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## Life cycle assessment of passive radiative cooling materials

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### ABSTRACT

Passive cooling has been a subject of research interest for decades, due to its ability to provide low-cost heat rejection. Recently, the science of passive radiative cooling materials has shown rapid progress, with the synthesis of raw materials that exhibit lower-than-ambient temperature even under direct sunlight, which however requires the use of pigments at very high-volume fractions. As the development of these materials is still in the research stage, it is important to have a deeper understanding of their environmental impact. For this purpose, in this study, a life cycle assessment is performed on different compositions of materials that have shown the potential for passive cooling under full sunshine with high emissivity and reflectance. The compositions studied are based on calcium carbonate, barium sulphate, P(VDF-HPF) and P(FA) polymers, with a life cycle assessment focused on the production of these materials. The results indicate that polymer-based material shows the highest environmental impact due to the use of PFA. In the meantime, the lower footprint is found for the calcium carbonate used as the base and water and ethanol as solvents. Overall, for the future fabrication of passive radiative cooling materials, it is suggested to use materials that do not require extensive processing and are abundant. Finally, organic solvents should be replaced by more environmentally friendly substances.

## 1. Introduction

### 1.1. Passive daytime radiative cooling materials

Cooling is one of the most prominent energy consumption sectors, accounting for 10 % of the global electricity demand (Scoccimarro et al., 2023) and has become the fastest growing energy need in the world (International Energy Agency, 2018). Passive cooling focuses on minimizing solar heat gains in order to improve indoor thermal comfort with low to zero energy consumption. In this context, the scientific community of materials has exhibited growing interest in materials that can realize passive daytime radiative cooling (PDRC), an innovative cooling technology that uses coatings that reflect and emit incoming radiation to reduce the surface temperature where applied. PDRC uses an "atmospheric window" in wavelengths between 8  $\mu\text{m}$  and 13  $\mu\text{m}$  in which most of thermal radiation can be directly dissipated to the cold outer space without getting absorbed by the atmosphere. As global temperatures are on the rise due to anthropogenic activity, passive cooling presents a

low-cost alternative that can reduce greenhouse gas pollutants and address the effects of climate change.

While passive cooling has been known for decades (Catalanotti et al., 1975; Harrison and Malton, 1978; Granqvist et al., 1981; Andretta et al., 1981; Orel et al., 1993), the cooling of the coated surface during daytime has remained an unsolved problem until recently. In most of the reported studies, titanium dioxide particles, TiO<sub>2</sub>, were used as a pigment (Buxbaum and Pfaff, 2005, p. 23–24; Mandal et al., 2020). Huang, Z. and Ruan, X. (2017) theoretically calculated passive daytime cooling with titanium dioxide-based paint using two layers of coating, a lower layer of black carbon and an upper layer of TiO<sub>2</sub> acrylic coating. Despite several attempts, prolonged passive daytime cooling with titanium dioxide as base proved challenging.

To address the UV absorption problem of TiO<sub>2</sub>, alternative methods have been studied. In one study, broadband gap materials such as ZnS and ZrO<sub>2</sub> (Nilsson and Niklasson, 1995) were considered, which however exhibit a lower reflection index. Another approach used by was the utilization of photonic coatings (Raman et al., 2014). This approach

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achieved passive daytime cooling in full sunlight for the first time, thus renewing the interest of the science community. However, photonic coatings proved quite complex to fabricate and therefore imposed a barrier for large-scale applications. A recent review study (Liu et al., 2025) has explored the band gap and the refractive index of typical inorganic materials.

Recent studies have considered a wide range of approaches to achieve daytime cooling. In a recent work by Cui et al. (2022) a P(VDF-HFP) based coating demonstrated a 9.0 °C. sub-ambient temperature drop during daytime. The authors of another study investigated the possibility of integrating Hollow Yttrium-Oxide Spheres (HYS) into a Polydimethylsiloxane (PDMS) matrix to improve Radiative Cooling and showcased a surface temperature differential of 8.3 °C lower than the non-HYS counterparts (Yu et al., 2024). A largely studied possibility is realizing PRDC fabrications using SiO<sub>2</sub> spheres with different binders such as PDMS (Lee et al., 2024; Park et al., 2023), cellulose acetate (Xiang et al., 2021) and polyurethane dispersions (Yu et al., 2025). These studies highlighted the potential of SiO<sub>2</sub> based syntheses in passive cooling by achieving temperature reductions up to 7.6 °C, 9.7 °C, 6.2 °C and 7 °C correspondingly. In another work, a thermoelectric generator was assembled that utilized a radiative cooling film based on a PDMS, SiO<sub>2</sub> spheres and thermochromic pigments with the potential to achieve up to 63.2 W/m<sup>2</sup> of energy savings during summer (Park et al., 2023).

The creation of PDRC coatings that are single-layer and ultra white, is a challenging issue. The studies discussed in this paper consider four different structures to achieve PDRC. Specifically, the first structure used calcium carbonate as the main component, the second and the third structures were synthesized using barium sulphate, while the fourth utilized different polymer particles and glass beads (Li et al., 2020, 2021; Gong et al., 2023). In detail, the development of the first paint was innovative in using a wide particle size distribution instead of a specific particle size, achieving a wider spectrum in which the radiation was scattered in accordance with the theoretical considerations of J. Peoples et al. (2019). CaCO<sub>3</sub> has a relatively low reflectivity index (Ghosh, 1999) and therefore, a high particle concentration of 60 % was used. This composition was exposed to direct sunlight and demonstrated an emissivity index of 0.94 in the atmospheric window and a reflectance of 95.5 % in the solar spectrum.

Correspondingly, the second composition follows a similar pattern to the previous one, with a broad particle size distribution of BaSO<sub>4</sub> particles instead of CaCO<sub>3</sub> and a particle concentration of 60 %. This synthesis achieved an emissivity of 0.95 in the atmospheric window and a reflectance of 98.8 % in the solar spectrum. In the third composition, a membrane with BaSO<sub>4</sub> particles was implemented and showcased an emission coefficient of 0.96 and a reflectivity of 97.6 %. Finally, the fourth composition consists of two thin layers, one layer of polymers with a random distribution of SiO<sub>2</sub> beads of 6 % concentration and a silver layer to enhance its reflectivity in the solar spectrum. This synthesis presented an emission coefficient <0.93 in the spectrum of interest and a reflectivity of 96 %. The thicknesses of the four compositions are 400 µm for the CaCO<sub>3</sub> paint, 400 µm for the BaSO<sub>4</sub> paint, 150 µm for the BaSO<sub>4</sub> film and 50 µm for the polymer layer. It is emphasized that passive daytime cooling was achieved by all four of the studies.

Life cycle assessment (LCA) is a comprehensive methodology for assessing the environmental sustainability of products or systems. It provides detailed environmental assessments for policy makers, the research community and decision makers. The LCA methodology has been applied a few times to passive cooling materials. In their study, Tang et al. (2022) evaluated the life cycle of a paint with daytime passive cooling capability based on Magnesium Oxide (MgO) using a cradle-to-grave approach. Although the proposed material showed some drawbacks in terms of terrestrial acidification and water eutrophication, in general the overall impact of the material on the midpoint categories was observed to be low. In another study, Pirvaram et al. (2022) presented methods that can be followed to use life cycle assessment to

calculate the global warming potential (GWP) of various passive cooling structures. The authors of a recent study (Choi et al., 2025) applied Life Cycle Assessment in their research to evaluate the footprint of the passive material they fabricated, a TiO<sub>2</sub>-based acrylic paint and confirmed a ~73 % reduction in CO<sub>2</sub> emissions in the production process when compared to a solvent-based counterpart.

## 1.2. Goal and scope

In general, there is a limited number of studies addressing the life cycle of radiative cooling coatings. The authors of a recent study emphasized the importance of life cycle assessment in ensuring real-world viability of RC technologies (Yang et al., 2025). The implementation of the LCA methodology on newly formed PDRC technologies can offer the opportunity to minimize the environmental footprint of these innovative materials, optimize material selection and fabrication methods, and achieve true net reduction in emissions and impacts. Given the importance of this methodology in PDRC materials, the aim of this study is to compare the environmental impacts of innovative passive daytime radiative cooling materials by conducting a life cycle analysis and carry out concrete proposals for future research in the domain of radiative cooling materials. The four formulations mentioned above were considered and a comparative assessment was developed. By evaluating the environmental footprint of these materials from the extraction of the raw materials to the point of production, this analysis aims to determine the most sustainable option for passive cooling applications. Various impact indicators were examined such as human health, natural resource depletion and toxicity in order to highlight the environmentally friendly solutions in the field of building energy efficiency and draw general indications for the production of more sustainable daytime radiative cooling coatings.

## 2. Materials and methods

The primary objective of the Life Cycle Assessment is to assign a numerical value to the energy consumption and environmental impacts associated with different processes or activities throughout the life cycle of a product. The LCA method can assess all or specific stages of the life cycle of a product, depending on the way it is carried out. The three main types of LCA assessments include:

1. Cradle-to-gate approach: This method focuses on evaluating the life cycle of the product up to the point that the manufacturing process is completed. It is often chosen when data is limited and provides important environmental guidance when developing new technologies and materials (Cao, 2017).
2. Cradle-to-grave approach: This approach considers the entire life cycle of a product, from production to the end of its life. This approach analyses the product's footprint in depth, offering a broader picture of its long-term impacts, but requires a lot of data that may not always be available or disclosed in the literature (Ntours et al., 2021).
3. Cradle-to-cradle approach: This approach studies the case where the disposal of the materials under study is replaced at the end of their life by recycling. Note that this approach is often applied in the context of the circular economy (Tamoor et al., 2022).

The LCA study of this paper adopts a cradle-to-gate approach, focusing on the production of passive cooling materials. The life cycle impact assessment (LCIA) method ReCiPe 2016 v1.01 was used to conduct the analysis. The database utilized in this method is obtained from the Ecoinvent database (Huijbregts et al., 2017; Finkbeiner, 2014).

ReCiPe 2016 includes three types of perspectives to group different sources of uncertainty and different choices, listed below:

1. The individualistic perspective relies on immediate interests, clearly defined impact types and a belief in technological optimism.
2. The hierarchist perspective is based on scientific consensus concerning timelines and feasibility of impact mechanisms.
3. The egalitarian perspective adopts the most cautious approach, considering the longest timelines and all available impact pathways for which there is data.

In this study, the hierarchist perspective is utilized in order to take into account both the short-term and the long-term viewpoint of assessed impacts. In terms of weighting, there are two choices, ReCiPe H/H and ReCiPe H/A. A unique set of weighting is provided for each perspective in H/H, while H/A encompasses the average outcome for all three perspectives. In this study, H/A weighting is used. For the comparative visualizations in Figs. 7–9, characterization results for each midpoint and endpoint category were normalized using the global average normalization factors provided in the ReCiPe 2016 (H/A) method implemented in SimaPro. These factors represent the total annual environmental load per impact category per capita on a global scale, allowing the results to be expressed in dimensionless form and enabling direct comparison across categories and formulations. No additional regional adjustments or system-level scaling beyond the default global ReCiPe dataset were applied.

The purpose of the ReCiPe methodology is to quantify the data into indicators that reflect environmental impacts in different categories. This is achieved at two different levels, namely at the 'midpoint' level in which 17 indicators are calculated and on 'endpoint' level, where these 17 indicators are grouped in 3 endpoint indicators are calculated.

The endpoint indicators sum up the individual midpoint indicators to assess the results in the three important categories of human health, ecosystems and resources. Such a generalization on the one hand serves to provide better comparisons of results but does not reveal the multifactorial effects of the materials under consideration in the midpoint subcategories listed above (Ismael, 2018). Results and conclusions are thus extracted at both levels to allow for both total and partial comparison of the results by endpoint category and midpoint subcategory, respectively.

The data used in the life cycle assessment are provided in units of kilograms (kg) and watt-hours (Wh). Some of the materials considered were found in the SimaPro software libraries. For the remaining materials, the life cycle was estimated using the software's synthesis function. Finally, it is mentioned that the LCA study was implemented using the SimaPro software, version 9.5.0.

### 3. Experimental

For each composition, the required proportions of all substances are calculated for the preparation of 1 kg of product. Some compositions lack some specific details regarding these proportions or the use of instrumentation, so certain assumptions are imposed. The electricity consumed is assumed to come from the Greek infrastructure, which includes fossil fuels such as oil, coal and natural gas and also renewables such as solar and wind energy generators.

The fabrication process for synthesis 1, the CaCO<sub>3</sub> paint, is adopted from Li et al. (2020), for synthesis 2 and 3, BaSO<sub>4</sub>, the paint and film, are adopted from Li et al. (2021) and for synthesis 4, the polymer film, is adopted from Gong et al. (2023). The materials, processes and assumptions for each synthesis are analytically listed in appendix A. The estimated inputs for each synthesis are presented in Table 1.

**Table A1**  
Input data for synthesis 1.

Synthesis	Category	Quantity
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(continued on next column)

**Table A1** (continued)

Synthesis	Category	Quantity
Synthesis 1	Calcium Carbonate	357g
	Dimethylformamide	580g
	Methyl Methacrylate	63g
	Low voltage electricity	211Wh
Synthesis 2	Barium sulphate	357g
	Dimethylformamide	580g
	Methyl Methacrylate	63g
	Low voltage electricity	253Wh
Synthesis 3	Barium sulphate	500g
	Deionized water	250g
	Ethanol	250g
	Low voltage electricity	18Wh
Synthesis 4	Acetylene glycol ethoxy	14,08g
	P(VdF-HFP)	84,50g
	Water	478,87g
	PFA	169,01g
	Acrylic acid	126,76g
	Glass	70,42g
	TPNB	28,17g
	Silica modified polydimethylsiloxane	28,17g
	Low voltage electricity	405Wh

## 4. Results

### 4.1. Environmental analysis of different syntheses

For the environmental evaluation of 1 kg of each passive cooling material, the LCA methodology was carried out using the SimaPro software and specifically ReCiPe Endpoint (H) V1.13/World ReCiPe H/A was applied to extract the following graphs. A characterization graph is presented for each composition, showcasing the normalized comparative results of individual materials and processes by midpoint impact category.

In Fig. 1, it is evident that in synthesis 1, dimethylformamide has the largest impact with an impact of 44 up to 88 % of the overall impact of the synthesis in each category. This is followed by calcium carbonate particles with an impact rate from 8 to 33 % and electricity consumed with a rate up to 41 %. Finally, methyl methacrylate exerts the lowest impact with a rate up to 16 %. More specifically, dimethylformamide has a greater impact in the categories "Ozone depletion" (88 %) and "Natural land transformation" (86 %). This is because it has high levels of volatile substances that enhance the formation of tropospheric ozone, thus destroying stratospheric ozone and is a harmful air pollutant and a key component of smog (Kim et al., 2015). After that, calcium carbonate particles show an increased effect in the categories "Agricultural land occupation" (33 %) and "Climate change Human Health" and "Climate change ecosystems" with 20 % each. Calcium carbonate itself has a low impact on climate change. Although its industrial production, particularly in the manufacture of cement, releases significant amounts of carbon dioxide contributing to global warming indirectly (Hanein et al., 2021), there are natural sources such as eggshells and oyster shells that have even been used in literature to manufacture PRDC paints, exhibiting temperature reduction of at least 4.1 °C and emissivity of at least 95 % and within the realm of practical feasibility and the potential for commercial viability, while demonstrating robust mechanical performance. (Lee et al., 2025; Wu et al., 2023). It must be noted that the manufacturing procedure of these syntheses complex in terms of required materials and processes. The electricity consumption process affects mostly the categories "Freshwater eutrophication" (41 %), "Marine ecotoxicity" (32 %) and "Freshwater ecotoxicity" (31 %). Finally, acrylate has the highest impact in the category "Photochemical oxidant formation" (21 %). Methyl methacrylate contributes to the formation of photochemical oxidants, mainly through the release of volatile organic compounds (VOCs) during its production and use. These VOCs may react with nitrogen oxides (NO<sub>x</sub>) in the presence of sunlight to form

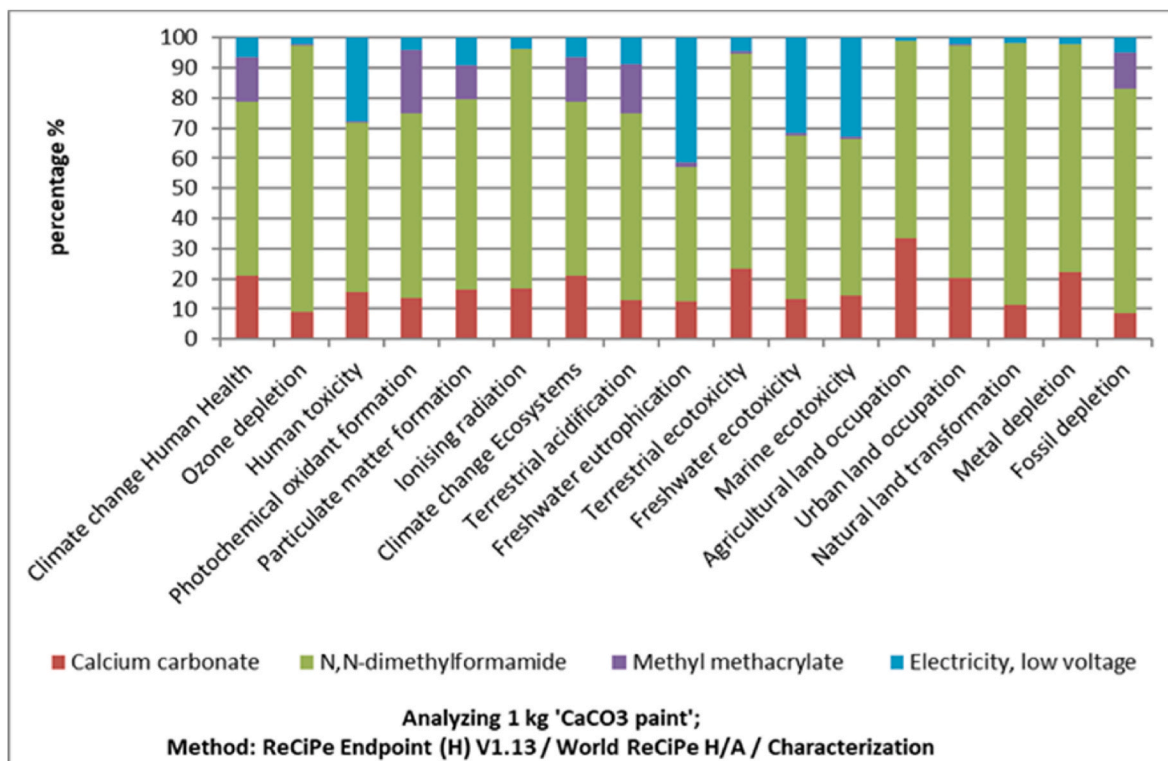


Fig. 1. Comparative results for synthesis 1, CaCO3 paint.

ground-level ozone, a key component of photochemical smog, which is harmful to human health and the environment (Department of Climate Change, 2022).

In Fig. 2, it is demonstrated that in composition 2, barium sulphate particles have the greatest influence across most categories, with an

impact up to 94 %. This is followed by dimethylformamide with an impact rate as high as 83 % and the electricity consumed with a rate up to 37 %. Finally, the acrylic base (methyl methacrylate) has the lowest impact with a maximum rate of 17 %. More precisely, barium sulphate particles show an increased impact in the categories "Human toxicity"

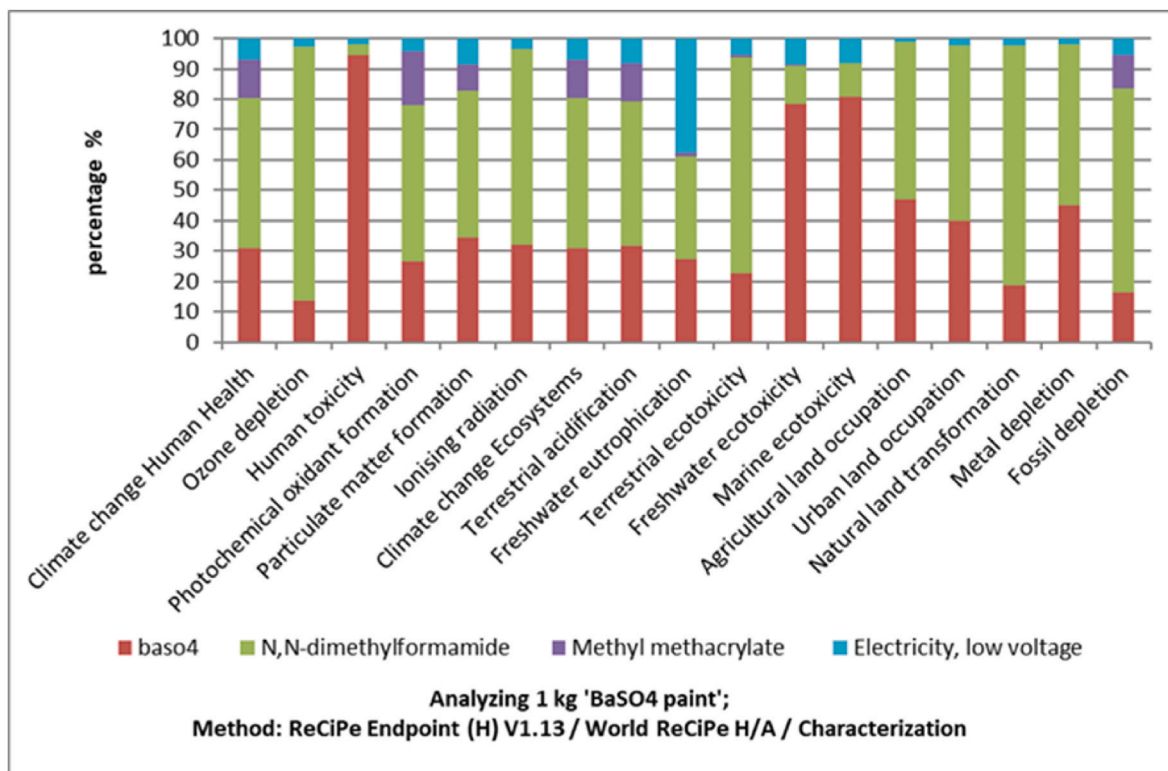


Fig. 2. Comparative results for synthesis 2, BaSO4 paint.

(94 %) and "Marine ecotoxicity" (80 %), that can be attributed to its manufacturing process. The mining and processing of BaSO<sub>4</sub> can lead to air and water pollution, releasing harmful by-products or particulates. It is generally inert and has low solubility, showing little direct marine ecotoxicity. However, there may be impacts on the sea from the by-products of its extraction and processing, which could contaminate water masses. Improper disposal of BaSO<sub>4</sub> waste and the involvement of more toxic chemicals in its production and use may also contribute to marine ecotoxicity, affecting aquatic life. Following this, dimethylformamide has a greater impact in the categories "Ozone depletion" (83 %) and "Natural land transformation" (79 %). This is due to the fact that it contains high levels of volatiles that are harmful air pollutants and a key component of smog, as mentioned in Synthesis 1 (Environmental Protection Agency of the U.S. Government, 2016). The electricity consumption process also affects this composition most significantly in the categories "Freshwater eutrophication" (37 %), "Marine ecotoxicity" (8 %) and "Freshwater ecotoxicity" (8 %). Finally, acrylate has the highest percentage in the category "Photochemical oxidant formation" (17 %). Methyl methacrylate, as reported in Composition 1, contributes to photochemical oxidant formation, mainly through the release of volatile organic compounds (VOCs) during its production and use.

In Fig. 3, it is evident that in composition 3, barium sulphate nanoparticles significantly influence a broad spectrum of categories, with an impact rate as high as 99 %. This is followed by ethanol, with the same maximum rate of 99 %, but unlike barium sulphate particles it typically does not exceed 50 %. Subsequently, the electricity usage reflects a significantly reduced percentage ranging from 0.1 to 5 %, while the least effect is attributed to deionized water, showing a negligible rate hardly exceeding 0.1 % across any of the categories. More particularly, barium sulphate nanoparticles show an increased impact in the categories "Human toxicity" (99 %) and "Marine ecotoxicity" (97 %), in accordance with the results shown in the previous synthesis with barium sulphate. Second to that, ethanol has the greatest impact in the categories "Terrestrial ecotoxicity" (99 %) and "Natural land transformation" (96 %). Ethanol can affect soil ecotoxicity through the processes involved in

its production, use and disposal. Agricultural practices used to produce ethanol feedstocks can lead to soil and water contamination due to the high use of agrochemicals they require. These chemicals can leak into the soil and the surrounding environment, damaging terrestrial ecosystems. In addition, during the production, transport and use of ethanol, accidental spills can introduce ethanol and related chemicals into the environment, potentially damaging soil health and biodiversity. Similarly, by-products and wastes generated during ethanol production, if not properly managed, can lead to soil contamination (Falano et al., 2014). The electricity consumption process also in this composition affects most the category "Freshwater eutrophication" with a percentage of about 5 %, while in the others the percentage of impact is less than 1 %. Finally, deionized water, which is used as a solvent in this synthesis, has the lowest impact, with an average impact of only 0.01 %.

Finally, Fig. 4 demonstrates that in Synthesis 4, PFA exerts the most significant influence, with a maximum impact rate of 99 %. This is followed by the electricity consumption process with a rate up to 45 %, silica modified polydimethylsiloxane with a maximum rate of 21 % and acrylic acid, with a rate up to 13 %. Lower levels are mainly found in flat glass, (P(VDF-HFP)) and TPNB (typically below 10 %). Water and acetylene glycol ethoxy showcase the lowest impact, showcasing rates below 1 %. In more depth, PFA exhibits an increased impact in the categories "ozone depletion" (99 %), "Climate change Human Health" (94 %) and "Climate change ecosystems" (94 %). PFAs pose significant risks to human health, mainly due to its classification as a potential carcinogen. Exposure to PFA can lead to a variety of negative health effects, including reproductive problems, liver damage, kidney diseases and an increased risk of cancer (Fenton et al., 2021). Furthermore, PFA can have harmful effects on ecosystems, especially if released into the environment during its production, use or disposal. In water, it can be toxic to aquatic organisms and ecosystems (Ohoro et al., 2024). In addition, it is persistent in the environment, meaning that it can persist in ecosystems for prolonged periods of time, potentially causing long-term ecological damage. Their environmental and health risks have invoked an EU proposal to ban PFAs (Sonne et al., 2023). The electricity

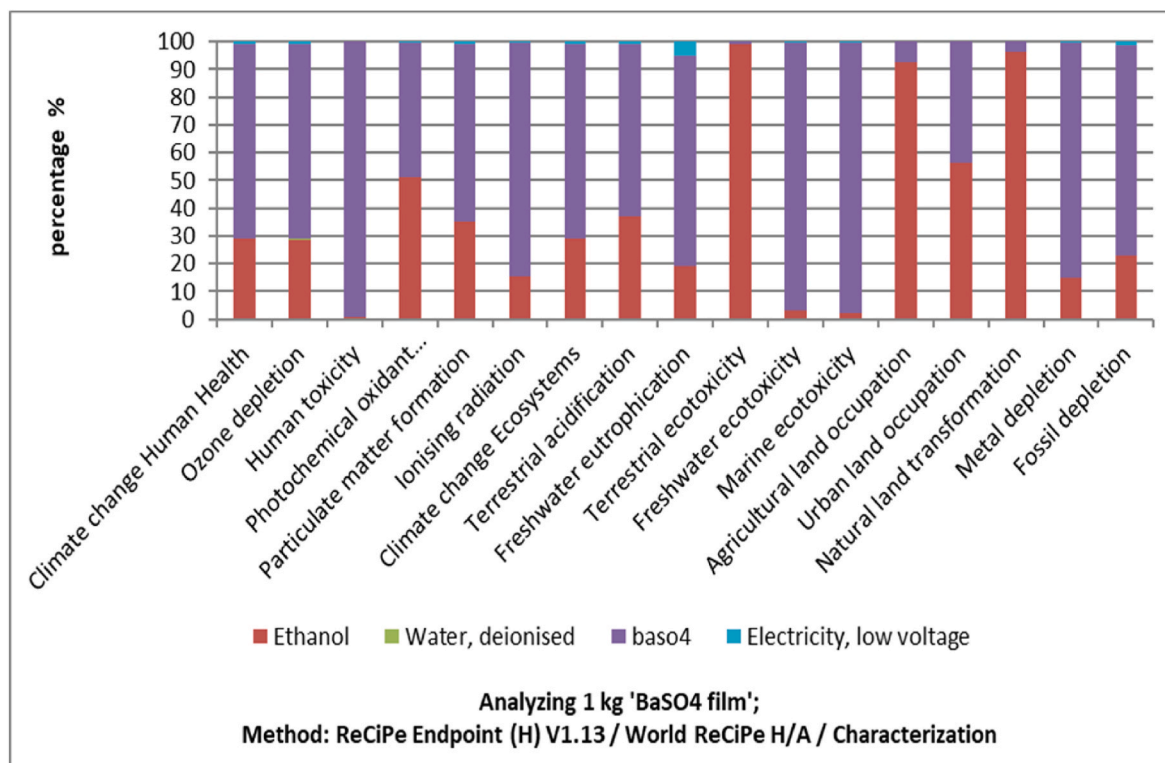


Fig. 3. Comparative results for synthesis 3, BaSO<sub>4</sub> film.

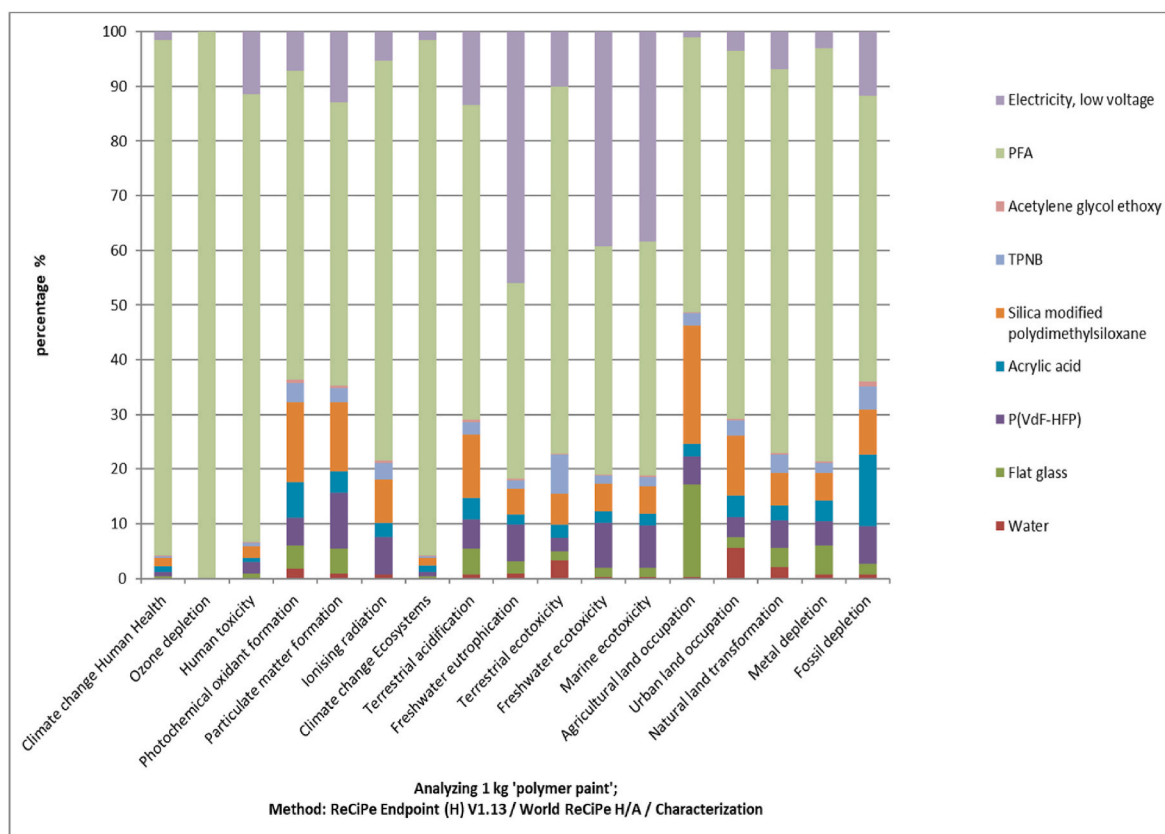


Fig. 4. Comparative results for synthesis 4, polymer film.

consumption process also has the highest impact in this composition in the categories 'Freshwater eutrophication' (45 %), 'Marine ecotoxicity' (39 %) and 'Freshwater ecotoxicity' (38 %). Further, silica modified polydimethylsiloxane has a greater effect in the categories 'Agricultural land occupation' (21 %) and 'Photochemical oxidant formation', (14 %) mainly through the release of volatile organic compounds during its production, usage, or degradation. Acrylic acid has a greater impact in "Fossil depletion" (13 %), since it comes mainly from petrochemical sources, such as propylene obtained from crude oil or natural gas. The production of acrylic acid involves energy-intensive processes that rely heavily on fossil fuels, both as raw materials and as energy sources. This reliance contributes to the depletion of fossil fuel reserves (Sun et al., 2017). Glass shows an increased impact compared to the other categories in "Agricultural land occupation" (16 %), while P(VdF-HFP) stands out in "Particulate matter formation" at 10 %. The manufacture of PVDF involves high temperature processes and the use of various chemicals, which can release nanoparticles and VOCs into the air. These emissions can lead to the formation of particulate matter (PM), a harmful air pollutant that can affect respiratory health and contribute to environmental issues such as smog. TPNB showcases the highest impact rate in the category "Terrestrial ecotoxicity" (7 %), due to its chemical properties and its potential to contaminate the environment. If released into the environment through spills, improper disposal or run-off, this solvent can contaminate soil and adversely affect terrestrial organisms. Its persistence and its potential for bioaccumulation may further exacerbate its effects on terrestrial ecosystems, leading to long-term ecological damage. Finally, the impact on the environment due to water and acetylene glycol ethoxy is considered negligible since it does not exceed 1 % in any category.

#### 4.2. Comparison of the syntheses with BaSO<sub>4</sub> particles as the main component (synthesis 2, synthesis 3)

In this section, a comparison of two different passive cooling material synthesis methods involving BaSO<sub>4</sub> paint and film respectively, is made to evaluate their comparative environmental impact. The results obtained from the analysis are presented in the following graphs.

In Fig. 5, both formulations are projected to have a significantly higher environmental impact in the category "Human health", 72 % of its overall impact for Synthesis 2 and 80 % for Synthesis 3. This result was expected since barium sulphate has an impact on health in an indirect way, as mentioned in the previous section. It is notable that the film has a 10 % higher impact when compared to the paint in this category. In "Resources", the paint appears to be more harmful, with an impact score 3 times higher than the film. Finally, in 'Ecosystems', both formulations exhibit similarly low impact rates of around 2 % of the overall. Overall, it can be observed that both compositional pathways have the same overall impact, with the barium sulphate paint having a rate 10 % higher than the film. This is because the paint utilizes DMF as a solvent, which, unlike water and ethanol used by the film, is particularly harmful to the environment due to its nature as a VOC. Fig. 6 confirms that the paint tends to showcase slightly larger environmental footprints throughout most midpoint categories, with the exceptions of toxicity related categories (human toxicity- 1.3 higher, terrestrial ecotoxicity- 30 times higher, freshwater ecotoxicity- 1.1 times higher, marine ecotoxicity-1.2 times higher) and land related categories (agricultural land occupation- 9 times higher, urban land occupation- 1.3 times higher, natural land transformation- 7 times higher), due to the usage of ethanol (see Fig. 6).

#### 4.3. Overall comparison of the syntheses

In this section an overall comparison is conducted between the four syntheses, using the SimaPro software. Significant differences between

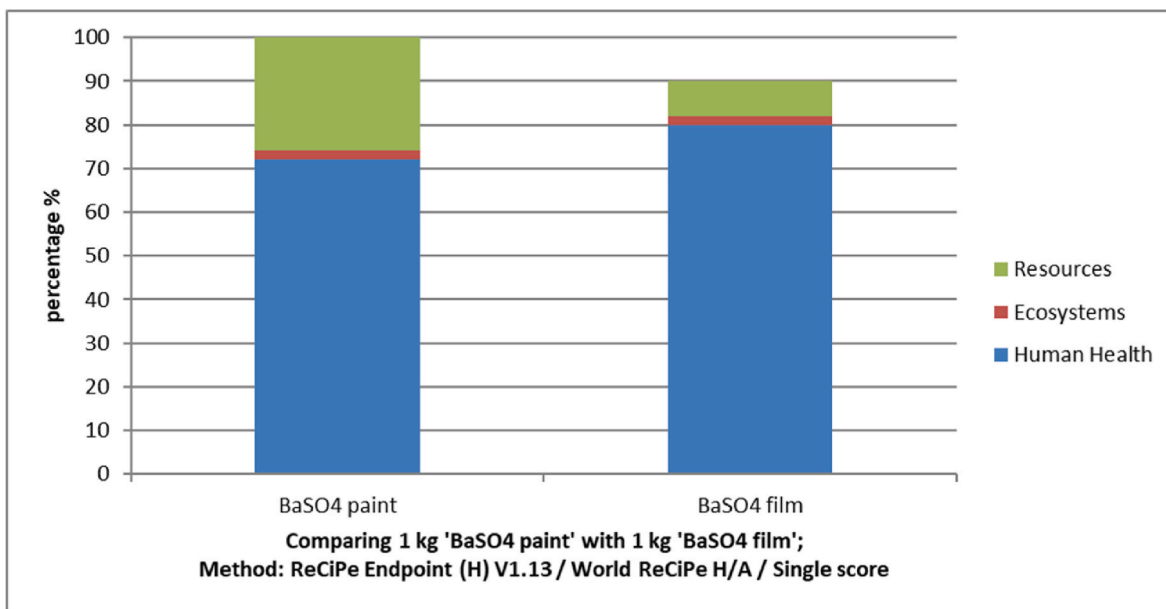


Fig. 5. Normalized comparison of the impact rates of the two BaSO4 syntheses per endpoint category.

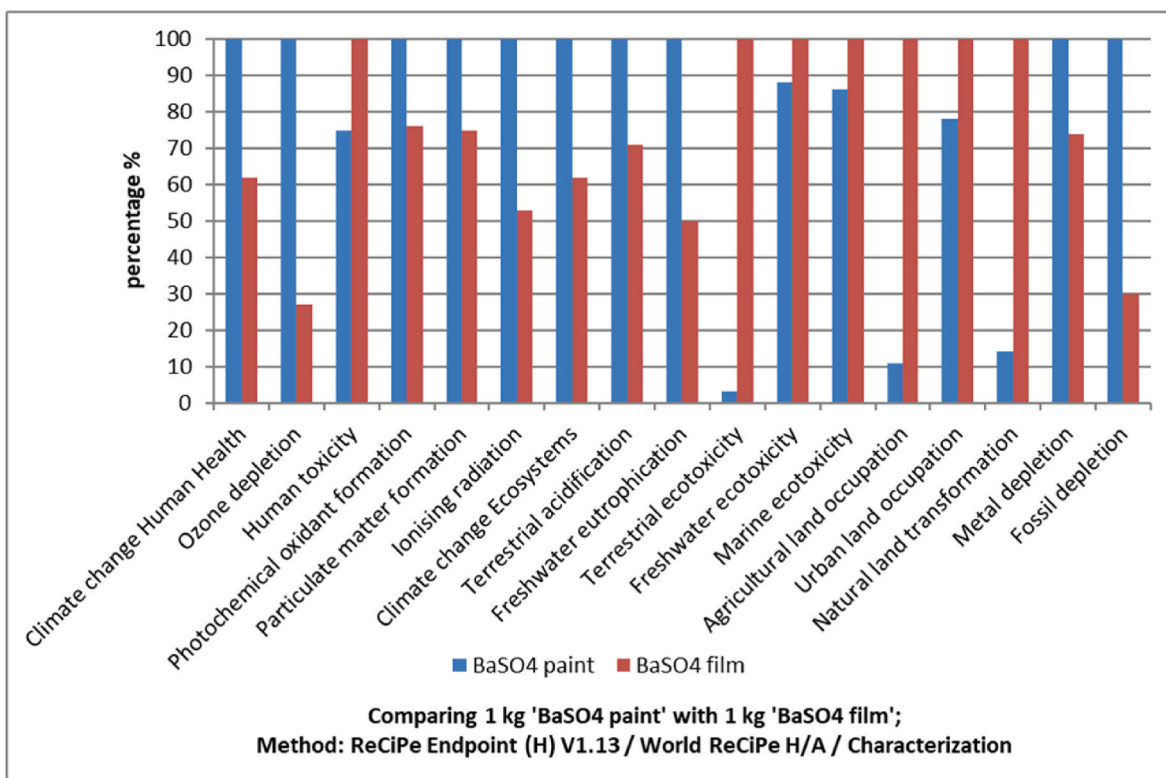


Fig. 6. Normalized comparison of the impact rates of the BaSO4 syntheses per midpoint category.

the four passive daytime radiative cooling materials can be identified in the following figures, which can be attributed primarily to the types and quantities of solvents used in the formulations and to the subsequent manufacturing process.

In Fig. 7 it can be noted that the formulation with the lowest environmental footprint is synthesis 1, CaCO3 paint, which reduces the impact by a factor of 75 % from the most harmful one in the study. This is followed by the two different compositions using BaSO4 with composition 3, the film, performing slightly better (55 % reduction),

since it uses deionized water instead of dimethylformamide contained in composition 2 (50 % reduction). The most harmful composition is Composition 4, which requires a large number of materials and processes for its preparation. It is further noted that all four formulations possess the highest percentage of harmful impacts in the Human Health category, followed by the Resources category with a considerable margin in between. Ecosystems appear to be the least affected category.

Fig. 8 identifies the environmental impacts of the compositions weighted by impact category. Composition 1 requires lesser amounts of

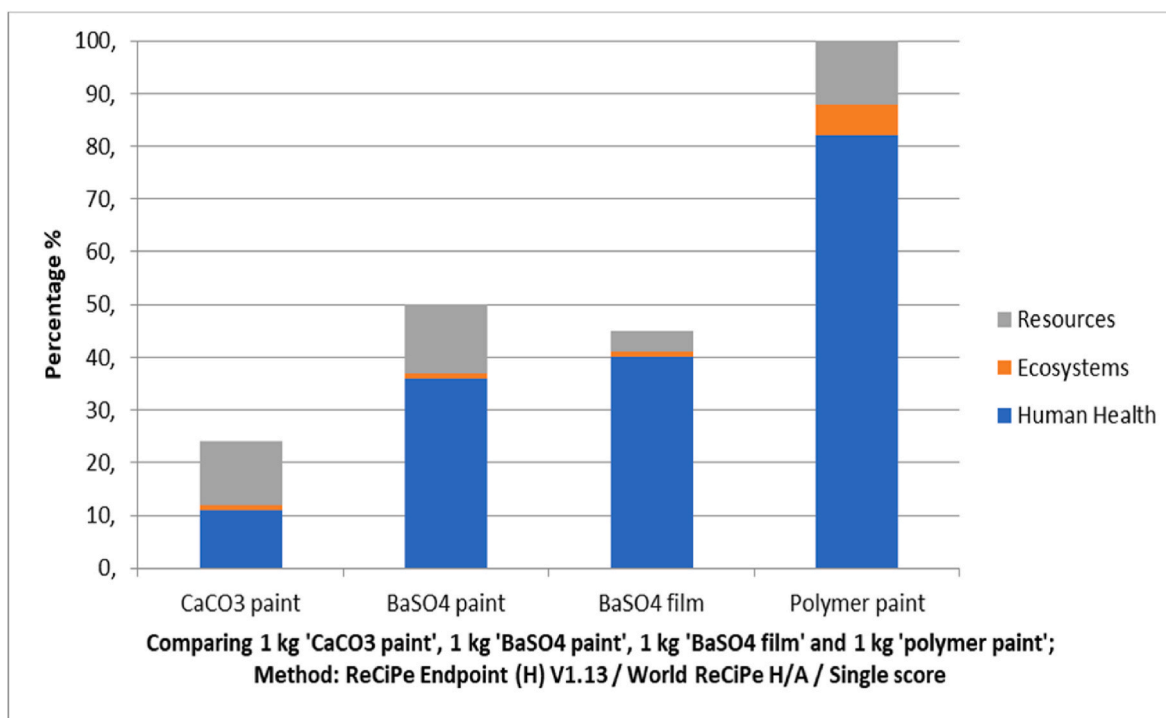


Fig. 7. Normalized comparison of the impact rates per synthesis.

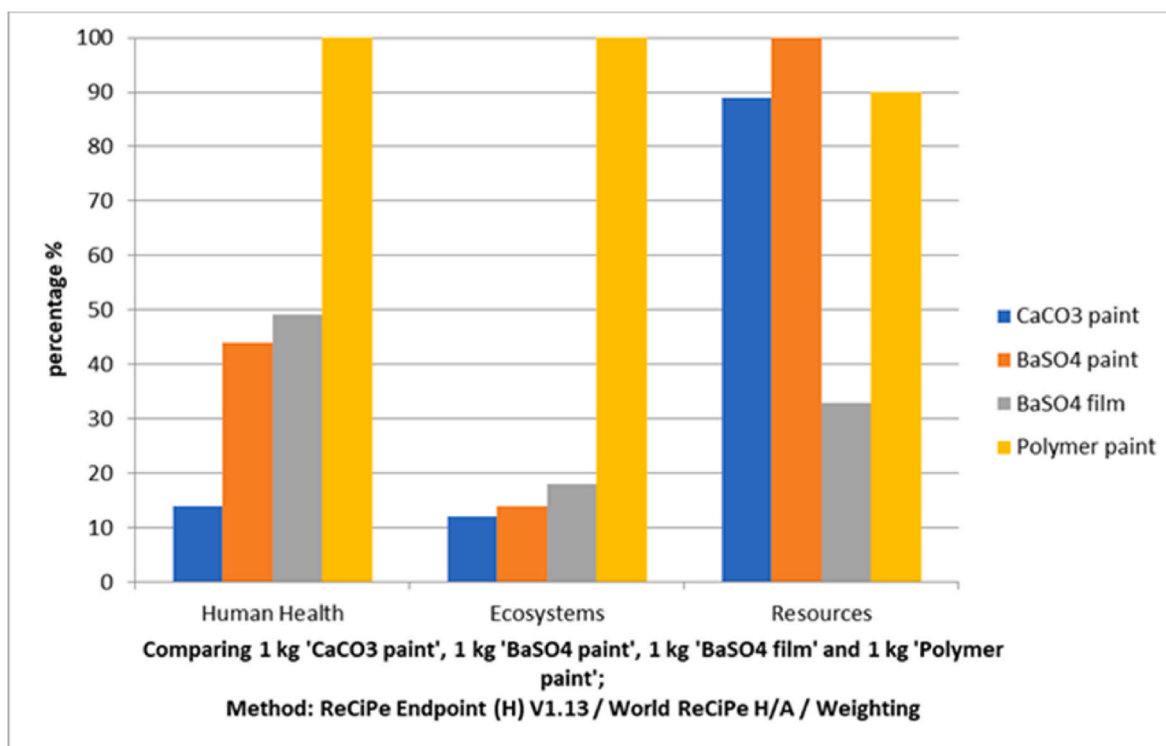


Fig. 8. Normalized comparison of the impact rates of each synthesis per endpoint category.

compounds compared to composition 4. This achieves an 88 % reduction in impact on ecosystems and 86 % on human health. Overall, significant reductions have been achieved in all 17 impact categories, as shown in Fig. 9. Subsequently, composition 3 shows a 51 % reduction in impact on human health, 82 % on ecosystems and 57 % on resources. In composition 2, it is noteworthy that although a 56 % reduction in impact

on human health and 86 % on ecosystems is identified, in the resources category it appears to be the most damaging with a 10 % reduction in impact compared to Composition 4. This can be attributed to the fact that barium sulphate affects resources mainly through its extraction and processing.

Fig. 9 shows that although Synthesis 3 is the third most harmful of

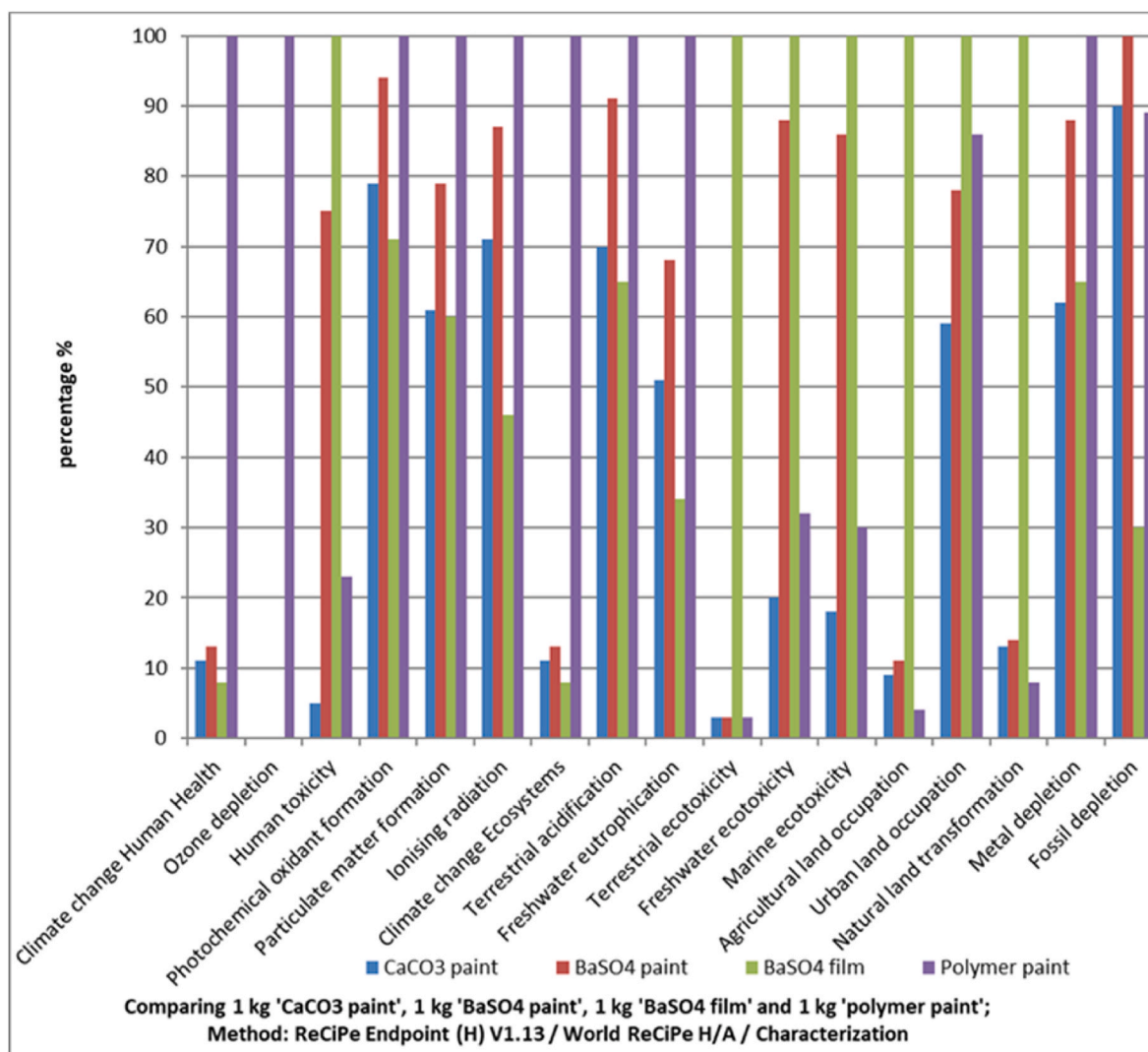


Fig. 9. Normalized comparison of the impact rates of each synthesis per midpoint category.

the four, it shows a significantly greater impact than the others in the categories of 'Terrestrial ecotoxicity', 'Agricultural land occupation' and 'Natural land transformation'. This is because this formulation requires ethanol for its fabrication, which contributes to these categories through agricultural and industrial processes, monoculture practices and deforestation.

Overall, the environmental impact of PDRC materials depends both on the energy consumed in the synthesis process, since it shows contributions to the categories of impacts, in terms of climate change, freshwater and marine eutrophication and land use and on amount and type of the chemical solvents used as well. It is therefore confirmed that Composition 4 has the highest footprint due to the increased amount of electricity required in the usage of a large amount of PFA.

## 5. Discussion

This study provides a comprehensive life cycle assessment analysis of four innovative PRDC materials, each using different compounds and processes, in order to evaluate their comparative environmental impact. The findings show that Composition 1, the CaCO<sub>3</sub> paint emerges as the most environmentally friendly option among the four, mainly due to the intrinsic properties of CaCO<sub>3</sub> itself. This compound is abundant, non-toxic and requires less processing compared to the other materials. In contrast, BaSO<sub>4</sub> also used in passive cooling applications, has a larger

environmental footprint and has been classified since 2017 as a critical raw material from the European Commission (ECC, 2017). The mining and processing of BaSO<sub>4</sub> can lead to higher emissions of hazardous by-products and particulates, posing greater risks not only to human health but also to the environment. These factors make CaCO<sub>3</sub> a more viable option for passive cooling products aligned with the goals of reducing ecological impacts and enhancing environmental sustainability. On the other hand, the fourth formulation, which incorporates a variety of polymers, has the highest environmental impact, mostly due to its extensive use of PFA, a fluorinated compound associated with significant environmental and health concerns. In addition, by using less energy, the CaCO<sub>3</sub> based product minimizes greenhouse gas emissions, the formation of fine particulate matter and other related pollutants.

Overall, the results for CaCO<sub>3</sub> are in accordance with literature. A recent study showcased that the impact of a CaCO<sub>3</sub> paint on the climate was always positive across all climate zones (Barber et al., 2025). However, a limitation that could prevent the widespread commercial adaptation of CaCO<sub>3</sub> in PRDC technologies is its low chemical resistance to acids, when compared to BaSO<sub>4</sub> and SiO<sub>2</sub>. Calcium carbonate-based RC coatings could be damaged when exposed to acid rain, resulting in a shorter lifespan and potentially an increase in the total life cycle environmental load. In order to weigh more precisely the long-term implications of chemical resistance in the formulations, it is highly recommended that a cradle to grave LCA analysis is conducted when

data regarding the lifespans of these formulations are available.

Regarding the solvents, DMF, used in the first and second formulations, is classified as a volatile organic compound that contributes to ground-level ozone formation and photochemical smog. Ethanol, used in the third formulation, which is classified as the second most environmentally friendly formulation, although less toxic than DMF, also has its drawbacks, particularly in its role in contributing to soil ecotoxicity and the potential for eutrophication of water bodies. In composition 3, water is also used as a solvent and stands out as the best choice due to its non-toxic nature, its abundance and its overall minimal environmental impact. The use of water as a solvent can significantly reduce the ecological footprint of passive cooling products. A recently published study examined the performance of BaSO<sub>4</sub> and CaCO<sub>3</sub> PDRC paints using both water and DMF as solvents and concluded that the performances of the materials were similar (Joseph et al., 2023). Thus, water could be used as an alternative solvent in future similar syntheses, with a scope of reducing the environmental footprint. However, DMF is still more suitable for exterior paints due to its higher resistance to extreme weather conditions and water-based solvents face are less effective in providing long-term protection against corrosion. In addition, water may undergo phase separation, resulting further damage of the formulation. Although there is considerable focus within the scientific community on replacing solvents with water, these problems still pose significant drawbacks to the large scale in the large-scale replacement of DMF by water as a solvent.

The present analysis uses the Greek electricity mix to reflect the geographical context of the source experiments, which combines fossil-based sources (oil, coal, natural gas) and renewables (solar, wind). While this choice may influence absolute values in impact categories such as climate change, eutrophication, and ecotoxicity, the relative ranking of the four formulations is not expected to change substantially because electricity-related impacts were generally secondary compared to those from solvents and raw material production. Future work should include a sensitivity analysis using alternative electricity mixes, such as the EU-27 or global average, to quantify the potential variation in results due to the energy source profile.

It is important to stress the limitations of current research. The availability of detailed environmental data for some of the materials and processes remains limited, which may affect the completeness and accuracy of the assessment. Future research should focus on collecting more comprehensive data, in particular on the long-term environmental and health impacts of new materials and their production processes. Extending the scope of life cycle analysis studies from the production process (cradle to gate) to commercial use (cradle to grave) and further to the recycling process of the materials used (cradle to cradle) is also necessary to develop a deeper understanding of their impacts over long periods of time. In addition, conducting a sensitivity analysis and assessing uncertainty propagation should be considered to ensure the accuracy of the results. Future studies should also examine alternative, environmentally friendly solvents and processing methods for non-water-soluble materials, enabling a more comprehensive assessment of potential formulations while ensuring comparable performance in PDRC applications.

In order to ensure the sustainable development of the passive cooling materials industry, the following approach is recommended. First, it is crucial to focus on materials with low energy requirements and minimal environmental impact, such as CaCO<sub>3</sub>. In addition, research should prioritize the use of environmentally friendly solvents, such as water, to reduce toxicity and ecological risks. Furthermore, the implementation of recycling and waste management strategies for these materials can reduce the environmental impact associated with disposal and minimizing resource extraction. Finally, assessing the life cycle of a material early in its design phase can help to identify potential environmental impacts and take necessary measures to mitigate them.

## 6. Conclusions

With a scope of minimizing the environmental footprint of cooling, alternative technologies such as materials with passive cooling capability during sunlight exposure are being sought. In this paper, the life cycles of four passive cooling materials were assessed by utilizing SimaPro software, the cradle-to-gate approach and the ReCiPe 2016 v1.01 method and their environmental footprints were compared. The results indicate that the calcium carbonate-based formulation has the least environmental impact among the four, while the formulation based on the different polymers has the highest. While passive cooling materials are presented as an emerging alternative to active cooling, few studies have focused on their life cycle assessment. In order for this technology to be harnessed for the transition to a more sustainable future, a more detailed study of the footprint of passive cooling materials is essential with cradle-to-grave and cradle-to-cradle approaches, which analyze the footprints of materials over a deeper time span. By continuing to explore and adopt greener materials and processes, the environmental performance of passive cooling technologies can be improved, contributing to a more sustainable and resilient to climate change future.

## CRedit authorship contribution statement

**Anastasia Athanasopoulou:** Writing – original draft, Software, Methodology, Investigation. **Dimitra Papadaki:** Writing – review & editing, Supervision, Methodology, Investigation, Conceptualization. **Chrysanthi Efthymiou:** Writing – review & editing, Methodology, Conceptualization. **Lorenzo Pattelli:** Writing – review & editing, Resources, Project administration, Funding acquisition. **Margarita-Niki Assimakopoulos:** Supervision, Project administration, Funding acquisition.

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## Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: none.

## Data availability

Data will be made available on request.

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