

**REMOVAL OF BISPHENOL A IN WATER SOURCES
CONTAMINATED WITH LEACHATE FROM OPEN
DUMPS OF MUNICIPAL SOLID WASTE USING
NANOMATERIAL-INCORPORATED FRAMEWORK**

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198105A

Doctor of Philosophy

Department of Civil Engineering

Faculty of Engineering

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Sri Lanka

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Thesis submitted in partial fulfillment of the requirements for the degree

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DECLARATION

I declare that this is my own work, and this thesis does not incorporate without acknowledgment any material previously submitted for a degree or diploma in any other University or Institute of higher learning, and to the best of my knowledge and belief it does not contain any material previously published or written by another person except where the acknowledgment is made in the text. I retain the right to use this content in whole or part in future works (such as articles or books).

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DEDICATION

To my beloved husband and parents, for your endless support and love.

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ABSTRACT

Bisphenol A (BPA), an anthropogenic organic compound widely used to manufacture polycarbonate plastics, epoxy resins, and thermal paper receipts, has received significant attention due to its negative health impacts and ubiquitous environmental presence. Its widespread use in consumer goods has led to widespread environmental contamination, with leachate from municipal solid waste (MSW) dumps representing a significant source of the release of BPA into aquatic ecosystems.

The investigation is the first comprehensive study to investigate the BPA levels in leachate emanating from MSW dumps in Sri Lanka. Leachate samples were collected from two prominent dumpsites in the Western Province of Sri Lanka, Karadiyana and Kerawalapitiya. Specifically, at the Karadiyana dumpsite, BPA concentrations ranged from 2.0–10.9 mg/L during the wet period and 8.7–13.5 mg/L during the dry period. In contrast, leachate generated from the Kerawalapitiya dumpsite revealed substantially higher concentrations of BPA, ranging from 13.3–52.3 mg/L in the wet period and 15.1–74.9 mg/L in the dry period. The Kerawalapitiya site exhibited markedly elevated concentrations compared to Karadiyana. The results of the paired-sample t-test indicated significant differences ($p < 0.05$) in BPA concentrations between wet and dry periods at all tested locations in the Karadiyana dumpsite, except for L_1 and L_3 . Similarly, at the Kerawalapitiya dumpsite, all sites except L_1 exhibited significant variations in BPA levels between the two periods. The findings suggest that BPA levels are typically elevated during dry periods in contrast to wet periods. This trend could be attributed to the effect of dilution during wet periods, where increasing water volume can lead to reduced concentrations of BPA.

This study further presents the first investigation into BPA levels in Sri Lanka's surface water bodies and their ecological risk assessments, particularly in fish populations. BPA levels were examined in samples from the Bolgoda Lake and the Kerawalapitiya canal network. In Bolgoda Lake, BPA levels ranged from 3.3 to 29.2 $\mu\text{g/L}$ in the wet period and 1.0 to 1.2 $\mu\text{g/L}$ in the dry period in the first sample set, while the second set showed levels of 1.0 to 10.6 $\mu\text{g/L}$ in the wet period, and 0.8 to 1.5 $\mu\text{g/L}$ in the dry period. Except for specific locations, these concentrations exceeded the Canadian-established threshold of 3.5 $\mu\text{g/L}$. Similarly, the Kerawalapitiya canal network exhibited BPA levels ranging from 0.4 to 42.6 $\mu\text{g/L}$ in the wet period and 0.2 to 4.9 $\mu\text{g/L}$ in the dry period. Significantly, the location of L_{in} and samples located near the

dumpsite exhibited elevated levels, indicating persistent contamination, particularly during the wet period due to leachate runoff.

The correlation analysis revealed that BPA concentrations correlate positively with pH and salinity while showing an inverse relationship with total suspended solids (TSS), temperature, and dissolved oxygen (DO). Furthermore, the ecological risk assessment identified significant risks to aquatic species during different periods. Specifically, Mozambique tilapia, grass carp, and orange chromide face a high acute risk in the wet period and a moderate acute risk in the dry period. Further, Nile tilapia face a high acute risk in both periods. These results underscore the urgent need for effective measures to prevent further contamination and protect aquatic ecosystems and human health.

Evaluation of various nanomaterials for their efficacy in BPA removal revealed that graphene oxide (GO) exhibited the highest efficiency, achieving a removal rate of 74.77%. In contrast, other nanomaterials demonstrated lower effectiveness. Despite their removal capacities, none of these materials reduced BPA levels below the threshold, rendering them unsuitable for significantly lowering BPA levels in treated leachate because of the insufficient removal efficiency under given conditions. Furthermore, powdered activated carbon (PAC) achieved a BPA removal efficiency of 94.79%, while granular activated carbon (GAC) achieved 72.13% efficiency.

In the case of biochar materials, various types demonstrated different removal efficiencies, with king coconut nuts exhibiting the highest at $60.2 \pm 0.6\%$. King coconut nuts contain 29.10% moisture, 33.45% volatile substances, 7.78% ash content, and 29.67% fixed carbon by weight based on proximate analysis. Chemical activation significantly improved the removal efficiency of BPA, particularly with HCl-activated KBC showing the highest efficiency ($80.1 \pm 0.9\%$). Additionally, reducing the particle size of KBC to 75–105 μm enhanced BPA removal efficiency. The efficiency of BPA adsorption remained consistently high within the acidic pH range, maintaining a 100% removal rate. An increase in KBC dose increased BPA removal, reaching 100% at a dosage of 5.0 g/L. Interestingly, the presence of competing ions did not impact the BPA removal efficiency. These findings emphasize the robustness and adaptability of the KBC-based removal method to effectively mitigate BPA contamination under diverse environmental conditions.

Scanning electron microscope (SEM) images revealed that KBC possesses a honeycomb-like porous structure with well-defined pores, resembling carbon nanotubes at the microscopic scale. Energy-dispersive X-ray spectroscopy (EDAX) analysis indicated the composition of KBC, comprising C, O, K, Cl, Si, Na, Ca, Mg, and P. Additionally, Fourier-transform infrared (FTIR) spectra confirmed BPA adsorption onto KBC. The adsorption isotherm results, best suited to the Langmuir isotherm model, suggest monolayer adsorption on KBC, with a maximum adsorption capacity of 39.53 mg/g. The pseudo-second-order model provided the most accurate description of the adsorption dynamics of BPA on KBC, suggesting that chemisorption plays a crucial role as the primary rate-limiting step.

Regeneration and desorption studies of KBC demonstrated that ethanol is the most suitable eluting agent. In studies of fixed bed columns, optimal performance for KBC was observed at a flow rate of 1.0 mL/min, an initial concentration of 100.0 mg/L, and a bed height of 3.0 cm. Moreover, the Thomas and Yoon-Nelson models are closely aligned with experimental data, demonstrating KBC's effectiveness in removing BPA under various conditions. The cost estimate to synthesize 1 g of KBC on a laboratory scale was Rs. 3.28. Lastly, a framework consisting of four layers of KBC pillows effectively treated a synthetic BPA solution at varied flow rates of 1.0, 2.0, and 3.0 mL/min over five days. It achieved 100 % removal of BPA from a total treated volume of 7.2 L, 14.4 L, and 21.6 L, respectively, showcasing its efficient BPA removal capabilities.

This study comprehensively analyses BPA contamination in the Sri Lankan environment, providing valuable information on its sources, distribution, and ecological impacts. The findings underscore the urgent need for effective remediation strategies to mitigate BPA contamination and protect the environment and human health. This research contributes to ongoing efforts to address this pressing environmental challenge by advancing our understanding of the dynamics of BPA contamination and remediation techniques.

Keywords: Bisphenol A, municipal solid waste, endocrine disruptor, king coconut biochar, nanomaterials, adsorption

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LIST OF ABBREVIATIONS

Abbreviation	Description
BPA	Bisphenol A
BPA-KBC	Bisphenol A adsorbed king coconut biochar
BTC	Breakthrough Curve
CEA	Central Environment Authority
CNTs	Carbon nanotubes
DCM	Dichloromethane
DO	Dissolved Oxygen
DSC	Differential Scanning Calorimetry
DW	Dry weight
ECHA	European Chemicals Agency
ESEM	Environmental Scanning Electron Microscopy
EU	European Union
FDA	Food and Drug Administration
FTIR	Fourier Transform-Infrared Spectroscopy
GAC	Granular Activated Carbon
GO	Graphene oxide
HCl-KBC	HCl-activated king coconut biochar
KBC	King coconut biochar
LOD	Limit of Detection
LOQ	Limit of Quantification
MSW	Municipal Solid Waste
MWCNTs	Multi-walled carbon nanotubes
nZVI	Nano zero valent iron

PAC	Powered activated carbon
PEI	Polyetherimide
pH _{PZC}	Point of zero charge
PSU	Polysulphone
PTFE	Polytetrafluoroethylene
RQ	Risk quotient
RSD	Relative Standard Deviation
SD	Standard deviation
SIM	Selective Ion Monitoring
SML	Specific Migration Limits
SONPs	Silver oxide nanoparticles
SPE	Solid Phase Extraction
SRM	Selected Reaction Monitoring
SWCNTs	Single-walled carbon nanotubes
TGA	Thermogravimetric analysis
TiO ₂	Titanium dioxide
TSS	Total Suspended Solids
XRD	X-ray Powder Diffraction

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