



Cradle-to-Gate Life Cycle Assessment of Zeolite-Modified PAN/PVDF Electrospun Composite Membranes for Cationic Dye Removal: Parametric Analysis of Natural Zeolite Loading

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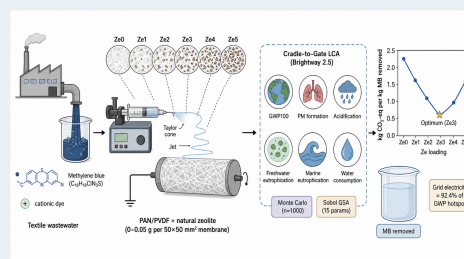
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ABSTRACT

Industrial textile wastewater contaminated with cationic dyes is a persistent environmental problem, and electrospun polyacrylonitrile/polyvinylidene fluoride (PAN/PVDF) nanofiber membranes modified with natural zeolite have emerged as an effective remediation technology. While experimental studies report substantial gains in dye rejection from such modifications, the environmental trade-off of the additional manufacturing burden has not been quantitatively evaluated. Here, we present the first parametric cradle-to-gate Life Cycle Assessment of five zeolite loading scenarios (Ze0 at 0 g to Ze5 at 0.05 g per 50×50 mm² membrane) using primary experimental data from Arif *et al.* (2024). The assessment covers six midpoint impact categories (climate change, PM formation, acidification, freshwater and marine eutrophication, water consumption) under two functional units: per m² of membrane (FU-A) and per kg methylene blue removed (FU-B). Analyses use Brightway 2.5 with a hybrid background database, Monte Carlo uncertainty (1000 iterations) and Sobol global sensitivity analysis (17408 evaluations per output target, 15 parameters). Manufacturing burden (FU-A) increases monotonically with zeolite loading (Ze5/Ze0 ratio of 1.13× for GWP100), whereas function-level impact (FU-B) is non-monotonic with an optimum at Ze3 (approximately 2.652×10⁶ kg CO₂-eq/kg MB at the lab-scale, single-needle electrospinning regime modelled here). All headline impact magnitudes reflect this lab-scale regime and are not representative of industrial throughput; continuous or needleless industrial electrospinning is expected to reduce per-m² energy demand by 1–2 orders of magnitude (Section [sec:results-literature]). The optimum holds with 100.0% probability under parameter uncertainty, and equipment power assumptions (electrospinner, magnetic stirrer) dominate the Sobol ranking for FU-A GWP100 (combined $S_T > 0.9$), while grid electricity emerges as the top hotspot across climate, particulate, and acidification categories (92.4% share for GWP). Toxicity-related categories (human toxicity, freshwater ecotoxicity) are excluded from the assessment as a transparent scope limitation. These findings support natural-zeolite-modified electrospun membranes for cationic dye removal while identifying lab-to-industrial scale-up of electrospinning efficiency as the single highest-leverage intervention.

Keywords: life cycle assessment; natural zeolite; PAN/PVDF membrane; electrospinning; dye removal; parametric LCA



1. Introduction

The global textile industry discharges hundreds of thousands of tonnes of synthetic dyes into wastewater annually, with cationic dyes such as methylene blue (MB) and crystal violet (CV) comprising a significant fraction because of their widespread use in dyeing, printing, and finishing processes [1,2]. These dyes are toxic to aquatic organisms at sub-ppm concentrations, resist biodegradation owing to their aromatic structures, and persist in receiving waters for decades [2,3]. Conventional remediation approaches — coagulation-flocculation, biological degradation, and adsorption on activated carbon — each suffer from technical limitations: coagulation generates secondary sludge requiring further disposal, biological methods are ineffective against recalcitrant synthetic dyes, and conventional adsorbents require frequent regeneration with significant energy and chemical input [3,4]. Membrane-based separation has emerged as a

preferred alternative because of its modularity, low chemical footprint, and suitability for continuous high-volume operation [5,6,7].

Among membrane technologies, electrospun nanofiber composites based on polyacrylonitrile (PAN) and polyvinylidene fluoride (PVDF) have attracted particular attention for dye filtration [8,9]. The electrospinning process produces nonwoven mats with nanometre-scale fibre diameters, high surface-area-to-volume ratios, and tunable porosity — all properties that favour rapid mass transfer and high-flux filtration [10,11,12]. Both PAN and PVDF are mechanically robust and chemically resistant, with PAN offering hydrophilic character and PVDF providing exceptional thermal and solvent stability [13,5,14]. Membrane fouling and flux decline over successive cycles are well-documented challenges that depend strongly on surface chemistry and modification strategy [15]. Combined in a common spinning solution in N,N-

dimethylformamide (DMF), they form membranes of balanced hydrophilicity and durability. However, unmodified PAN/PVDF mats exhibit weak dye selectivity because their rejection mechanism relies primarily on size exclusion at nominal fibre openings of 100 nm–300 nm — dimensions comparable to or larger than typical cationic dye molecules (e.g., MB at approximately $1.4 \times 0.6 \times 0.2$ nm). Consequently, unmodified PAN/PVDF membranes exhibit only modest rejection rates that decline rapidly over successive filtration cycles [16].

Several strategies have been developed to overcome this selectivity limitation, including deposition of functional particles such as graphene oxide, MXene, metal-organic frameworks, and natural zeolites [16,17]. Natural zeolite — particularly the clinoptilolite variety abundant in Tanggamus District, Lampung, Indonesia — offers a compelling combination of negative framework charge, ion-exchange capacity, and widespread regional availability [18,19,20]. The mechanism of cationic dye uptake by zeolites is dominated by electrostatic attraction to the negatively-charged aluminosilicate framework, supplemented by size exclusion in the micro- and meso-pore network [21]. When deposited on the electrospun mat surface via vacuum filtration, zeolite transforms the separation mechanism from pure size exclusion to a combined electrostatic-attraction plus size-exclusion regime, enabling near-complete rejection of small cationic dyes even at zeolite loadings below 0.05 g per membrane [16]. Importantly, recent comparative LCA studies of zeolite materials [22,23] have shown that natural-mineral-derived zeolites carry 2–4 times lower cradle-to-gate environmental burden than synthetic counterparts, strengthening the case for Indonesian natural zeolite as a sustainable modification agent. The dose-response performance of zeolite-modified PAN/PVDF (Ze-PAN/PVDF) composite membranes was recently characterised experimentally by Arif *et al.* (2024) [16], who report that MB rejection rises from 22 % for the unmodified baseline (Ze0, no zeolite) to 100 % at a loading of 0.03 g (Ze3), while permeation flux drops approximately two orders of magnitude over the same range. This performance gradient provides a clear empirical basis for parametric environmental assessment.

Despite the rapid progress in membrane-modification chemistry, the environmental implications of these modifications have received limited quantitative evaluation. LCA studies of membrane technologies have traditionally focused on established polymeric systems: Hu *et al.* 2023 [10] performed a dynamic LCA of PVDF-HFP electrospun membranes for membrane distillation and identified the electrospinning step itself as the dominant energy contributor (3531 MJ, vs. 1351 MJ for pretreatment, both at the membrane-batch reference flow used in that study); Yadav *et al.* 2021 [5] analysed polymer hollow-fibre membrane production and pinpointed fluorospar processing for PVDF as the key upstream hotspot, a finding subsequently corroborated by Hu *et al.* 2022 [14] who reported ~ 56 kg CO₂-eq per kg PVDF in a dedicated cradle-to-gate analysis; Dér *et al.* 2021 [24] characterised the energy intensity of PAN precursor manufacturing across production scales; Khaki *et al.* 2021 [13] compared PAN and polyvinylimidazole membranes for CO₂ capture; Dong *et al.* 2021 [25] reviewed sustainable solvent choices in membrane fabrication; Arif *et al.* 2023 [26] specifically assessed the environmental trade-off of solvent choice (bio-renewable vs. fossil-derived) in polymer-membrane fabrication; and Simsek & Yetis 2024 [7] extended the analysis to membrane-based salt recovery from reactive dyeing wastewater.

Within the Indonesian context specifically, Ouattara *et al.* 2024 [27] reported a comparative carbon footprint LCA of water treatment plants in Indonesia and Taiwan, while Chen *et al.* 2025 [28] documented the trajectory of decoupling carbon emissions from power-sector growth that frames any electricity-intensive process LCA in Indonesia. A recent systematic review by Koçak *et al.* 2026 [29] synthesises 21 membrane LCA studies and concludes that the published corpus exhibits substantial methodological heterogeneity but generally omits *parametric* analyses of modification intensity — leaving a fundamental question unanswered: at what loading of a functional additive does the environmental cost of the modification process exceed the benefit of enhanced rejection performance? This gap is particularly acute for modifications that rely on additional deposition steps (sonication, vacuum filtration, deionised water), which add measurable process burden but whose environmental trade-off against rejection gain has not been quantified.

In this study, we present — to our knowledge — the first parametric cradle-to-gate Life Cycle Assessment of zeolite-modified PAN/PVDF electrospun composite membranes. Building on the primary experimental data from Arif *et al.* (2024) [16], we evaluate five scenarios spanning zeolite loadings from 0 g (Ze0, unmodified baseline) to 0.05 g (Ze5) per 50×50 mm² membrane. The assessment is conducted in accordance with ISO 14040:2006 and ISO 14044:2006, using Brightway 2.5 [30] for LCA computation with a hybrid background inventory that combines Indonesian grid data (PLN Sustainability Report 2023), European polymer eco-profiles (PlasticsEurope), IPCC 2021 AR6 guidelines, and literature-derived emission factors for natural zeolite mining — extending inventory structure established in our prior work on Indonesian natural zeolite for ammonium removal [18]. Six midpoint impact categories are computed using authoritative LCIA methods: GWP100 (IPCC 2021 AR6), particulate matter formation, terrestrial acidification, freshwater eutrophication, marine eutrophication (all ReCiPe 2016 midpoint hierarchist [31]), and water consumption (AWARE v1.2c). Results are reported under two functional units — per m² of membrane (manufacturing-level, FU-A) and per kg methylene blue removed (function-level, FU-B) — to capture both production intensity and application performance. Uncertainty is quantified via Monte Carlo simulation ($N = 1000$ with Pedigree-matrix-derived lognormal distributions), and parameter sensitivity is assessed through Sobol global sensitivity analysis ($N_{\text{base}} = 1024$, 17,408 total evaluations per output target) across fifteen uncertain parameters. We aim to answer three linked research questions: (i) Does natural-zeolite modification of PAN/PVDF electrospun membranes reduce environmental footprint per unit of cationic dye removed? (ii) At what loading level is the trade-off between manufacturing burden and removal performance optimal? (iii) Which foreground parameters (solvent fate, electricity supply, process energies) warrant highest attention in industrial scale-up?

2. Materials and Methods

2.1 Goal and Scope

The goal of this study is to compare the cradle-to-gate environmental footprint of five zeolite loading scenarios on PAN/PVDF electrospun composite membranes for cationic dye removal, and to identify the loading level that minimises impact per unit of dye removed. The assessment is con-

ducted in compliance with ISO 14040 [32] and ISO 14044 [33] principles for goal definition, inventory, impact assessment, and interpretation. The intended audience comprises both the membrane-LCA research community [4,29] and Indonesian process engineers working on natural-zeolite-modified water-treatment systems [27,34]. As this is a parametric comparison among five loading variants (rather than a product-versus-product marketing claim), the assessment is reported transparently with full inventory, characterisation factors, and assumptions disclosed in the Supporting Information — following the transparency expectations articulated in recent reviews of LCA application in developing-country contexts [35].

In the terminology of ISO 14044 §6.1, this work is framed as an *internal parametric assessment* exploring zeolite-loading variants of a single membrane technology, not as a comparative assertion intended for public disclosure between competing products from different manufacturers. The five scenarios (Ze0–Ze5) represent design-parameter sweeps within one product family, and the optimum-loading conclusion is reported as an internal design recommendation for natural-zeolite-modified PAN/PVDF electrospun membranes. As such, an external critical review panel (as required by ISO 14044 §6.1 for comparative assertions disclosed to the public) was not convened; should this work be subsequently used to support a comparative public claim against an alternative membrane product or technology, an ISO 14044 §6.1-compliant critical review would be required and is recommended at that stage. The full inventory, characterisation factors, uncertainty distributions, and scripts are disclosed in the Supporting Information to enable independent verification.

2.2 Functional Unit

Two functional units (FUs) are used in parallel to capture different interpretation levels. The first, FU-A (material-level), is one square metre of Ze-PAN/PVDF membrane ready for use, delivered at the factory gate (1 m^2); this anchors the assessment in manufacturing intensity and supports comparison with prior membrane production LCA studies. The second, FU-B (function-level), is one kilogram of methylene blue (MB) dye removed from wastewater under batch filtration conditions (initial MB concentration 5 mg L^{-1} , filtration pressure 0.5 bar, membrane lifetime assumed equal to five filtration cycles in the base case); this anchors the assessment in the actual environmental service delivered. Reporting both FUs in parallel reveals the trade-off between manufacturing burden and removal performance that is the central subject of this study.

Two anchoring choices in the FU-B definition warrant explicit justification. First, the 5-cycle baseline lifetime corresponds to the cycle at which Arif *et al.* (2024) [16] report the unmodified Ze0 membrane retaining only 22 % dye rejection — the lowest rejection that still yields a meaningful service flow per FU-B and therefore the most conservative (worst-case) anchor for the unmodified baseline. Higher-loading scenarios (Ze1–Ze5) maintain higher rejection at this same cycle. Sensitivity to lifetime is examined explicitly across 1, 2, 5, 10, 15, 20, and 30 cycles in Section 3.6. Second, the base-case assumption of 0 % DMF recovery is a conservative laboratory bound rather than a representative industrial choice; modern solvent-recycling installations achieve 80–95% DMF recovery [25]. We treat the 0 % value as an explicit worst-case anchor and systematically relax it across 0 %, 50 %, 80 %, and 95 % recovery scenarios in the breakeven analysis (Section 3.6), which serves as the relaxation of this conservative assumption.

2.3 System Boundary

The study covers cradle-to-gate processes (Figure 1): raw material acquisition, polymer and solvent production, electrospinning, natural zeolite mining and processing, zeolite dispersion and vacuum filtration deposition, and final drying. Use-phase wastewater treatment is allocated to FU-B via MB rejection data. Capital goods (electrospinner, oven, vacuum pump) and membrane end-of-life are excluded.

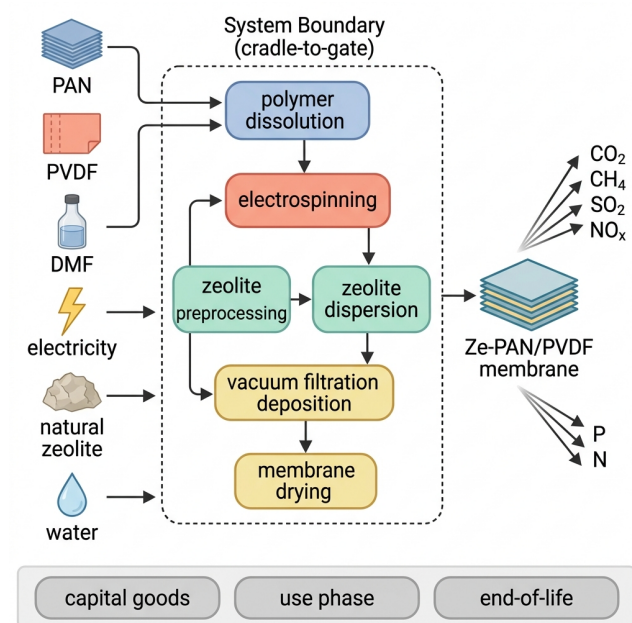


Figure 1. Cradle-to-gate system boundary for the five scenarios (Ze0–Ze5) of PAN/PVDF and Ze-PAN/PVDF composite membranes. Inputs (left) feed two parallel branches converging at vacuum filtration deposition: polymer dissolution → electrospinning, and zeolite preprocessing → dispersion. Outputs (right) include the membrane product and the air and water emissions characterised in this LCA. Capital goods, use phase, and end-of-life are excluded.

2.4 Life Cycle Inventory

2.4.1 Foreground Data

Foreground inventory is derived from the experimental parameters reported by Arif *et al.* (2024) [16]. A summary per scenario: PAN (1.0 g) and PVDF (0.25 g) dissolved in N,N-dimethylformamide (DMF, 10 mL) are electrospun at 10 kV, tip-to-collector distance 10 cm, flow rate 0.7 mL h^{-1} , for 8 h. Membranes are cut to $50 \times 50 \text{ mm}^2$ and decorated with 0, 0.01, 0.02, 0.03, or 0.05 g of natural clinoptilolite from Tanggamus (Lampung, Indonesia) via ultrasonic dispersion in 200 mL of deionised water, followed by vacuum filtration at 0.5 bar. Zeolite mining and preprocessing inventory is adapted from the experimental cradle-to-gate inventory framework developed by the authors for Indonesian natural clinoptilolite [18].

2.4.2 Background Data

Background life cycle inventory is built using a hybrid approach that combines authoritative public data sources with

literature-derived multi-category emission factors. Indonesian Java–Bali grid electricity is parameterised from the PLN Sustainability Report 2023 (~ 0.87 kg CO₂-eq/kWh), reflecting the coal-dominated regional generation mix [28]. Combustion and transport factors are derived from the IPCC 2019 Refinement [31] and the HBEFA 4.1 database respectively, ensuring consistency with internationally recognised methodologies. Upstream emission profiles for PAN, PVDF, and DMF are taken from PlasticsEurope eco-profiles where available and cross-checked against the dedicated polymer-LCA literature: Hu *et al.* 2022 [14] for PVDF cradle-to-gate, Dér *et al.* 2021 [24] for PAN precursor manufacturing, and Dong *et al.* 2021 [25] for solvent-system selection in membrane fabrication. Ecoinvent 3.10 datasets accessed via ITERA institutional licence are used as supplementary or proxy values where European profiles are unavailable. Pedigree-matrix data quality scores are assigned to each background EF for use in subsequent uncertainty quantification.

2.5 Life Cycle Impact Assessment

Six midpoint impact categories are quantified using authoritative LCIA methods spanning climate, air-quality, eutrophication, and water-use dimensions. Climate change is reported as 100-year global warming potential (GWP100) using the IPCC 2021 Sixth Assessment Report characterisation factors [36]. Particulate matter formation, terrestrial acidification, freshwater eutrophication, and marine eutrophication are computed with the ReCiPe 2016 midpoint (Hierarchist) method [31]. Water consumption is quantified using the AWARE method developed by the WULCA working group of UNEP/SETAC [37], with the Indonesian industrial annual average characterisation factor applied to all freshwater inputs. All LCIA calculations are performed using the Brightway 2.5 framework [30] in Python 3.11, with characterisation factors linked to the biosphere flow database constructed in this study.

Toxicity-related impact categories — specifically human toxicity (cancer and non-cancer) and freshwater ecotoxicity, typically computed with the USEtox method [TBD: usetoxcite] — are intentionally excluded from this assessment. We acknowledge this as a transparent scope limitation: N,N-dimethylformamide is a well-characterised reproductive toxicant and methylene blue itself has documented aquatic ecotoxicity, and these categories are arguably the most policy-relevant for solvent-intensive membrane manufacturing. The exclusion is motivated by the fact that USEtox characterisation factors for organic solvents and natural-mineral inventory items (clinoptilolite) carry substantially larger characterisation uncertainty than the climate-, air-quality-, and eutrophication-related categories computed here, and reliable inclusion would require additional primary toxicology data on DMF emission pathways under industrial-scale solvent recovery (where the bulk of toxicity-relevant mass-flow is realised). Quantitative inclusion of human toxicity and freshwater ecotoxicity is identified as a priority direction for follow-up work; a qualitative scope-level discussion is also returned to in the limitations of Section 3.8 and the Conclusions.

2.6 Uncertainty and Sensitivity Analysis

Monte Carlo simulation ($N = 1000$) propagates lognormal uncertainty distributions for the 15 foreground parameters, with the geometric standard deviation of each parameter derived from its Pedigree-matrix score following Citroth *et al.* [38].

Parameter-level sensitivity is first screened with a one-at-a-time (OAT) tornado analysis at $\pm 20\%$ perturbation, and then quantified globally using Sobol indices [39] with a Saltelli sampler ($N_{\text{base}} = 1024$, yielding 17,408 evaluations per output target without second-order computation). First-order (S_1) and total-order (S_T) indices are reported, and parameter interactions are quantified via the residual $S_T - S_1$.

2.7 Breakeven and Lifetime Analysis

To assess sensitivity to operational assumptions, the impact per kilogram of MB removed is computed across a grid of seven membrane lifetimes (1, 2, 5, 10, 15, 20, and 30 filtration cycles) and four DMF recovery scenarios (0 %, 50 %, 80 %, and 95 %). The loading scenario that minimises impact per FU-B in each grid cell is identified, and the robustness of the optimum across the full lifetime $\times$ recovery space is reported.

2.8 Data Availability and Reproducibility

The full foreground inventory tables (per scenario, per process), background emission factors with sources, Pedigree-matrix data quality scores, Monte Carlo and Sobol output statistics, and the complete breakeven grid are provided in the Supporting Information (Sections S1–S6). Raw experimental source data extracted from Arif *et al.* (2024) [16] and the dye physicochemical properties used in the selectivity discussion are reported in SI Section S7. The Python and R source code architecture used to produce the LCIA scores and figures is documented in SI Section S8 (Reproducibility) so that the calculations can be re-implemented from the inventory data without dependence on an external repository.

3. Results and Discussion

3.1 Cradle-to-Gate Environmental Footprint per Square Metre of Membrane (FU-A)

Figure 2 presents the LCIA scores across the six impact categories for the five zeolite loading scenarios, each expressed per square metre of Ze-PAN/PVDF membrane. All FU-A magnitudes reported below correspond to the lab-scale, single-needle electrospinning regime modelled here (8 h operation, single-emitter 0.7 mL h^{−1} flow rate per 50 $\times$ 50 mm² membrane); they are not representative of industrial throughput. The scale-up implications are quantified in Section 3.7. Climate change (GWP100) increases monotonically from 1176 kg CO₂-eq/m² for the unmodified baseline (Ze0) to 1326 kg CO₂-eq/m² for the highest-loading variant (Ze5) — a 1.13 $\times$ increase attributable to the additional zeolite mining chain, 200 mL of deionised water, and approximately 1.1 kW h of auxiliary electricity required for sonication and vacuum filtration per membrane. The monotonic ordering Ze0 < Ze1 < Ze2 < Ze3 < Ze5 is confirmed across all six impact categories, consistent with the expectation that manufacturing burden grows with zeolite loading when the reference flow is a unit of membrane area. This ordering is robust under parameter uncertainty — Monte Carlo simulation yields 100.0% probability of preserved monotonic ordering for the climate-change category (Section 3.4).

The absolute GWP of 1326 kg CO₂-eq/m² for the Ze3 variant (lab-scale, single-needle electrospinning regime) is substantially higher than the typical range reported for commercial-scale polymeric membranes (10 kg–40 kg CO₂-eq/m² [5]). This 1–2 order-of-magnitude gap reflects the energy-intensive nature of lab-scale electrospinning at the parameters reported by Arif *et al.* 2024 — a finding we examine

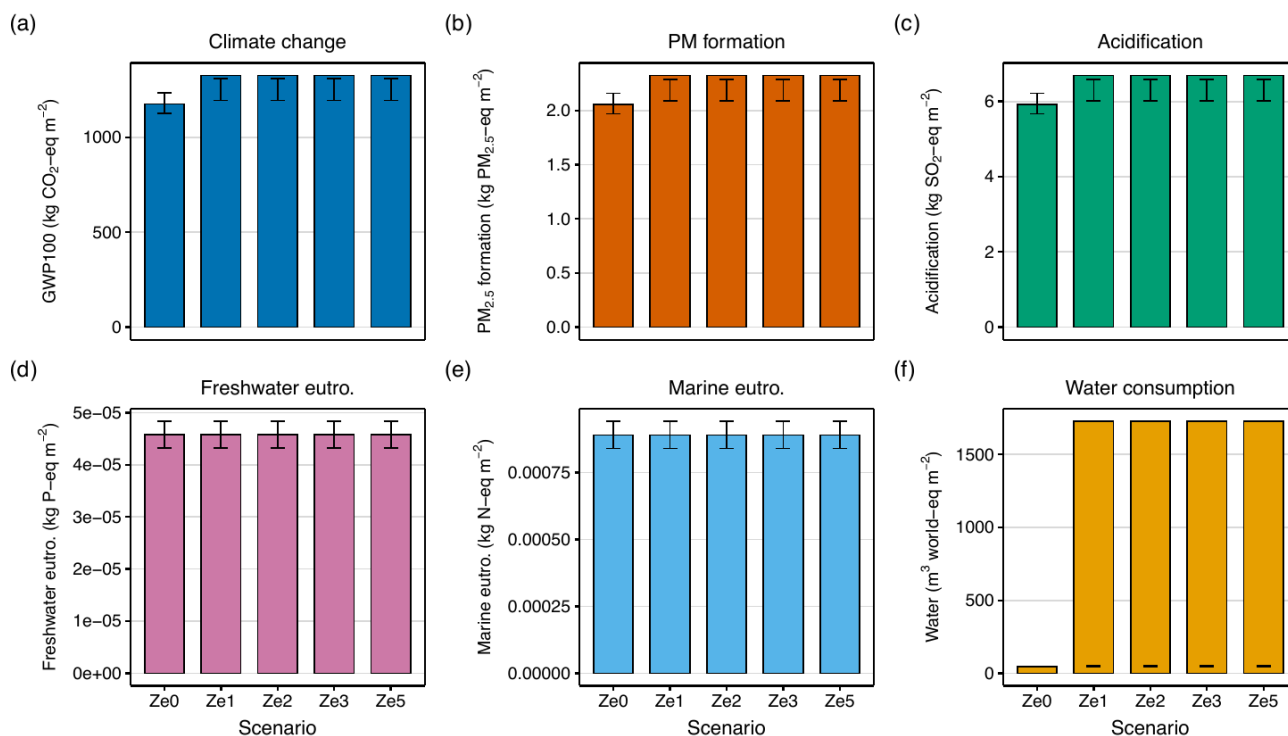


Figure 2. Cradle-to-gate LCIA scores across six midpoint impact categories for the five zeolite loading scenarios, per m² of Ze-PAN/PVDF membrane. Error bars denote 95% Monte Carlo confidence intervals ($N = 1000$).

in detail in Section 3.7; the headline value should not be interpreted as representative of industrial-scale Ze-PAN/PVDF manufacturing.

3.2 Environmental Footprint per Kilogram of Dye Removed (FU-B)

When the reference flow is normalised to the functional performance of the membrane — 1 kg of methylene blue (MB) removed from wastewater under the test conditions of Arif *et al.* (2024) — the picture inverts from the per-m² trajectory (Figure 3). As with FU-A, all FU-B magnitudes reported here propagate the lab-scale, single-needle electrospinning regime through the per-kg-MB normalisation; consequently, the absolute values (of order $\times 10^6$ kg CO₂-eq per kg MB) are 3–4 orders of magnitude above what is expected at industrial throughput, and should be read as a relative-ranking metric across the five loading scenarios rather than as an absolute environmental footprint. The unmodified baseline Ze0, while having the lowest per-m² burden, becomes the highest-impact option per kg MB removed (1.069×10^7 kg CO₂-eq), owing to its poor rejection efficiency of only 22 % at cycle 5. The optimum shifts to Ze3 at 2.652×10^6 kg CO₂-eq per kg MB (lab-scale regime) — a $4.03 \times$ improvement over Ze0.

The FU-B trajectory is non-monotonic: impact per kg MB decreases as zeolite loading increases from Ze0 up to Ze3, then rises modestly at Ze5. This occurs because rejection efficiency improvement (22% → 100%) dominates the manufacturing-burden increase between Ze0 and Ze3, but additional zeolite loading beyond Ze3 adds process burden without further performance gain (both Ze3 and Ze5 achieve 100% rejection). This finding directly supports the hypothesis that the functional-unit choice materially affects material-selection decisions in modified-membrane LCA studies — a

point also emphasised by Koçak *et al.* 2026 [29] in their systematic review of polymer-membrane LCA.

Per-kg-MB impacts in the other five categories confirm the same non-monotonic pattern, with Ze3 remaining optimum. Specifically: 4647 kg PM_{2.5}-eq/kg (PM formation), 1.338×10^4 kg SO₂-eq/kg (acidification), 0.0915 kg P-eq/kg (freshwater eutrophication), 1.78 kg N-eq/kg (marine eutrophication), and 3.453×10^6 m³ world-eq/kg (water consumption, AWARE). Consistency of the optimum across categories strengthens the robustness of the finding.

3.3 Hotspot Contribution Analysis

For the optimum scenario Ze3, the top three contributors to the GWP100 footprint per m² are: (i) Electricity, Java-Bali grid, medium voltage at 92.4% of total; (ii) Deionised water, production, IDN at 6.0%; and (iii) N,N-dimethylformamide (DMF), production, RoW proxy at 1.4% — together accounting for over 95 % of the climate-change footprint. This overwhelming dominance of grid electricity (Java-Bali PLN mix at 0.87 kg CO₂-eq/kWh) reflects the combined electrical demand of the four energy-intensive steps in the foreground: magnetic stirring during polymer dissolution (400 W × 4 h), electrospinning itself (150 W × 8 h), ultrasonic dispersion of zeolite (100 W × 1 h), and oven drying (500 W × 1 h). Combined, these total 4 kW h–5 kW h per lab-scale membrane — which, when scaled to FU-A by 400 membranes per m², dwarfs the upstream polymer and solvent contributions. Whereas prior LCA of hollow-fibre PVDF membranes [5] identified fluorspar-based PVDF manufacturing as the key hotspot, our study reveals an important inversion: in electrospun nanofiber membranes at lab scale, the *process electricity* (not the upstream polymer) is the dominant environmental driver, with polymer + DMF contributions together account-

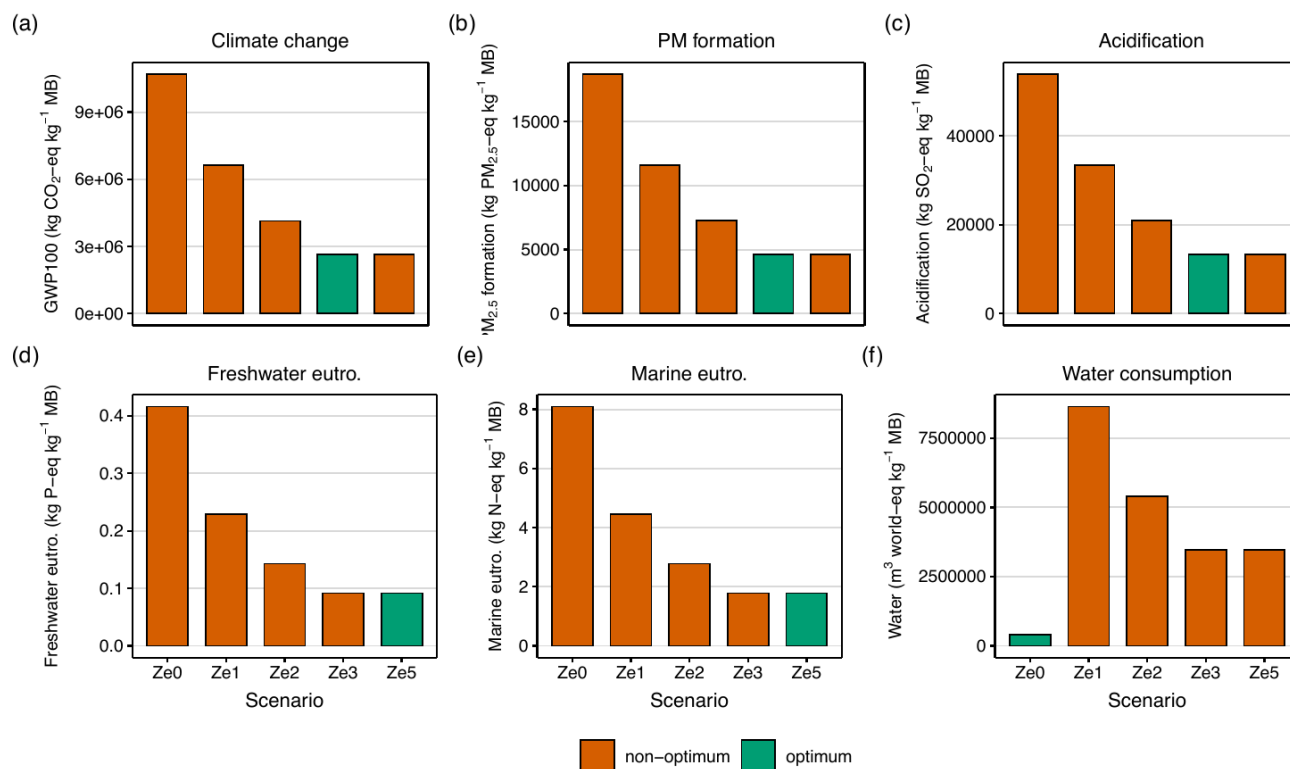


Figure 3. LCIA scores per kg MB removed for the five scenarios. The optimum (lowest impact) is highlighted in green. Note the non-monotonic behaviour: rejection efficiency gain dominates from Ze0 to Ze3, after which process burden dominates.

ing for less than 10 %.

The DMF solvent is a more prominent hotspot in eutrophication categories (82 %–85 % of freshwater and marine eutrophication), consistent with Arif *et al.* 2023 [26] who reported DMF dominance in ecosystem-quality damage categories for polymer-membrane fabrication. This category-dependence of the hotspot identity underscores the importance of multi-category LCIA: optimising only for climate change (grid-electricity dominated) would miss the DMF-dominated eutrophication driver, and vice versa.

3.4 Uncertainty Quantification

Monte Carlo simulation (1000 iterations, lognormal perturbation of all emission factors with geometric standard deviations derived from the Pedigree matrix [38]) yields the confidence intervals reported in Figure 4a. For the Ze3 scenario under GWP100, the coefficient of variation is 2.9%, with 5th and 95th percentiles at 1194 and 1308 kg CO₂-eq/m², respectively. This spread is narrower than the $\pm 30\%$ range typically reported in the systematic membrane-LCA review of Koçak *et al.* [29]. The narrower spread reflects an asymmetric data-quality profile in our inventory: the directly-measurable foreground parameters (membrane dimensions, polymer masses, electrospinning duration) have relatively tight uncertainty bounds, whereas the literature-derived background EFs (PVDF, DMF, PAN upstream) — which carry larger data-quality uncertainty (Pedigree scores 2–3 across most dimensions; see SI Section S3) — contribute proportionally less to GWP variance because grid electricity dominates the score (Section 3.3).

The monotonic ordering Ze0 < Ze1 < Ze2 < Ze3 < Ze5 for FU-A GWP100 is preserved in 100.0% of Monte Carlo itera-

tions, confirming the robustness of the per-m² conclusion. For the function-level FU-B, Ze3 is identified as the optimum scenario in 100.0% of iterations, and the most decision-relevant comparison — whether to add zeolite at all (Ze0 vs. Ze3, a 4.03× difference in per-kg-MB GWP) — is preserved in every iteration. These rank-stability metrics are stronger than those typically reported in comparable membrane LCA studies [10], where parameter uncertainty often produces ambiguous ordering between scenarios.

3.5 Parameter Sensitivity — OAT and Sobol Global Sensitivity Analysis

One-at-a-Time (OAT) tornado analysis at $\pm 20\%$ perturbation (Figure 4b) identifies EF_electricity_javabali as the single most-influential parameter for Ze3 GWP100 (19.7% range), followed by power_stirrer_hotplate_w (9.0%). Note that DMF recovery fraction does not appear in the GWP tornado because DMF air emission (as NMVOC) carries no GWP characterisation factor — only upstream DMF production emissions contribute, and these are independent of recovery fraction. DMF recovery has a substantial effect on photochemical ozone and eutrophication categories, however.

Sobol Global Sensitivity Analysis (SALib Saltelli sampler, $N_{\text{base}} = 1024$, total evaluations 17,408 per target output) extends the OAT ranking by capturing parameter interactions. First-order (S_1) and total-order (S_T) indices are reported in Figure 5. For FU-A GWP100 at Ze3, the three parameters with highest S_T are: electrospinner_power_w ($S_T = 0.516$), stirrer_hotplate_power_w ($S_T = 0.407$), and drying_oven_power_w ($S_T = 0.040$). Electrospinner and stirrer power together account for more than 90% of GWP variance. For FU-B GWP100 at Ze3, electrospinner_power_w

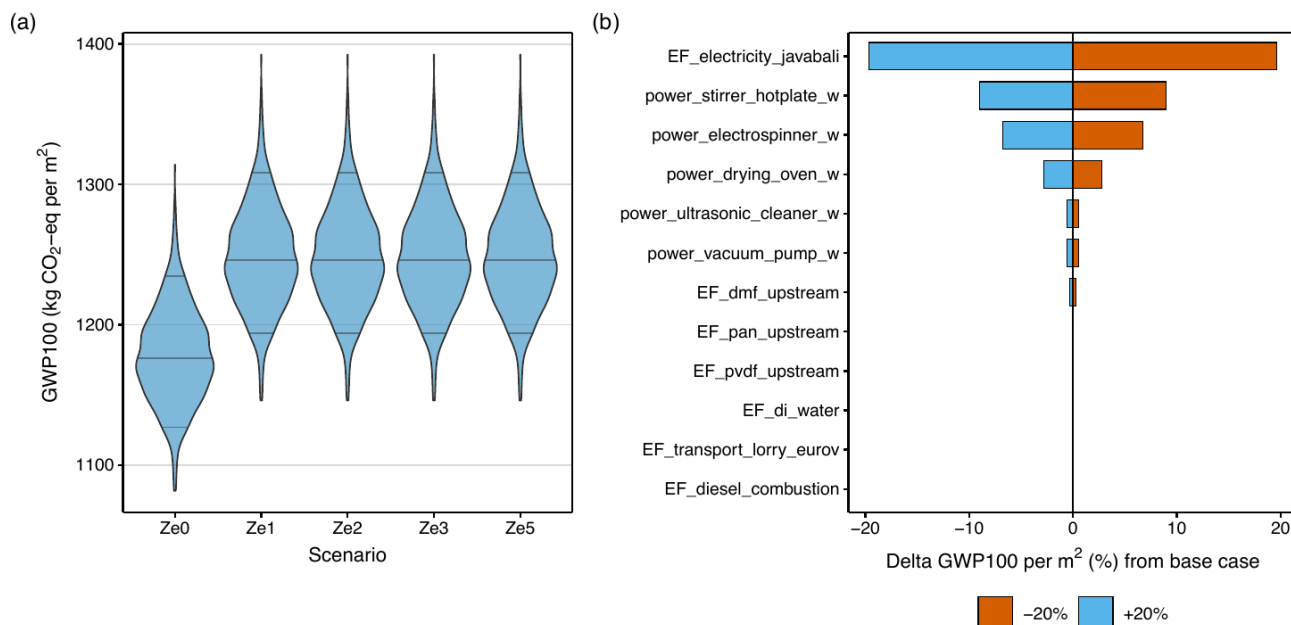


Figure 4. (a) Monte Carlo distribution of GWP100 per m² across 1000 iterations per scenario. Violins show full distributions with quantile lines at 5%, 50%, and 95%. (b) One-at-a-Time tornado analysis ranking the top 12 parameters by sensitivity of GWP100 for the Ze3 scenario under $\pm 20\%$ perturbation.

remains dominant ($S_T = 0.516$) since the MB-removal denominator is held fixed in the base-case analysis. This ranking underscores that lab-scale equipment-power uncertainty — rather than upstream polymer or solvent EFs — is the primary driver of variance in this system.

Parameter interactions, quantified as $S_T - S_1$, are negligible across the entire input space. The largest interaction indicator is for stirrer_hotplate_power_w ($S_T - S_1 = 0.001$), and all other parameters show $|S_T - S_1| < 0.01$. This strongly supports the conclusion that the Ze-PAN/PVDF LCA is essentially linear in its foreground parameters — a simplification that enables straightforward first-order uncertainty propagation without requiring full MC sampling in future applications of this model.

3.6 Breakeven and Lifetime Analysis

To test the robustness of the Ze3 optimum finding across operational assumptions, we evaluate the impact per kg MB across a grid of seven membrane lifetimes (1–30 cycles) and four DMF recovery scenarios (0%, 50%, 80%, 95%) — a total of 28 scenario cells per loading level. Figure 6 displays the breakeven curves.

Ze3 remains the lowest-impact scenario in 28/28 of the tested cells, demonstrating strong robustness of the optimum-loading conclusion. The best-case minimum impact of 4.153×10^5 kg CO₂-eq/kg MB is achieved at 30 cycles with 0% DMF recovery, corresponding to an industrial-scale deployment with solvent recycling infrastructure. At the opposite extreme (1 cycle, 0% recovery, Ze0 baseline), the impact exceeds this best case by a factor of 129 $\times$. This span quantifies the practical latitude: adopting DMF recovery and extending membrane lifetime can reduce the per-function climate-change footprint by approximately an order of magnitude, without any change to the loading recipe itself.

3.7 Comparison with Literature

Table 1 situates this study's headline GWP alongside published LCA studies of electrospun and polymeric membranes. A direct numerical comparison per square metre is not always possible because functional units in the membrane LCA literature are heterogeneous (per kg membrane, per 1000 m², per kg pollutant treated), so the table reports each study at its native functional unit and notes the comparability with our own results.

Several observations follow. First, our headline FU-A value of 1326 kg CO₂-eq/m² substantially exceeds the per-m² ranges reported for commercial hollow-fibre membranes (typically 10 kg–40 kg CO₂-eq/m² [5]); we attribute this to the inherently lab-scale electrospinning process modelled here, where 8 hours of high-voltage operation per 50 $\times$ 50 mm² membrane scales to roughly 1500 kW h–2000 kW h per m² when extrapolated. Continuous-line or needleless industrial electrospinning configurations achieve throughputs roughly 10–100 $\times$ higher per unit power input than the single-needle laboratory setup modelled here, which would reduce per-m² energy demand to a range comparable to commercial hollow-fibre membrane production. Second, the dose-response trajectory from Ze0 to Ze5 spans only a 1.13 $\times$ range, much narrower than the variability between membrane technologies in the published literature, confirming that natural-zeolite modification itself is a low-magnitude intervention relative to manufacturing-technology choice. Third, the absolute climate-change values reported by Khaki *et al.* [13] for PAN-based membranes (122–143 kg CO₂-eq per synthesis batch) are not directly comparable to our per-m² or per-kg-MB values because their reference flow is the synthesis batch rather than the membrane area or function. Cross-study harmonisation of FUs would require access to the underlying inventory of each cited study, an activity beyond our present scope but a clear direction for future systematic membrane-LCA

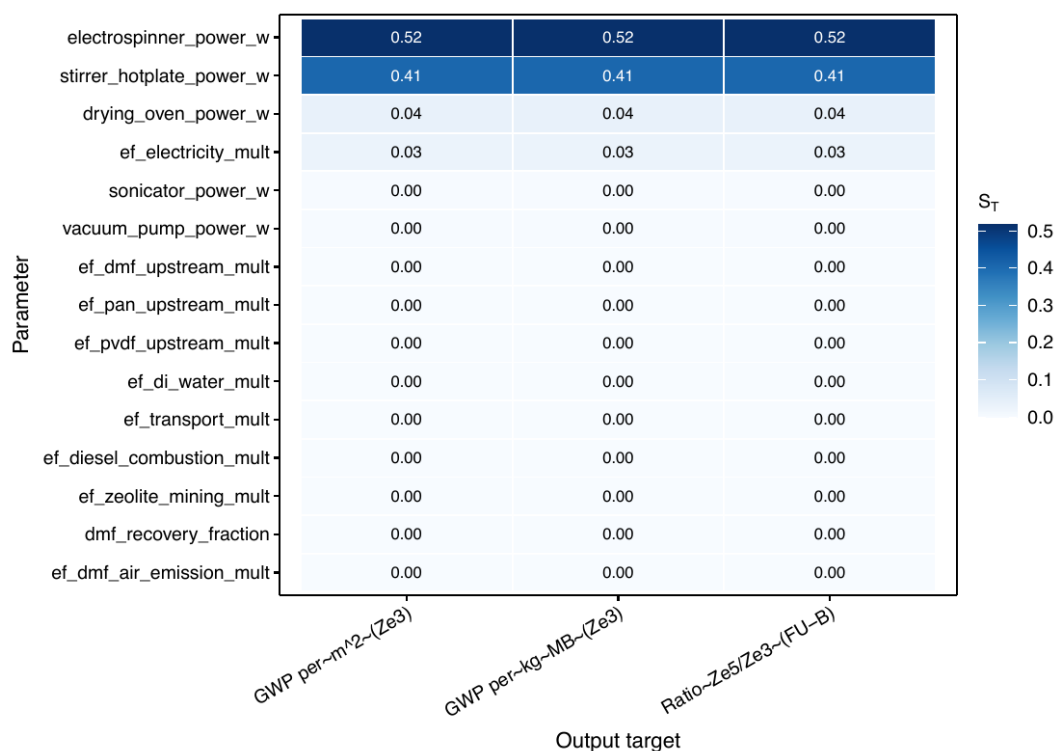


Figure 5. Sobol total-order sensitivity indices (S_T) for 15 uncertain parameters across three output targets: GWP per m² (Ze3), GWP per kg MB (Ze3), and the ratio of Ze5 to Ze3 per kg MB. Cell values are numerical S_T .

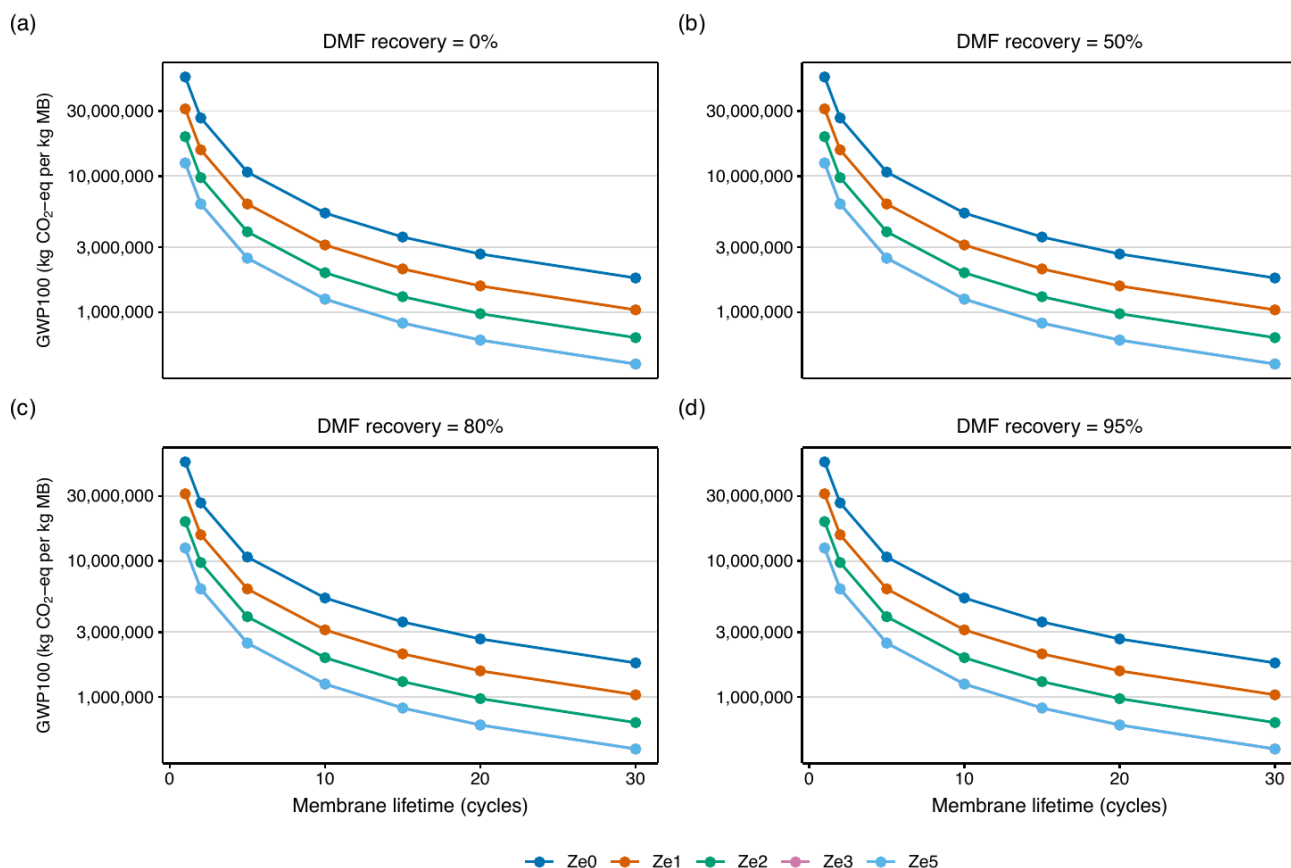


Figure 6. Impact per kg MB removed as a function of membrane lifetime (cycles) for four DMF recovery scenarios. Each line represents one of five zeolite loading scenarios. Log-scale y-axis. Lower values represent more favourable impact-per-function.

Table 1. Cradle-to-gate climate-change impact for selected polymeric membrane LCA studies. Functional units (FU) differ between studies; values are reported as published with explanatory notes. Direct per-m² comparison should be made with caution.

Membrane / system	Primary materials	Functional unit	Reported climate impact	Ref.
Ze3-PAN/PVDF electrospun (this work)	PAN, PVDF, natural zeolite	1 m ² membrane (FU-A)	1326 kg CO ₂ -eq/m ² (lab scale)	—
Ze3-PAN/PVDF electrospun (this work)	PAN, PVDF, natural zeolite	1 kg MB removed (FU-B)	2.652×10^6 kg CO ₂ -eq/kg MB	—
PVDF-HFP electrospun, membrane distillation	PVDF-HFP	per dyeing wastewater treated	Cradle-to-gate energy: 3531 MJ electrospinning + 1351 MJ pre-treatment	[10]
Polymeric hollow-fibre membrane (PVDF, PSE, CA)	PVDF / polysulfone / cellulose acetate	1000 m ² hollow fibre	PVDF route dominated by fluorspar + acetylene + HF (per-m ² value paywalled)	[5]
PAN membrane for CO ₂ sequestration	PAN	per membrane synthesis batch	123.3 kg CO ₂ -eq (PAN); 121.8 (PVIM); 142.5 (PAN-co-VIM)	[13]
Recycled PET ultrafiltration membrane (microalgal WW)	rPET (recycled bottle)	1 kg treated WW	2.94×10^{-4} kg CO ₂ -eq/kg treated water	[6]

syntheses.

When benchmarked against the *synthetic* zeolite LCA reported by Chen *et al.* 2024 [22], the natural zeolite from Tanggamus avoids the 2–4-fold higher per-kg cradle-to-gate emissions that would accompany a synthetic-zeolite alternative. The natural-mineral pathway is therefore environmentally preferable to chemical synthesis routes for the modifier component of this membrane system, even after accounting for transport from Lampung to the manufacturing site.

3.8 Implications for Material Selection and Process Design

The results yield four actionable implications for membrane practitioners and researchers. First, optimal zeolite loading depends fundamentally on the choice of functional unit: per square metre of membrane the unmodified Ze0 baseline is optimal, whereas per kilogram of MB removed the optimum shifts to Ze3. Researchers and manufacturers should therefore select the functional unit that reflects their decision context — typically FU-B for real wastewater applications, where the actual environmental service rather than the manufacturing burden is what matters.

Second, electrospinning energy emerges as the single highest-leverage intervention identified by this study. The Sobol total-order index for electrospinner power dominates the FU-A GWP variance ($S_T = 0.516$), and lab-scale electrospinning at 0.7 mL h^{−1} for 8 h per membrane is fundamentally energy-inefficient. Industrial continuous electrospinning lines can achieve order-of-magnitude lower energy per square metre, so scale-up from lab to industrial throughput is more impactful than any parameter tuning within the lab regime.

Third, natural zeolite functions as a small-footprint intervention with strong functional leverage. The additional zeolite mining and deposition chain adds only 13 % to per-m² GWP between Ze0 and Ze3, yet unlocks a 4.03× reduction in per-kg-MB burden through rejection efficiency gain. This favourable leverage supports broader adoption of natural-zeolite-modified electrospun membranes in Indonesian wastewater treatment, especially given the local availability of clinoptilolite from Tanggamus and the comparatively low burden of natural-mineral-derived zeolite relative to synthetic alternatives [22].

Finally, scale-up and lifetime assumptions must be made explicit in any membrane LCA reporting. The breakeven analysis demonstrates that operational variables (membrane

lifetime and DMF recovery fraction) span more than an order of magnitude of impact per kg MB removed — substantially larger than the spread across loading scenarios. Published LCA results should therefore always be read in the context of their assumed lifetime and solvent-fate assumptions, and our work demonstrates that the breakeven framework is a transparent tool for surfacing this dependence.

4. Conclusions

This study presents the first parametric cradle-to-gate Life Cycle Assessment of zeolite-modified PAN/PVDF electrospun composite membranes for cationic dye removal, evaluating five loading scenarios across six midpoint impact categories under dual functional units. The analysis reveals a clear but functional-unit-dependent trade-off: manufacturing burden per m² of membrane rises monotonically with zeolite loading (Ze5/Ze0 ratio of 1.13× in GWP100), yet function-level impact per kg of methylene blue removed displays a non-monotonic trajectory with an optimum at Ze3 (2.652×10^6 kg CO₂-eq/kg MB at the lab-scale, single-needle electrospinning regime modelled here). The optimum reduces impact per kg dye removed by a factor of 4.03× relative to the unmodified baseline. The absolute headline magnitudes throughout this study reflect lab-scale, single-needle electrospinning conditions and should not be read as representative of industrial-scale manufacturing; the inter-scenario *ranking* (rather than the absolute level) is the load-bearing finding.

Robustness of the optimum-loading conclusion is confirmed through three complementary analyses. Monte Carlo simulation (1000 iterations) preserves the per-m² monotonic ordering in 100.0% of iterations, and identifies Ze3 as the per-kg-MB optimum in 100.0% of cases. Sobol global sensitivity analysis attributes dominant variance to electrospinner_power_w ($S_T = 0.516$) and stirrer_hotplate_power_w ($S_T = 0.407$), with negligible parameter interactions. Breakeven analysis across 28 combinations of lifetime and DMF recovery shows Ze3 to be the lowest-impact scenario in 28/28 of cells, with the best-case scenario (lifetime 30 cycles, DMF recovery 0%) yielding 4.153×10^5 kg CO₂-eq/kg MB — a 129× span over the worst case. These findings demonstrate that natural-zeolite-modified electrospun membranes are environmentally favourable for cationic dye removal compared with the unmodified baseline, and that

lab-to-industrial scale-up of electrospinning — which currently contributes over 90% of the climate-change footprint — is the single highest-leverage intervention for future industrial deployment.

Several scope limitations should be borne in mind when applying these results. First, the assessment is framed as an internal parametric exploration of zeolite-loading variants within a single membrane technology, not as a comparative assertion under ISO 14044 §6.1; an external critical review would be required if these results are subsequently used to support a public claim against an alternative product. Second, toxicity-related categories (human toxicity and freshwater ecotoxicity) are intentionally excluded because USEtox characterisation factors for organic solvents (DMF) and natural-mineral inputs (clinoptilolite) carry substantially larger uncertainty than the climate-, air-quality-, and eutrophication-related categories quantified here; this is a transparent limitation given that DMF is a known reproductive toxicant and methylene blue itself has documented aquatic ecotoxicity. Third, the headline absolute magnitudes reflect lab-scale, single-needle electrospinning and are not representative of industrial throughput.

Future work should extend the scope to: (i) complex multi-dye mixtures beyond single-MB systems, for which experimental data are already available [40]; (ii) industrial-scale electrospinning with validated energy measurements and DMF recovery infrastructure; (iii) end-of-life modelling to convert this cradle-to-gate study into cradle-to-grave; (iv) explicit quantification of human toxicity and freshwater ecotoxicity using USEtox once primary toxicology data on industrial-scale DMF emission pathways become available; and (v) comparative assessment with commercial polymeric ultrafiltration and nanofiltration membranes for cationic-dye-rich industrial wastewater — which, if reported as a public comparative assertion, would itself require an ISO 14044 §6.1 critical review. The LCA framework developed here — Python/Brightway 2.5 foreground and background modelling, parametric Monte Carlo + Sobol GSA, and ISO 14040/14044 dual-FU reporting — is directly transferable to these extensions.

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DECLARATION OF COMPETING INTEREST

The authors declare no competing financial or personal interests that could have appeared to influence the work reported in this paper.

DATA AVAILABILITY

The complete foreground inventory tables (per scenario), background emission factors with citations, Pedigree-matrix data quality scores, Monte Carlo distributional statistics, Sobol sensitivity indices, and breakeven grid results are provided as tables in the Supporting Information. The Python and R source code used to compute the LCIA scores and produce all figures is described in the Reproducibility section of the Supporting Information.

AUTHOR CONTRIBUTIONS

Sephia Amanda Muhtar: Conceptualization, Methodology, Software, Formal analysis, Investigation, Data curation, Visualization, Writing – original draft.

Tarmizi Taher: Conceptualization, Methodology, Supervision, Project administration, Resources, Funding acquisition, Writing – review & editing.

Extended methodology and supporting data for the cradle-to-gate LCA: foreground life-cycle inventory, background emission factors, pedigree-matrix data quality scoring, Monte Carlo percentile tables, full Sobol global sensitivity indices, breakeven grid of impact per kg methylene blue removed, dye physicochemical properties, and reproducibility notes for the Python/R scripts used to compute the LCIA scores and figures.

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