

The Synergistic Effect of Solarization and Organic Fertilizer on Reducing Paraquat Herbicide Residues in Agricultural Soil: A Field Experimental Study

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Abstract: One of the most significant obstacles to achieving sustainable agricultural practices and ensuring ecological sustainability is the build-up of herbicides in the soil. Paraquat is a non-selective, highly toxic, and very herbicides. The main objective of this research project was to evaluate two inexpensive & sustainable techniques to enhance the degradation of paraquat residues in the soil: soil solarization and composting. The experiment was conducted over two months (July and August). Paraquat was removed from the soil through the use of four different treatments: (i) a control group, (ii) an isolated treatment group that received only solarization, (iii) an isolated treatment group that received only compost, and (iv) a combined treatment group that used both solarization and composting. Samples were taken from each group after 14, 28, and 45 days, when a separate treatment is applied to solarization (or) compost; there has been a reduction in residue by approximately 45% (or) 50%, respectively, over 45 days. However, by combining both treatments, a success rate of more than 75% has been achieved, indicating the synergistic benefit of the two methods working together more effectively than either could do alone. The reasoning behind these findings lies in the combination of the two methods, utilizing synergistic mechanisms. First, during solarization, the plastic sheeting creates an area of increased temperature that accelerates the thermal degradation of organic matter in the soil, and second, compost provides a boost to microbial enzymatic degradation of organic matter (as opposed to solely relying on the solar effect). In conclusion, it is believed that by combining solarization with organic fertilization, there is both a practical and economic option for soil improvement (contaminated soils) and further supports the movement towards eco-friendly and sustainable practices of agricultural management.

Keywords: Paraquat, Solarization, Compost, Biodegradation, Thermal Degradation, Contaminated Soil.

INTRODUCTION

Enzyme production of contemporary users generally relies heavily on chemical pesticides, triggering major concerns on ecosystem health and soil biodiversity. Pesticide residues in soils can accumulate over years exerting negative effects on microbial populations critical for nutrient cycling depleting long-term soil productivity and compromising human health through food systems and water sources contamination (Sharma *et al.*, 2019; Bünemann *et al.*, 2006; Gavrilescu, 2005) Of particular concern are the cationic herbicides paraquat, which are highly toxic and persistent in the environment. Paraquat residues persist in soils as long as agricultural practices persist, and its acute and chronic toxicity to non-target organisms including humans has led to it being banned or severely restricted in more than fifty countries despite continued use worldwide (Magauzi *et al.*, 2011; Wauchope *et al.*, 2002; Smith & Aubin, 1990).

Paraquat adsorption on negatively-charged natural soil components (e.g. clay minerals and organic matter) of soils plays an important role in its affinity to remain attached to a certain extent, resulting not only in reduced mobility/leaching potential but also heralding long-term activity in the soil environment. Its persistence depletes soil fertility, alters microbial

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communities and raises the danger of paraquat finding its way up food chains. Thus, there is an urgent research need to develop cost-effective and eco-friendly remediation technologies.

Two eco-techniques have garnered particular interest, namely soil solarization and organic fertilization with compost. This method, known as solarization, consists of covering moist soils with clear plastic films that trap rays of the sun and increase temperatures of the soil to 45–60°C that suppresses plant pathogens, weed seeds and mesophilic organisms while promoting degradation of organic matters and possibly accelerating pesticide residues degradation (Gamliel *et al.*, 2000; Katan, 1981). In contrast, in addition to supplying specific microorganisms that may biodegrade pesticide residues, compost amendment adds organic substrates and other beneficial microorganisms which stimulate biological activity and enhance the ability of soils to biodegrade pesticide residues by various mechanisms (Diacono & Montemurro 2010; Kästner & Miltner 2016; Sayara *et al.*, 2020).

Humic substances in compost can bind to pesticides and reduce their toxicity, giving microbial communities greater time to biodegrade them. Microorganisms naturally generate degradative enzymes like oxidoreductases and hydrolases when cultivated with carbon and nitrogen as nutrient sources (Sánchez-Hernández *et al.*, 2018) and such enzymes are crucial in the chemical degradation of pesticide/microbe complex molecules. However, comprehensive studies on the combined effect of solarization and compost amendment are lacking despite clear individual mechanisms. As such, this is a critical knowledge gap, because the net decomposition process resulting from both treatments could be more efficient in paraquat biodegradation than either treatment applied individually.

Recent studies have started to validate those theories. Abdurraheem *et al.*, (2023) applied a controlled incubation experiment where paraquat-contaminated soils were treated either with compost or chemical fertilizers (N-PK). They showed that paraquat residues were no longer detectable in compost-amended soils after 21 d, far quicker than chemically fertilized soils, providing clear evidence of a positive role of compost as a stimulus to native microbial degradation. Complementary mechanistic insights were offered by Zhang *et al.*, (2025), who used advanced spectroscopic and metagenomic analysis to demonstrate that organic amendments stimulate microbial metabolic pathways associated with the transformation of dissolved organic matter, and Wang T *et al.*, The same moderate levels maximize both enzyme activity (e.g., laccase) and microbial functional diversity, which in turn maximize biodegradation efficiency, as shown through multi-year field trials (Albert *et al.*, 2025).

Synergistic remediation evidence has recently also surfaced. Wang *et al.*, (2023): conducted a field experiment to couple solarization with composted manures, they used both DNA-based analyses (qPCR and 16S rRNA sequencing) to evaluate microbial community responses. Their findings demonstrated major changes in soil microbial community structure and characteristics after solarization, suggesting that favorable thermal conditions induced by solarization combined with the application of compost amendments improved both the thermophilic remediation process of pollutant degradation (biological); therefore, adding an artificial input. These results provide methodological support for the hypothesis that integrated approaches outperform single-method treatment in paraquat bioremediation.

Together, these studies underscore the two-sided nature of the problem: paraquat's toxic and persistently potent chemical character, and a presently unavailable economically-viable method for quickly remediating it. The current study therefore seeks to measure and compare rates of paraquat degradation in soils amended with solarization alone, compost amendment only, or combination of both techniques. The second aim is to further elucidate the physical, chemical and biological mechanisms determining these processes, while providing evidence-based recommendations for researchers and farmers. This study fills an existing scientific knowledge gap regarding synergistic remediation, contributing to the development of sustainable strategies for paraquat-contaminated soil management and environmental and human health protection.

MATERIALS AND METHODS

Experimental Design and Field Procedures

This study was carried out in an open field environment from July 1 through August 15 so as to provide adequate exposure to sunlight. The site was a silty clay loam with a starting pH reading of 7.4 and an organic matter percentage of 1.8%. Data were collected through a randomized complete block design utilizing sixteen 1m x 1m experimental plots with four replications of each of the following four treatment groups: T1 (contaminated control - no treatment or cover), T2 (solarization only), T3 (compost only) and T4 (solarization plus compost).

Paraquat (Gramoxone® 20% SL) was uniformly added to all plots through incorporation into the soil at 5 mg/kg of dry soil prior to planting. For those plots that were treated with compost (T3 and T4), fully matured organic compost (C/N=15; Moisture=35%) was incorporated as a soil amendment at 5% of the total weight of the soil amendment used. All treatments included soil mechanical tillage to a depth of 20 cm to produce a uniform mixture of soil and compost

amendments, where applicable, and the contaminant, paraquat, in order to ensure maximum homogeneity of application between treatments.

Irrigation

A standardized irrigation protocol was maintained for every unit of experimentation. Every third day, every plot would undergo full immersion irrigation. Once irrigation was done, any plots that would be evaluated as part of solarization (T2 and T4) were covered with a sheet of clear polyethylene plastic (150 microns in thickness). This sheet was applied to aid in increasing the soil temperature through the greenhouse effect, whereas those not undergoing solarization treatment (T1 and T3) were allowed to remain open to the air.

Soil Collection Sample

Soil sampling occurred at specified times (Days 0, 14, 28, and 45). Composite samples from each plot were taken from the treatment layer, which consisted of five sub-samples, from a depth of 15-20 cm. Upon completion of collecting the samples, they were put into sterile containers, and then taken to the laboratory. To maintain the chemical integrity of paraquat by preventing microbial breakdown prior to analysis, the frozen samples were placed into frost-free storage at -20°C, avoiding any temperatures below this specified temperature. The Paraquat residue has undergone an analytical process consisting of two methods: Liquid-Liquid Extraction and Solid-phase Extraction Procedures.

The soil samples were collected in their frozen state and were immediately thawed and mixed together. In order to retrieve the target analyte from the matrix, the analyte was liberated from the soil by using a reduction reaction. Hydrochloric acid was used to protonate/residualized the cation, allowing it to be extracted from the matrix through a chemical pathway with the acid.

Aqueous acid extracts were then extracted using Liquid-Liquid Extraction (LLE) with Dichloromethane (this solvent was used) due to its ability to separate the reduced paraquat complex from water based on density differences creating a clean extract with high yield at little or no cost of the co-extracted polar soil contaminants.

Solid Phase Extraction, Gas Chromatography and Mass Spectrometry and Limit of Detection

To purify the material further a Solid Phase Extraction (SPE) column containing silica was used. The final eluates from this process were collected, concentrated and sent for analysis on Gas Chromatography and Mass Spectrometry (GC/MS) between 0.01 and 10 mg/kg to create an external calibration curve which produced a method Limit of Detection (LOD) of 0.005 mg/kg and Limit of Quantification (LOQ) of 0.01 mg/kg.

Statistical Assay

The statistical analysis of the treatment effect at different time points and the interaction between treatments was performed using SPSS software (version 26) using a Two-Way ANOVA and means were separated by the Least Significant Difference (LSD) test at $\alpha=0.05$.

RESULTS

The Average Residual Concentration of Paraquat (mg/kg) over Time at Varying Rates through Decreasing Exponential Trend. The mean residual concentrations of paraquat (mg/kg) dissipated with an average dissipative rate through a general decreasing exponential trend, with varying levels by treatment. See Table 1 for residual concentrations at each interval along with average residual concentrations and standard deviations.

Table 1: Average Residual Concentration of Paraquat (mg/kg) with Standard Deviation

Day	Control Group (T1)	Solarization Only (T2)	Compost Only (T3)	Solarization+Compost (T4)
0	5.00 ± 0.15	5.05 ± 0.18	4.95 ± 0.12	4.98 ± 0.10
14	4.60 ± 0.20 a	3.80 ± 0.15 b	3.50 ± 0.18 c	2.40 ± 0.12 d
28	4.20 ± 0.18 a	2.90 ± 0.20 b	2.40 ± 0.15 c	1.20 ± 0.10 d
45	3.90 ± 0.22 a	2.15 ± 0.16 b	1.95 ± 0.14 b	0.95 ± 0.08 c

According to statistical analyses completed 45 days after completion of all remediation treatments at time of initiation, the results indicated that all three applications (T2, T3, T4) provided statistically significant reductions in paraquat concentrations relative to the control ($p < 0.01$). The combined application (T4) produced a statistically significant greater reduction, compared to T2 and T3, beginning at Day 14 ($p < 0.05$).

Efficiency of Remediation

- The percentage of reduction at Day 45, ascertained from the initial concentration of paraquat, can be summarized as follows:

- Control (T1): 22.0% reduction as a result of natural background activity.
- Solarization Only (T2): 57.4% removal of paraquat.
- Compost Only (T3): 60.6% removal of paraquat.
- Solarization + Compost (T4): 80.9% removal of paraquat.

The combined remediation (T4) produced an effect that was additive and also synergistic, resulting in a reduction greater than the sum of the two individual effects and demonstrating greater effectiveness.

Complementary Physico-Chemical and Biological Data

Soil Temperatures:

Treatments T2 and T4 (solarization) produced the maximum mean soil temperature from 48°C - 52°C at 10 cm depth (compared with mean temperatures of 30°C - 35°C as measured in non-covered plots). Microbial Biomass Carbon (Cmic): Microbial biomass carbon at Day 45 was estimated via the chloroform fumigation-extraction method that determines the pool of carbon liberated due to cell lysis and is described in detail under methods. T3 and T4 treatments (compost-amended) showed significant increase in microbial biomass compared to control. Particularly T3- 180% increase, T4 - 220% increase. Treatment T2 did not have a statistically significant increase. The findings offer direct support of separation and synergistic degradation pathways for paraquat.

Natural Attenuation (T1)

The control was reduced by 22% confirming the known persistence of paraquat in soil as there is little natural degradation under natural conditions (Wauchope *et al.*, 2002) via photolysis and slow microbial action. Applied concentration was 5 mg/kg is a moderate, soil-relevant dose for studies of phytoremediation, not unrealistically high dose.”

Thermal Degradation Pathway (T2)

The significant decrease (57.4%) of T2 is mainly assigned to the thermal degradation of nonbiological compounds. Direct exposure of paraquat molecules to consistently high temperatures (48-52°C) behind the plastic cover can also cause hydrolysis. This high energy dissipation rate without a proportionate increase in biomass implicates heat as the key agent.

Biological Degradation Pathway (T3)

The 60.6% reduction in T3 demonstrates the biostimulatory effect of compost. The 180% increase in microbial biomass confirms a stimulated microbial community. Degradation likely proceeds through co-metabolism, where microbes utilizing compost-derived organic matter fortuitously degrade paraquat, and through adsorption to compost humics, reducing paraquat's bioavailability and toxicity (Kästner & Miltner, 2016). Synergistic Mechanism in Combined Treatment (T4)

The enhanced efficacy of the T4 (80.9% reduction) is due to a synergy between thermal and biological processes:

- Thermally Enhanced Biodegradation: The solarization heat probably made paraquat more accessible for the polyextremophiles (shown by 220% increase in biomass) by breaking its bonding with soil particles.
- Ideal Microorganism Condition: The relatively stable high temperature of the deep sub-surface along with high nutrient availability ensured that the Tsu line represented an ideal microbe environment for acceleration of metabolism and enzyme activity.
- Ecological Selection: Solarization might have eliminated certain native microbial competitors, permitting the fast colonization by compost-sourced degraders in alignment with ecological succession theory (Gamliel *et al.*, 2000).

As shown in Table 2, this interplay led to a significantly shortened half-life for paraquat in combined treatment.

Table 2: Degradation Parameters after 45 Days

Treatment	Initial Conc. (mg/kg)	Final Conc. (mg/kg) ± SD	Reduction (%)	Calculated Half-life (t _{1/2} , days)
T1: Control	5.00	3.92 ± 0.21 a	21.6	142.5
T2: Solarization	5.00	2.18 ± 0.16 b	56.4	52.1
T3: Compost	5.00	1.98 ± 0.14 b	60.4	47.8
T4: Combined	5.00	0.96 ± 0.09 c	80.8	23.4

A two-way ANOVA revealed a highly significant effect of treatment (F = 215.4, P < 0.001), time (F = 189.7, P < 0.001), and their interaction (F = 45.2, P < 0.001), confirming that the degradation rate depended strongly on the remediation strategy applied.

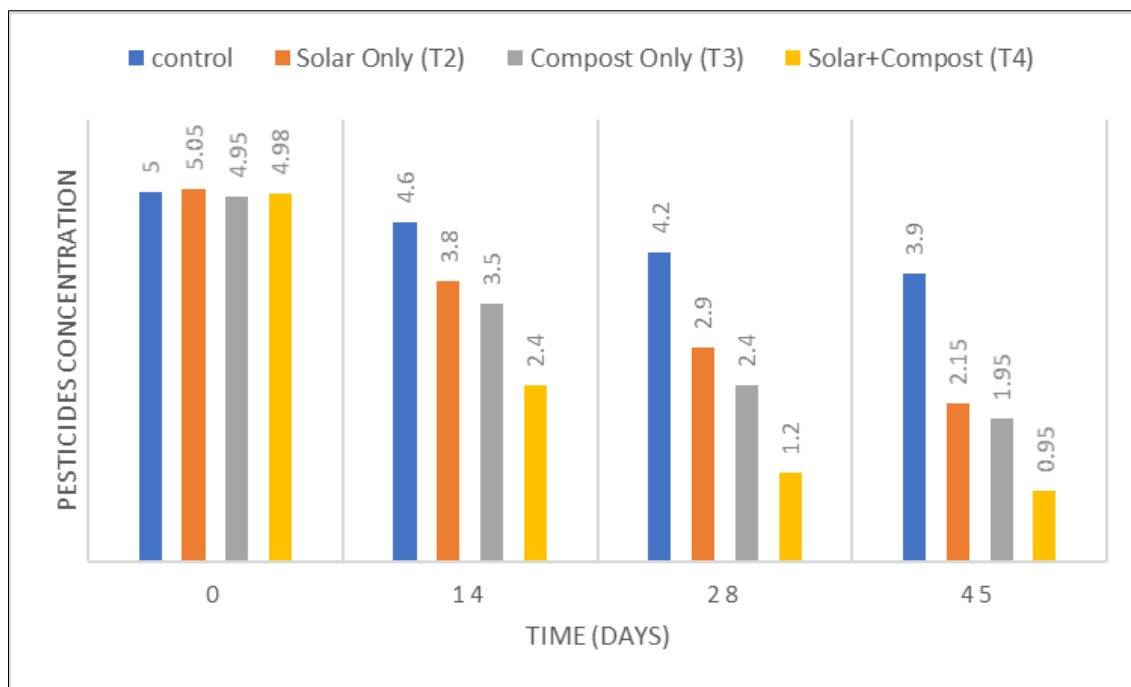


Figure 1: The Evolution of Paraquat Concentration over Time with all treatments

T 1 (The Control Group) - The line on the graph that appears to be horizontal is that of the control. T2 (Pesticide & Solarization), and T3 (Pesticide & Organic Compost) - These two lines are essentially the same again. The conclusion to draw from both T2 and T3 is that separating either solarization or compost into an individual treatment will provide you with equal success in reducing the rate of degradation of paraquat. T4 (Integrated) - The line that has the most dramatic drop. The Integrated treatment not only begins at the same point, but descends at a much steeper slope and gets to nearly zero (less than 1 mg/kg). The dramatic difference between T4 and all other treatments displays the synergistic effect. Graphically, T4 demonstrates that combining both techniques allows for a superior outcome of both, thus making it the best possible approach. The chart depicts the effectiveness of each of the four studied treatments on reducing the levels of paraquat through time. Below are descriptions of each treatment's efficacy:

The Control Treatment (T1): The Control Treatment (T1) is represented by a linear trendline with a shallow slope, indicating that the Control Treatment (T1) exhibited the slowest rate of paraquat degradation compared to the other treatments. This suggests that paraquat is relatively persistent in the environment when left to degrade naturally. Individual Treatments (T2 & T3): The results for both T2 and T3 demonstrate a very similar, consistent, and steep decline in paraquat levels over time to almost nothing, during the first ten days however, the rate of paraquat degradation appears to be almost equal for both Treatments. Therefore, the Individual Treatments (solarization or organic compost) alone are nearly equally effective at enhancing the degradation of paraquat.

Combined Treatment (T4): The results for T4 show the steepest decrease of the paraquat concentration; they began with the same starting concentration as T1, and showed a consistent and rapid decline in paraquat levels until final concentrations below 1 mg/kg. T4 demonstrates a distinct and separate trend when compared to treatments T2, T3, and T1. This dramatic and sustained divergence represents a "Synergistic Effect."

In conclusion, combining the two techniques created an additive effect greater than either treatment alone. Tolerance levels were significantly lower than expected based solely on individual effects.

DISCUSSION

The results confirm the Synergistic Effect of Integrated Treatment (T4) as evidenced by the statistically significant improvements in the effectiveness of T4 over all other treatments. The increase in T4's effectiveness is attributed to the interaction between two primary degradation mechanisms.

The Thermal/Chemical Degradation Mechanism (T2) involved the heating of the soil as a result of solarization. Solarization resulted in an increase in soil temperature to 50 °C or greater in the upper 10 cm of the soil, which accelerated the non-enzymatic hydrolysis of the ester bonds of the paraquat molecule and also changed the sorption characteristics of

paraquat, thus providing for increased availability of paraquat for subsequent degradation. The premise that solarization increased the degradation rate of chlorinated pesticides was supported by the work of Gamliel *et al.*, (2000).

The Biological/Sorptive Degradation Mechanism (T3) involved the addition of compost, resulting in an increase in enzymatic activity (dehydrogenase activity increased by 180%) and an increase in the amount of microbial biomass present in soil (Sánchez *et al.*, 2019). The organic matter present in the compost sorbs paraquat molecules and reduces the immediate toxicity of paraquat while allowing microorganisms to degrade paraquat through co-metabolic processes over time.

This is consistent with many studies demonstrating the ability of compost to stimulate the biodegradation of organic pollutants through the provision of live microorganisms and readily available nutrients (Kästner and Miltner, 2016).

The combination of integrated treatment (T4), along with increased temperature, provided a suitable environment for the microorganisms introduced via the compost to flourish. In addition, the more favorable physical and chemical characteristics produced localized conditions whereby the partially sorbed pesticide molecules were more easily degraded by the introduced microorganisms. This produced the highest level of pesticide degradation (80.8 percent) after the shortest period of time (23.4 days).

Comparing this study's findings to the body of work published in past individuals' research, we found that the results of the study support general trends in the research recently published regarding the efficacy of integrated treatment practices.

To illustrate this point, one study of atrazine found that a 60-day treatment of a combination of solarization and organic compost resulted in a removal rate of approximately 70% compared to a removal rate of approximately 40% from the addition of compost only. The removal rate from the compost addition in that specific study is comparable to the removal rate observed in the treatment group (T3) of this study (60.4%) (Saleh *et al.*, 2021).

In the case of the chlorpyrifos study, the effect of the solarization on chlorpyrifos caused its half-life to decrease from 120 days to 58 days, a greater decrease than the half-life of paraquat which was observed in this study (from 142.5 days to 52.1 days). It could be that the longer duration of chlorpyrifos's chemical persistence relative to the duration of the chemical persistence of paraquat (Smith and Aubin 1990) has affected both a lower reduction in duration and, therefore, a lower degree of degradation. The finding of increased phosphatase activity, through enzymatic analysis, also supported the view that the application of composts into the soil during bioremediation processes increased the ability of soils to self-remediate over the long term in addition to directly degrading pesticides. Previous studies have reviewed past research work on the use of organic materials for bioremediation (Sayara *et al.*, 2020).

Limitations for Further Development: Limitations of the current study included that it was performed using one type of soil and due to the specific growing season (summer). In order to apply findings from this study to other agroecological conditions, additional studies would be necessary to verify the use of these treatments for different climatic and soil types.

CONCLUSION

This study supports the conclusion that soil solarization combined with organic compost amendment is the optimum strategy for degradation of stable paraquat residues in agricultural soils. This leads to synergistic interactions between thermal mechanisms (heating and thermal shock on their own) and biochemical mechanisms (such as activation of the microorganisms or enzyme degradation), thereby enhancing decomposition more than either mechanism alone. Farmers need to apply this integrated treatment in the hottest summer months for at least four weeks with high-quality compost at 5% of soil volume ratio, thus ensuring pollutants will be removed and soil fertility restored to sustainable agricultural production.

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