

# Advanced Oxidation Processes For Lindane Degradation: A Leap Toward Water Purification Excellence

Noor Zulfiqar<sup>1\*</sup>, Hafiz Muhammad Yameen<sup>2</sup>, Muhammad Asad Ali<sup>3</sup> and Fawad Inam<sup>4,5</sup>

<sup>1</sup>Department of Chemistry, Faculty of Science, University of Agriculture, Faisalabad, Pakistan.  
Email: chemistnoor94@gmail.com

<sup>2</sup>Department of pharmaceuticals, Government College university, Faisalabad.  
Email: yameenhm425@gmail.com

<sup>3</sup>Department of chemistry, Riphah International University Faisalabad.  
Email: aliasadmuhammad0308@gmail.com

<sup>4</sup>School of Architecture, Computing and Engineering, University of East London, Docklands Campus, University Way, London, UK.  
Email: f.inam@uel.ac.uk

<sup>5</sup>Oxford Business College, Macclesfield House, New Road, Oxford, UK.

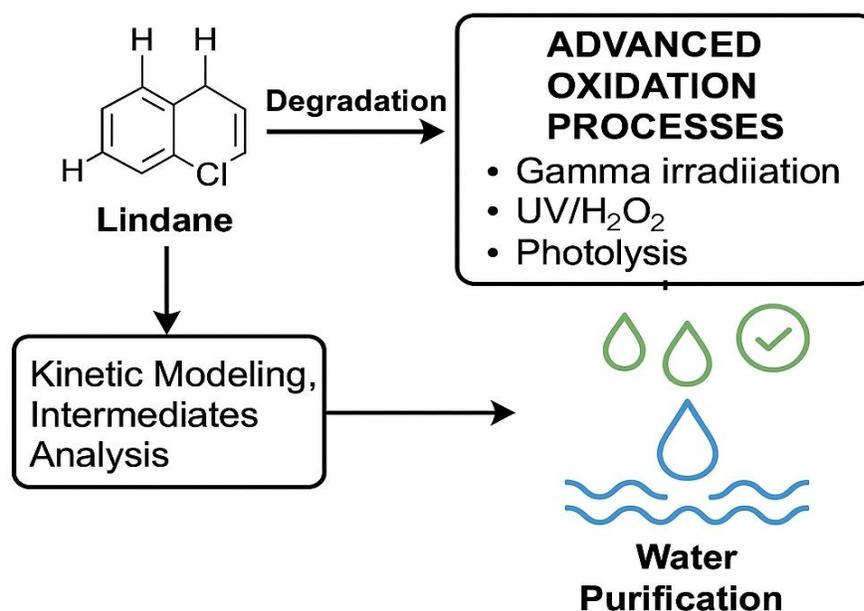
## \*Corresponding Author

Noor Zulfiqar, Department of Chemistry, Faculty of Science, University of Agriculture, Faisalabad, Pakistan.

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## Graphical Abstract



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## Abstract

*Lindane, a persistent organochlorine pesticide and potent environmental pollutant, poses significant risks to ecosystems and human health due to its toxicity, bioaccumulation, and resistance to degradation. Conventional water treatment methods often fail to achieve complete mineralization of such contaminants, necessitating the development of more effective remediation strategies. Advanced Oxidation Processes (AOPs), particularly those involving hydroxyl and other reactive radicals, have emerged as promising technologies for the complete breakdown of hazardous organic compounds like lindane. This study explores the degradation of lindane in aqueous media using various AOPs, including gamma radiation, UV/H<sub>2</sub>O<sub>2</sub>, and photolysis, under controlled laboratory conditions. Comprehensive kinetic modeling, intermediate identification, and analytical evaluations were performed using GC-ECD, HPLC, and IC methods. The degradation efficiency, optimal conditions, and influence of natural radical scavengers were examined in detail. Additionally, real-world water samples from Peshawar Valley and industrial effluents from Hayatabad were analyzed for lindane contamination and subsequently treated using gamma irradiation. The findings underline the potential of radiation-induced AOPs in achieving near-complete mineralization of lindane, offering a robust and scalable solution for environmental detoxification and safe water production.*

**Keywords:** Lindane Degradation, Advanced Oxidation Processes (AOPs), Gamma Irradiation, Water Purification, Persistent Organic Pollutants (POPs), Organochlorine Pesticides, Kinetic Modeling, Environmental Remediation, GC-ECD, UV/H<sub>2</sub>O<sub>2</sub>

## 1. Introduction

Environmental pollution by toxic chemicals is a global problem, particularly organochlorine compounds (OCs), such as Persistent Organic Pollutants (POPs), are of great concern because of their persistent, toxic, bio accumulative and long-range transportable nature. Among different types of pollution, water pollution is more important since water is a key component in determining the quality of our lives. Although water covers more than 70% of the Earth, only 1% of the Earth's water is available as a source of drinking water. Yet, our society continues to contaminate this precious resource and, therefore, protecting the integrity of our water resources is one of the most essential environmental issues of the 21st century.

Hexachlorocyclohexane (HCH, also called benzene hexachloride) is an organochlorine insecticide and is considered as one of POPs. HCH is available in two formulations: technical HCH and lindane. Technical HCH is a mixture of different isomers (which predominantly consists of  $\alpha$ ,  $\beta$ ,  $\gamma$ , and  $\delta$  isomers) while lindane is almost pure  $\gamma$ -isomer (> 99% pure). Annual global production levels for lindane and technical HCH were estimated as high as 6,000,000 and 11,000,000 metric tons respectively in 1980s [1]. Lindane was first produced at the beginning of 1940s and since that time, it has been widely used in both agricultural and pharmaceutical commercial applications throughout the world, mainly in developing countries due to its low cost and high efficiency, and has caused serious environmental problems. Some of the uses of lindane are on crops, in warehouses, in public health measures to control insect-borne diseases and, together with fungicides, as a seed treatment agent. Lindane is also used in a variety of domestic and agricultural applications, such as dips, sprays and dust for livestock and domestic pets. The forestry industry also uses lindane to control pests on cut logs. Other uses of lindane include lotions, creams and shampoos for the control of lice and mites (scabies) in human [2]. China is the world's second largest pesticide manufacturer, and the neighbor countries are likely to be highly contaminated. Several studies have reported the elevated concentrations of organochlorine pesticides (OCPs) in

fishes, mussels and birds collected from Asian countries including India, Vietnam and China [3]. In Pakistan, it was estimated that the consumption of technical HCH has increased from 3 tons per year in 1996 to 20 tons per year in 1999 [4]. High levels of HCH and other OCPs have been detected in human body and in soil samples from different areas of Pakistan also [5].

The use of lindane was prohibited in most countries in 1990 for treatment of cereal crops, but it is still in use in United States of America (USA), Canada and most European countries for wood treatment. In Africa and Asia, about 3000 tons of lindane was used in 1998 [6]. Because of the generally slow rate of transformation in the environment, lindane pollution has been found in almost all types of environment, with most of the pollution found in water besides contamination of soil, sediments and air. The most contaminated areas are locations where lindane is formulated, stored, used or disposed of. Lindane residues have been found in water samples from different resources, in human and wildlife tissues as well as in food products, including fruits, vegetables, meat, milk products, fish and other seafood [7]. The estimated degradation half-lives of lindane in rivers and lakes range from 3 to 300 days depending on alkalinity, pH, temperature, Dissolved Oxygen (DO), Total Organic Carbon (TOC) and Biological Oxygen Demand (BOD) etc., while in soil and sediment, it is 120 and 90 days, respectively.

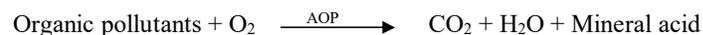
Lindane is considered to be highly toxic to aquatic organisms, and moderately toxic to birds and mammals. Lindane and certain HCH isomers have been found to cause endocrine disrupting effects as well as reproductive and central nervous system damage. Both United States Environmental Protection Agency (US EPA) and International Agency for Research on Cancer (IARC) have classified lindane as a possible human carcinogen [8]. Maximum Residue Limits (MRL) recommended by World Health Organization (WHO) for lindane vary from 0.05 to 3 ppm for various fruits and vegetables, 0.01 ppm for milk and 0.003 ppm for water [9]. US EPA has set a Maximum Content Level (MCL) of 0.0002 ppm for drinking water [8].

Up to now, significant research has been carried out to remove lindane and other POPs from water, and many conventional methods already existed, such as air stripping, activated carbon filtration, membrane technologies and biodegradation etc., however, none of the methods give satisfactory and efficient results [10].

Some innovative and ultimate treatment technologies have shown encouraging results for treatment of several contaminants in environmental samples [11,12]. As a group, these technologies are called Advanced Oxidation Technologies (AOTs) or Advanced Oxidation Processes (AOPs). AOPs lead to degrade pollutants to simpler fragments and often result in complete mineralization. This is an improvement over many conventional options used now a day that are merely phase transfer processes. Similarly, land disposal merely transfers pollutants that will eventually require further attention. However, mineralization represents the final and ultimate treatment, which is the goal in many areas of pollutant control. The AOPs use free radicals (principally hydroxyl radical, •OH) which attach and decompose pollutants. In some cases, oxidation via the •OH is slow and the application of reducing radicals, such as the hydrated or solvated electron (e-aq) or hydrogen atom (H.) is advantageous. These, still newer, technologies are called

advanced oxidation reduction technologies (AORTs). An excellent source of both reducing and oxidizing radicals for water treatment is ionizing radiation, such as radioactive isotopes or fast electrons from electron beam. The simultaneous generation of both reducing and oxidizing species allows for a versatile approach to the ultimate treatment of a wide variety of pollutants [12].

The overall process of AOPs in presence of oxygen can be summarized as:



Several types of AOPs can be used for lindane degradation, such as UV/H<sub>2</sub>O<sub>2</sub> photolysis, photo catalytic degradation, photochemical degradation, microwave decomposition and radiation treatment [13-15].

In the present study, a detailed radiation induced degradation of lindane in water, in presence of several contaminants is planned. Kinetic modeling of the results using computer simulation would also be carried out.

Method	Mechanism	Degradation Efficiency	Advantages	Limitations	End Products
Air Stripping	Transfer of volatile lindane from water to air	Low to moderate	Simple setup, inexpensive	Phase transfer only, secondary air pollution	Lindane in air phase
Activated Carbon Filtration	Adsorption onto carbon surface	Moderate	Effective for organics	Regeneration required, saturation issues	Adsorbed lindane residues
Membrane Filtration	Physical separation	Moderate	Selective removal	Expensive membranes, fouling	Lindane in retentate
Biodegradation	Microbial breakdown	Variable	Eco-friendly	Slow rate, toxic to microbes	Partial breakdown intermediates
UV Photolysis	Photodegradation via UV photons	Moderate	Direct breakdown	Requires high energy UV, low quantum yield	Partially oxidized products
UV/H <sub>2</sub> O <sub>2</sub> AOP	Generation of hydroxyl radicals	High	Efficient mineralization	Sensitive to pH and H <sub>2</sub> O <sub>2</sub> dose	CO <sub>2</sub> , H <sub>2</sub> O, Cl <sup>-</sup>
Photocatalysis (e.g., TiO <sub>2</sub> )	Light-activated oxidation	High	Reusable catalyst, green	Needs catalyst separation	CO <sub>2</sub> , H <sub>2</sub> O, organics
Microwave Decomposition	Thermal excitation	High	Rapid heating, bulk treatment	High power required	CO <sub>2</sub> , organic residues
Gamma Irradiation	Oxidation by radiolysis products (•OH, eaq <sup>-</sup> )	Very High (>95%)	Powerful, deep penetration, scalable	Safety & infrastructure requirements	CO <sub>2</sub> , H <sub>2</sub> O, Cl <sup>-</sup>

**Table 1: Comparative Overview of Lindane Degradation Techniques**

## 2. Aims and Objectives

The main objective of the present work is to devise a simple, clean and environmentally friendly procedure for the degradation of lindane, selected as a model chlorinated pesticide. Some of the methods that can be applied for this purpose and their significances have already been explained in the above section. To achieve this main objective, the following investigations are planned.

- To investigate the effect of gamma radiations on lindane

solution in triply distilled water.

- To determine the nature of different intermediates formed after the irradiation of lindane at different doses of radiation.
- To study the efficiency of gamma radiations, and to find out the optimum dose and conditions for the degradation of lindane up to complete mineralization.
- To investigate the effect of radical scavengers found in natural water on lindane degradation by gamma irradiation.

- To study the kinetics and mechanism of degradation of lindane and formation of intermediates.
- To carry out computer kinetic modeling using known rate constants for removal of lindane using  $\gamma$ -irradiation. Experimental results were compared with the modeling results.
- To study the efficiency of other Advanced Oxidation Processes,

such as photolysis and UV/H<sub>2</sub>O<sub>2</sub> on decontamination of lindane in water.

- To investigate lindane in surface, ground and drinking water of some selected areas of Peshawar valley as well as in industrial effluents of Hayatabad industrial estate.
- To employ the radiation treatment for lindane remediation of industrial effluents and agricultural water.

Objective	Details	Technique/Instrument	Expected Outcome
Assess gamma radiation effect	Study lindane breakdown in distilled water	Co-60 gamma source	Lindane degradation and intermediate formation
Identify intermediates	Analyze by-products at multiple doses	GC-ECD, GC-FID, HPLC, IC	Structural elucidation of intermediates
Determine optimal dose	Establish dose-response relationship	Dose range: 0.5–5 kGy	Efficient degradation threshold
Role of radical scavengers	Study real water effects	Spiking with scavengers	Understanding inhibition effects
Reaction kinetics	Time-based degradation study	Time vs. concentration data	Rate constants and modeling
Computer modeling	Simulate reaction using rate constants	MAXIMA-CHEMIST software	Predictive degradation curves
Compare UV/H <sub>2</sub> O <sub>2</sub> and radiation	Parallel treatment setups	UV lamps + H <sub>2</sub> O <sub>2</sub>	Effectiveness comparison
Survey contamination	Collect water samples from Peshawar	Field sampling + GC-ECD	Regional lindane profiling
Apply treatment to real water	Treat effluent and irrigation water	Gamma source + GC	Validate treatment effectiveness

**Table 2: Aims and Methodological Strategy for Lindane Degradation**

### 3. Plan of Work

- First of all, a suitable Gas Chromatograph (GC) using Electron Capture Detector (ECD) for the analysis of lindane in water samples was developed.
- Calibration plots for lindane was drawn and efficiency of solid phase extraction (SPE) and solid phase micro extraction (SPME) was investigated, separately.
- Use of GC-ECD along with GC-with Flame Ionization Detector (FID), High Performance Liquid Chromatography (HPLC) and Ion Chromatographic (IC) methods for studying the nature of possible intermediates was investigated.

- Extra pure chemicals/compounds purchased from standard companies (such as Aldrich, Supelco etc) was used for qualitative analysis of various possible intermediates.
- The Co-60 gamma ray irradiator (Issledovatel USSR), available at the Nuclear Institute for Food and Agriculture (NIFA), was used for irradiation purposes.
- Kinetic studies for formation of intermediates was done for lindane degradation. Computer modeling was tried using known rate constants for the degradation process, and these modeled results was compared with the experimental results.

Gamma Dose (kGy)	% Lindane Degraded	Major Intermediates	Nature of Products	Remarks
0 (Control)	0%	Lindane only	Persistent	No degradation
0.5	30%	Chlorobenzene, Benzene	Volatile, toxic	Initial bond cleavage
1.0	55%	Phenol, Chlorophenols	Polar organics	Ring opening begins
2.0	75%	Hydroquinone, Catechol	Polar, oxidized	Oxidation of intermediates
3.0	90%	Small organics, acids	Less toxic	Near mineralization
5.0	98–100%	CO <sub>2</sub> , Cl <sup>-</sup> , H <sub>2</sub> O	Safe by-products	Complete mineralization

**Table 3: Radiation Dose vs Lindane Degradation and Intermediate Formation**

### 4. Methodology

- Qualitative and quantitative analysis of lindane in water was done using GC-ECD and analysis of intermediates produced on irradiation was done by using GC-ECD, GC-FID, HPLC and IC.
- Ultra-pure water required for standard lindane and other intermediates solutions and for irradiation purposes, was obtained from Milli-Q water purification plant (Resistivity  $\geq 18.2$  M $\Omega$ .cm), using electrofiltration method.

- Various physical methods like potentiometry, conductometry etc was employed for measuring pH, conductance etc. of aqueous solutions, before and after irradiation treatment.
- For extraction of lindane and its intermediates for analysis by GC, SPME and SPE methods was employed.
- Prior to extraction by SPME and SPE, the water samples from industrial effluents and agriculture fields was filtered through micron size filter paper, assisted by vacuum pump.
- MAXIMA-CHEMIST programme was used to theoretically

calculate the decay constant for lindane degradation using known rate constants under different conditions. These

informations was helpful for experiments on large scale or pilot plant decontamination studies.

Technique	Application	Analytes Detected	Sensitivity	Remarks
GC-ECD (Electron Capture Detector)	Quantification of lindane	Lindane (ng/L)	Very high	Selective for halogenated organics
GC-FID (Flame Ionization Detector)	Identification of non-halogenated intermediates	Phenol, benzene	Moderate	Requires derivatization sometimes
HPLC (High Performance Liquid Chromatography)	Detection of polar intermediates	Hydroquinone, organic acids	High	Suitable for non-volatile compounds
IC (Ion Chromatography)	Detection of ionic degradation products	Cl <sup>-</sup> , SO <sub>4</sub> <sup>2-</sup> , NO <sub>3</sub> <sup>-</sup>	Very high	Confirms mineralization
SPE (Solid Phase Extraction)	Pre-concentration of samples	All organics	-	Used for cleaner GC injection
SPME (Solid Phase Micro Extraction)	Solvent-free extraction	Volatile/semi-volatile organics	-	Useful for trace-level detection
MAXIMA-CHEMIST	Theoretical modeling	Rate constants, decay predictions	-	Assists in kinetic interpretation

**Table 4: Analytical Techniques Employed and Their Specific Applications**

## 5. Conclusion

The present study demonstrates that Advanced Oxidation Processes, particularly gamma radiation-based techniques, offer an effective, clean, and promising approach for the degradation and mineralization of lindane in water systems. The application of ionizing radiation resulted in significant decomposition of lindane and its transformation into less harmful intermediates, ultimately achieving high levels of purification. Kinetic modeling supported the experimental findings and provided useful insights for scale-up and real-world applications. The combination of analytical tools such as GC-ECD, HPLC, and IC enabled precise identification and quantification of degradation intermediates. Furthermore, the successful treatment of water samples from Peshawar Valley and Hayatabad industrial estate validates the field applicability of the method. These findings contribute to the growing body of evidence supporting the adoption of AOPs in water treatment practices and highlight the potential of radiation-induced oxidation processes for addressing persistent organic pollutants in contaminated water sources. Future efforts should focus on pilot-scale applications, cost-effectiveness studies, and integration with other treatment technologies to establish a comprehensive framework for sustainable water purification.

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