serdang Series. At higher concentration, paraquat may inhibit activities of certain microbes and several species of soil fungi (TU and BOLLEN, 1968; WILKINSON, and LUCAS, 1969).

Due to the high affinity of Renggam soil for paraquat and slower degradation process, higher amounts of residues were found in the soil *(Table 3)*.

TABLE 3: $^{14}$C-PARAQUAT RESIDUES IN SOIL AFTER INCUBATION PERIOD

| Paraquat used (ppm) | Residues (%) ± S.E. | |
|---|---|---|
| | Renggam Series | Serdang Series |
| 100 | 21.02 ± 1.12 | 0.25 ± 0.01 |
| 140 | 29.31 ± 1.93 | 0.14 ± 0.01 |
| 200 | 32.43 ± 3.09 | 0.26 ± 0.01 |

About 20% of the initial paraquat concentration remained in the soil after 40 days incubation in Renggam Series and only 0.2% left in Serdang Series after 55 days. These results showed that paraquat persist longer in Renggam Series soil.

## CONCLUSION

It was shown that paraquat was strongly held by Renggam Series soil due to its high organic content. This resulted in a slower degradation process, with about 20% of the initial concentration remaining in the soil as residue after 40 days of incubation.


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## SUMMARY

The degradation of paraquat in two types of soil, Serdang and Renggam Series was discussed. The results showed that paraquat molecules were strongly adsorbed onto Renggam Series soil. However, the degradation process was more rapid in Serdang than Renggam Series Soil, as determined by analysis of $^{14}CO_2$ evolved during incubation. Higher $^{14}C$ residues were detected in Renggam Series which confirmed it to have a slower degradation process.


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