Cleaner Environmental Systems 11 (2023) 100139

Contents lists available at ScienceDirect

Cleaner Environmental Systems

journal homepage: www.journals.elsevier.com/cleaner-environmental-systems

Evaluation of abatement options to reduce formaldehyde emissions in vehicle assembly paint shops using the Life Cycle methodology

Daniel Granadero [a,b,*], Aida Garcia-Muñoz [b], Renate Adam [b], Francisco Omil [a], Gumersindo Feijoo [a]

[a] CRETUS, Department of Chemical Engineering, School of Engineering, Universidade de Santiago de Compostela, 15782, Santiago de Compostela, Spain
[b] Environmental Compliance and Sustainability, Stellantis, 65423, Rüsselsheim am Main, Germany

ARTICLE INFO

Keywords:
Vehicle paint shop
Formaldehyde emissions
Recuperative thermal oxidation
Regenerative thermal oxidation
Life cycle assessment (LCA)
Eco-efficiency

ABSTRACT

Recuperative thermal oxidizers, consisting of a single combustion chamber where volatile organic compounds are oxidized, and regenerative thermal oxidizers comprising several ceramic beds where the oxidation takes place, are the most common abatement technologies applied in vehicle paint shops to reduce formaldehyde emissions. In this work, a “cradle-to-grave” Life Cycle Assessment and an eco-efficiency analysis were carried out for a real paint shop to compare these two abatement technologies and identify the most environmentally sustainable option. The results show that the regenerative oxidizer leads to a decrease of the human toxicity impact category from 1329 kg 1,4-DB eq. in the initial situation without abatement to 1284 kg 1,4-DB eq., while an alternative with recuperative oxidizers achieves a significantly higher reduction to 1176 kg 1,4-DB eq. Considering the most relevant selected impact categories, the results demonstrate that the recuperative oxidizers cause a reduction from the initial situation of 2.6% of the normalized index, whereas the regenerative oxidizer implies a raise of 3.1%. This indicates that the installation of recuperative oxidizers is the most environmentally sustainable alternative from the two investigated technologies. Nevertheless, the eco-efficiency analysis confirms that the costs of the recuperative oxidizers option are 2.2 times higher.

1. Introduction

Vehicle manufacturing is one of the most complex industrial manufacturing processes because it requires large spaces as well as the sequential and coordinated operation of multiple technologies (Bysko et al., 2020; Giampieri et al., 2022). Painting operations have been identified as the stages with the greatest environmental impacts of the entire production process (VDI guideline 3455:2013-08; Onofre et al., 2020). The high demand for energy and resources, such as natural gas and raw materials for the production of coatings and paints, water consumption along with the generation of waste and wastewater are the most significant environmental aspects associated with the painting operations of vehicle assembly plants (STS BREF, 2020). In particular, when it comes to identifying the most polluting compounds in this type of facility, the environmental impact is marked by the emission of volatile organic compounds (VOCs), which represent the most relevant direct emissions. They are derived from spray booths, drying ovens and cleaning of equipment with organic solvents (Rivera and Reyes-Carrillo, 2014, 2016; Ou et al., 2022). The European Union (EU) car manufacturers emitted in 2021 an average of 2.2 kg/car and a total of 25.7 thousand tons of VOCs due to their vehicle painting activities (ACEA, 2022), which corresponds to 2.5% of the total VOCs emitted in Europe (German Environment Agency, 2023).

An exhaustive analysis of the paints and coatings used in a vehicle painting plant allows the identification of complex mixtures formulated from numerous components (Akafuah et al., 2016). In general, organic solvents, water, resins, plasticizers, dyes and pigments are the basic components of paints, being the binder (i.e. the resins) the predominant constituent defining the paint's characteristics (McMahon et al., 2023). Additionally, new developments in automotive paints include the use of nanoparticles as fillers (Nayane de Queiroz et al., 2022). The solvent content of color basecoats is in the range of 12–17% in water-based and 55–82% in solvent-based paints, while clearcoats typically have a solvent content up to 50% (STS BREF, 2020). One of the most commonly used binding agents in industrial coatings are amino resins and, mainly, those derived from melamine (Pizzi and Ibeh, 2022). Melamine resins used in automotive coatings are produced by the reaction of melamine

---

[*] Corresponding author. CRETUS, Department of Chemical Engineering, School of Engineering, Universidade de Santiago de Compostela, 15782, Santiago de Compostela, Spain.
E-mail address: daniel.granadero@rai.usc.es (D. Granadero).

https://doi.org/10.1016/j.cesys.2023.100139
Received 17 July 2023; Received in revised form 25 August 2023; Accepted 7 September 2023

Abbreviations

ALO	Agricultural Land Occupation
CC	Climate Change
COD	Chemical Oxygen Demand
COT	Total Organic Carbon
DNPH	2,4-Dinitrophenylhydrazine
EU	European Union
FD	Fossil Depletion
FE	Freshwater Eutrophication
FET	Freshwater Ecotoxicity
HPLC	High-Performance Liquid Chromatography
HT	Human Toxicity
IR	Ionizing Radiation
LCA	Life Cycle Assessment
LCI	Life Cycle Inventory
LCIA	Life Cycle Impact Assessment
MD	Metal Depletion
ME	Marine Eutrophication
MET	Marine Ecotoxicity
MSDS	Material Safety Datasheet
NLT	Natural Land Transformation
OD	Ozone Depletion
PMF	Particular Matter Formation
POF	Photochemical Oxidant Formation
RTO	Regenerative Thermal Oxidizer
S1	System 1
SS1	Subsystem 1
SS2	Subsystem 2
SS3	Subsystem 3
STS	Surface Treatment Using Organic Solvents
TA	Terrestrial Acidification
TET	Terrestrial Ecotoxicity
ULO	Urban Land Occupation
VOC	Volatile Organic Compound
WD	Water Depletion
WWTP	Wastewater Treatment Plant

and formaldehyde (Poth, 2008). Paint manufacturers indicate melamine resin contents varying from 0.1% to 18%, depending on the type of coating. Other amino resins used in coatings, such as those derived from urea or (meth)acrylamide, are also obtained by the reaction of different chemicals with formaldehyde (Pizzi and Ibeh, 2022). Thus, formaldehyde is present in automotive coatings as a residue associated with the resin manufacturing process (with a concentration below 0.1%, as can be taken from the material Safety Datasheets) but also as a component bound in melamine polymers.

Concerning the identification of formaldehyde sources, the emission of small quantities of this compound into the atmosphere present in the exhaust gas flow of the spray booth has been quantified at very low levels (< 1 mg/m^3) (Kim et al., 2011). However, also the bound formaldehyde from melamine resins is released from inside the drying chambers, which operate at elevated temperatures typically between 140 °C and 190 °C (Salthammer et al., 2010; Salthammer, 2019); therefore, depending on the configuration of the coating line and the efficiency of the end-of-process abatement equipment connected to the drying ovens, the formaldehyde emission can be significant (Sorrels et al., 2017). At present, not all vehicle painting facilities located in Europe have abatement equipment installed in the drying ovens (STS BREF, 2020). This depends on the specific legal requirements of each country, their specific set-up and the subsequent retrofitting steps from the plant assembly to the present time. Besides, the use of abatement equipment can be connected to the type of paints used in the paint shop. For instance, in 2016, 43% of the EU paint shops were estimated to apply solvent-based primer or basecoats, or a combination of both. These paint shops are more likely to have air emissions abatement equipment (STS BREF, 2020).

In 2015, formaldehyde was reclassified as a substance with carcinogenic potential (category 1B) and mutagenic potential (category 2) in the amendments to the so-called CLP Regulation on classification, labelling and packaging of substances and mixtures (Regulation (EC) No 1272/2008; Regulation (EU) No 605/2014; Regulation (EU) No 2015/491). With this change, all EU industries affected by the Industrial Emissions Directive 2010/75/EU shall, as far as possible, replace formaldehyde with less harmful substances. If formaldehyde emissions cannot be avoided, what is the case for painting operations in the automotive industry due to the use of the melamine resins, operators need to cope with significantly more stringent Emission Limit Values (Directive, 2010/75/EU).

Different technologies are available for the removal of VOCs from exhaust gases. These include biological treatment, recuperative thermal oxidation, regenerative thermal oxidation and catalytic oxidation (Mulholland and Dyer, 1999; Berenjian et al., 2012; Tomatis et al., 2019; Li et al., 2023). In the automotive industry, for the treatment of VOCs in vehicle paint shops, regenerative thermal oxidation and recuperative thermal oxidation, the latter sometimes in combination with catalytic oxidation, have been commonly applied (VDI guideline 3455:2013-08; STS BREF, 2020). Both technologies have proven to be efficient in VOC destruction with efficiencies higher than 95% (STS BREF, 2020). The introduction of catalytic oxidation to recuperative oxidizers helped reducing the high operating temperatures from 700 °C–740 °C to 400 °C necessary to obtain the desired abatement efficiency, with the consequent reduction in natural gas consumption (Yang et al., 2019; STS BREF, 2020; Brummer et al., 2022). However, this technique is not in use in vehicle paint shops because of the high maintenance requirements of the catalyst (STS BREF, 2020).

Given that both regenerative and recuperative thermal oxidation present important differences, the selection between one of them will depend on various aspects such as the VOC levels in the inlet gas, the flowrates to be treated, the spaces available in the paint shop, and the investment restrictions of the paint shop operator (Borwankar et al., 2012). In the selection of the most suitable technology, it is also crucial to consider the environmental impact of both technologies from a life cycle perspective in order to search for the most sustainable treatment method and take it into account in the decision-making process (Klöpffer and Grahl, 2014).

Life Cycle Assessment (LCA) is a powerful tool to analyze the environmental impacts of a product or process (ISO 14040:2006; ISO 14044:2006). There are several LCA studies related to environmental impacts in the automotive industry (Lopes Silva et al., 2018; Gebler et al., 2020), but most of them are focused on the vehicle itself, new engines, electric cars and automotive lithium-ion batteries (Hernandez et al., 2017; Bouter and Guichet, 2022; Winjobi et al., 2022; Guo et al., 2023). To the best of our knowledge, there are hardly any studies available in the scientific literature on LCA associated with the vehicle painting process. Papasavva et al. (2001) studied different coating materials commonly used in automotive painting, focusing on the use of powder paints as an alternative to solvent-borne paints with the advantage of not containing solvents and being easily reusable. Rivera and Reyes-Carrillo (2016) evaluated the overall environmental challenges of a paint shop, with special emphasis on metal and plastic surfaces. The areas with the greatest contribution to environmental impact were identified and potential process improvement alternatives focused on reducing energy consumption were proposed. Bianco et al. (2020)

provided a lifecycle inventory and LCA of an Italian automotive painting process, identifying the high energy consumption, the direct emissions of VOCs and the waste production and treatment as the hot points in several analyzed impact categories. Overall, it is likely that the scarcity of articles published on this topic is associated with confidentiality issues, with the consequent difficulty in collecting reliable data.

Beside the technical evaluation of alternative abatement technologies also an economic assessment of potential solutions is of importance for decision making. Eco-efficiency assessment is a quantitative tool to study the environmental impacts of a system in conjunction with its economic value (ISO 14045:2012) and helps entities to make conscious decisions (Torregrossa et al., 2018; Ramírez-Melgarejo et al., 2021). Thus, eco-efficiency analysis establishes a connection between the ecological impact of a system and its economic dimension (Desli et al., 2021). Although this identifies the solution with the most economic value and the lowest possible ecological impact, only a few eco-efficiency studies are available in the literature. Recent examples share research results on wastewater treatment plants (WWTPs) (Sala-Garrido et al., 2023); however, publications describing an eco-efficiency analysis for air emissions treatment equipment in the industry could not be found.

The objective of this work is to advance with real data on the environmental analysis of two different technologies for the reduction of formaldehyde emissions from the topcoat drying ovens of a real vehicle paint shop located in Europe. A “cradle-to-grave” LCA was carried out for the vehicle painting process in the initial situation of the paint shop and in two proposed scenarios corresponding to the two different abatement systems. As an essential step after the LCA, an eco-efficiency analysis was performed. The relevant cost data needed were obtained by an economic evaluation of the two abatement systems. This type of analysis is not available in the literature and it aims at demonstrating that the LCA methodology and eco-efficiency analysis can be used by vehicle manufacturing companies and paint shop operators to evaluate different process alternatives and favor the most environmentally sustainable technologies.

2. Materials and methods

2.1. Description of the paint shop

This study evaluates a real paint shop in a vehicle manufacturing plant located in Europe. The vehicle painting process consists of a sequence of dipping processes as well as the application of paints and coating materials using atomization equipment (Streitberger and Dössel, 2008). The different layers applied provide chemical and corrosion protection, weather and scratch resistance, as well as color and visual characteristics (Akafuah et al., 2016).

Before the main coating layers are applied to the vehicle body, a series of pre-treatment stages are carried out to clean and condition the body for the next painting steps (Debnath, 2013). The first stage is the pre-cleaning, followed by two or three degreasing phases. These steps remove oil, greases and lubricants from the previous manufacturing processes. After cleaning, an activation, a phosphating, a passivation and, finally, various rinsing and drainage stages are applied to increase the corrosion resistance of the vehicle body and prepare it for the next coating processes (Giampieri et al., 2020).

After the pre-treatment, the first main coating process is the electrocoating, in which the vehicle body is coated in a bath by applying the cataphoretic paint, providing additional corrosion resistance (Giampieri et al., 2020). In this electrochemical process, the paint is deposited on the metal by passing electric current through the body, causing paint to deposit on the surface in a uniform protection layer throughout the whole body (Marder and Goodwin, 2023).

Once the electrocoating has been completed, additional coating processes are applied for waterproof sealing of body and weld seams and to protect the underbody from damage with an elastic top layer. After sealing and underbody coating, the primer surfacer, the basecoat and the clearcoat paints are applied in spray booths for different purposes. The successive application of the basecoat and the clearcoat is commonly referred to as the topcoat. While the primer provides the vehicle body with further corrosion resistance and protection against stone chip, the basecoat and the clearcoat are mainly applied to add the required color and gloss effects to the vehicle. Moreover, the basecoat and the clearcoat give durability and protection against scratches and chemical aggressions (Jurgetz, 1995). Between the application of the different layers from electrodeposition to the topcoat each layer is cured or dried before the application of the subsequent coating. This happens in drying ovens which are typically connected to emissions control equipment to eliminate VOCs and hazardous substances released from the ovens (STS BREF, 2020).

Additional waxing, sealing and repairing operations on the dried topcoat complete the process and ensure that the desirable quality and corrosion protection requirements are met (Giampieri et al., 2020).

The paint shop under study has two parallel baths for the electrocoating process with two drying ovens per bath. To eliminate VOC emissions and unpleasant odors generated in the ovens, each oven is connected to two individual recuperative thermal oxidizers. The oxidizers are operated on natural gas as fuel, and the excess heat of the oxidizers is used to heat the oven by means of heat exchangers. As a result of the oxidation process, nitrogen oxides (NOx) and carbon monoxide (CO) are emitted into the atmosphere through the abatement installations exhaust systems (STS BREF, 2020).

For the primer surfacer application, the paint shop has a line with several paint booths in which a coating of waterborne primer is applied to the vehicle bodies using robots. In order to dry all vehicle bodies after the priming process, the line has three ovens which, similar to the electrocoating process, are connected to the abatement installations, i.e. recuperative thermal oxidizers, with the aim of treating the VOC emissions (including formaldehyde) produced during the drying process.

Primer coating is followed by the finishing processes, which consist of the application of a basecoat and a clearcoat that are applied in successive processes separated by a short flash off area. The paint shop studied uses solvent-based basecoat and clearcoat and has four coating lines with several booths where the paints are applied by robots. At the end of each clearcoat line, two ovens are installed in order to dry the painted vehicle bodies. Unlike the electrocoating and primer processes, the ovens are not connected to abatement installations. As a result, VOC emissions, including the formaldehyde released in the ovens, are emitted into the atmosphere. Part of the exhaust air from the basecoat paint booths is treated in two recuperative thermal oxidizers. In a previous stage, the volatile organic compounds present in the exhaust gas are partially removed in an activated carbon unit (Berenjian et al., 2012).

Proposed abatement scenarios. To remove or reduce formaldehyde emissions from the topcoat (basecoat and clearcoat) ovens, two alternative treatment methods corresponding to two different abatement technologies were evaluated from both economic and environmental viewpoints.

The first alternative was the installation of a regenerative thermal oxidizer (RTO) that can treat all waste gases from the topcoat ovens. In regenerative thermal oxidation, organic solvents are oxidized at a temperature between 800 °C and 850 °C (VDI guideline 3455:2013-08). The raw gas enters a first bed filled with ceramic medium and is preheated before entering the combustion chamber. The first bed is cooled down while the gas is heated. After leaving the combustion chamber, the hot gas passes through a second bed, releasing the energy to the ceramic packaging. The clean gas is then cooled down and released into the atmosphere. The process is carried out alternately so that in the next cycle, the inlet gas is preheated in the second bed and the first bed is where the hot gas is cooled. Using ceramic materials, the heat is stored and most of the energy produced by combustion is reused in the system itself without the need for additional heat exchangers (VDI guideline 3455:2013-08).

The number of ceramic beds in an RTO varies and follows a certain sequence of preheating and cooling steps taking place alternately in the different beds. The destruction efficiency of the regenerative thermal oxidation is 95% (STS BREF, 2020). For the paint shop studied, a 5-bed RTO was assumed for the treatment of the waste gas volumes from the topcoat ovens.

The second alternative technology considered was the installation of recuperative thermal oxidizers. In recuperative thermal oxidation, VOCs are oxidized in a simple oxidation chamber by chemical reaction with oxygen present in the exhaust air (STS BREF, 2020). The chamber has an auxiliary burner running on natural gas in which the waste gas is heated up to 700 °C–740 °C, leading to an almost complete oxidation, i.e. a destruction efficiency close to 100% of the contained organic compounds (VDI guideline 3455:2013-08; STS BREF, 2020). Energy is saved by preheating the raw gas with the hot flue gas in an intermediate heat exchanger. Additionally, the energy excess of the clean gas is used to heat the drying ovens. For this study, eight oxidizers are assumed to treat the waste gas volume from the topcoat ovens.

2.2. LCA methodology

An attributional LCA of the painting process in the paint shop being studied was performed from a “cradle-to-grave” perspective following the requirements and guidelines of the ISO 14040:2006 and ISO 14044:2006.

Goal definition and strategy. The goal of this LCA was to evaluate the alteration of the environmental impact of the painting process when introducing two different abatement systems for the reduction of formaldehyde emissions, allowing the comparison of both technologies and supporting the selection of the most suitable system during the planning process. This LCA is situated at the micro strategic level of the vehicle manufacturing plant since the step to reduce the emissions of formaldehyde is one of the numerous actions to increase its environmental performance and support corporate environmental strategies.

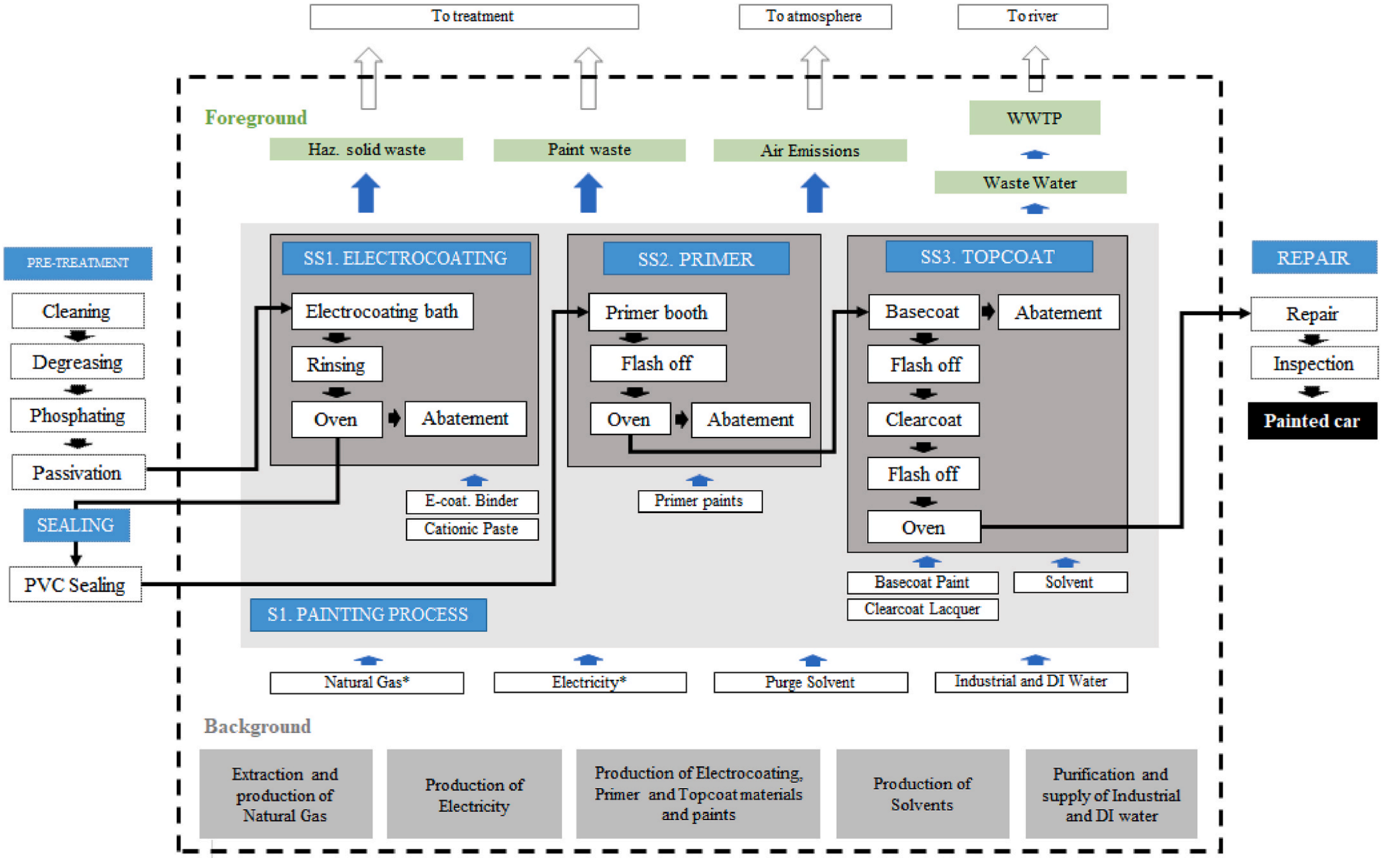
Functional unit. All the inputs and outputs of the painting process are related to the functional unit. The vehicle paint shop processes 360,000 vehicles per year, which corresponds to about 75% of its total capacity. The functional unit selected was 1 h of operation for each subprocess investigated.

System boundaries. From the entire painting process electrocoating, primer and topcoat were evaluated as they are the most relevant subprocesses in terms of atmospheric emissions and, in particular, formaldehyde releases. The study considered only the operation of the equipment while the construction of the installations was left out of the LCA. The environmental impact of pre-treatment, sealing, waxing and repairing operations was considered not relevant and thus is not included in the scope of this work (VDI guideline 3455:2013-08).

The scope of the study is visualized by the system boundaries shown in Fig. 1. Additionally, the figure illustrates the inputs and outputs to the sub-processes within scope as well as their background and foreground systems, used as the basis to elaborate the Life Cycle Inventory (LCI).

As inputs to the painting operations natural gas, electricity, purge solvents, water and electrocoating and painting materials were considered. The background processes are those related to the production and supply of materials and energy necessary to operate the installations:

- *Extraction and production of Natural Gas*: Natural gas is needed to run the burners in order to heat the ovens and to operate the abatement equipment. It is provided by the natural gas supplier of the manufacturing plant. Its extraction and production processes cause environmental impacts, such as emissions to air, water depletion and wastewater generation (Shamoon et al., 2022).



* Total painting processes consumption was split to the three subsystems based on allocation factors

Fig. 1. Scheme of the system boundaries and background and foreground processes for the paint shop under evaluation.

- *Production of electricity*: Electricity is necessary to operate the paint shop installations, such as robots, air supply for spray booths, compressed air and lighting. The contribution of each energy source for the electricity generation was taken from the country mix where the paint shop is located.
- *Production of Electrocoating, Primer and Topcoat materials and paints*: The production of the various materials and paints used in the painting processes conforms a significant background process to this system. It implies raw material extraction, the use of energy and water for the production process, direct emissions, wastewater and waste generation (da Silva et al., 2016; Nair K et al., 2021; Shi et al., 2023).
- *Production of Solvents*: Similar to the production of materials and paints, the manufacturing of solvents used for viscosity adjustment of the paints as well as for cleaning of painting equipment has associated environmental impacts.
- *Purification and supply of industrial and DI (deionized) water*: Industrial and deionized water is used to carry out the electrocoating process and to wash out the overspray from the spray booths. Water coming from the river is processed and purified to be used in the paint shop in a series of decantation, decarbonization and filtration processes and, in the case of deionized water, a deionization process. The deionization is performed in an ion-exchange resin system.

As outputs from the painting operations the study reflects air emissions, waste and wastewater, whereof wastewater is treated onsite in a WWTP before discharge. In more detail, the foreground processes are:

- Emissions into the atmosphere of VOC, dust and formaldehyde from paint booths, ovens and air emissions abatement equipment and CO, CO_2 and NOx from burners and abatement equipment.
- Release of wastewater pollutants in concentrations below given thresholds, i.e. COD, COT, Zinc, Iron, Phosphorus, Oil & greases, suspended solids and Fluorides, after the treatment in an onsite WWTP discharged into the river. Wastewater treatment is included in the system boundaries of this work.
- The generation of hazardous solid waste and other paint waste which leave the site to be treated in an off-site waste treatment facility. Waste treatment is outside of the system boundaries of this study.

Data collection and data quality. For the elaboration of the LCI a process-based bottom-up approach was followed. Real consumption and emissions data for inputs and outputs of the painting process were directly obtained from the paint shop. Materials consumption data are based on logistical and financial databases, as well as information obtained from the operational control systems of the paint facility. The composition of the materials is available through the material Safety Data Sheets (Regulation (EC) No 1907/2006) and additional information provided by the suppliers. Paints used in the topcoat process differ in composition and solvent content. In order to elaborate the LCI for the topcoat process, an exhaustive evaluation of all materials was performed to create several clusters representative for the variety of products used.

The consumption of natural gas and electricity was obtained from meters installed in the paint shop, while the allocation of energy consumption by sub-process was made using bibliographic values from the STS Best Available Techniques Reference Document.

VOC emissions were calculated using annual mass balances in accordance with the relevant European legislation (Directive, 2010/75/EU). Data on the rest of the atmospheric emissions and wastewater parameters were obtained from actual measurement campaigns and installed standard monitoring equipment commonly accepted in industry (VDI guideline 3455:2013-08; STS BREF, 2020). In cases where continuous monitoring results or data for the whole year were not available, the average results obtained during the monitoring periods were used. Formaldehyde concentrations in emission sources were determined using the DNPH method (VDI 3862 Part 2:2000-12), whose protocol states that from a known volume of gas samples withdrawn using glass impingers containing a solution of 2,4-dinitrophenylhydrazine (DNPH), the samples are transferred to dark glass vials for HPLC analysis. The emission loads of the different pollutants measured in each emission source of the analyzed sub-processes were summed up to calculate the total emissions of each pollutant for the elaboration of the LCI.

Finally, the waste data were collected from the monthly waste transfer records of the paint shop for the different identified waste streams.

2.3. Assessment methodology and impact categories

A Life Cycle Impact Assessment (LCIA) was conducted using commercial software SimaPro v.8.2 with European ReCiPe Midpoint V1.12 methodology. This method is scientifically sound, easy to use and interpret, and internationally accepted. The method focuses on environmental impact and damage, distinguishing 18 different impact categories (Geldermann and Rentz, 2005; ILCD, 2010a; ILCD, 2010b; Goedkoop et al., 2013).

After the selection of the impact categories and the classification of the inventory results as the initial steps of the LCIA, the characterization and normalization steps defined in the ISO 14040 and ISO 14044 standards were followed to carry out the impact assessment. The characterization of the system in its initial situation without abatement was performed in order to identify the sub-process with the greatest environmental impact. In addition, a characterization of each individual sub-process, i.e. electrocoating, primer and topcoat, was conducted to obtain the inputs and outputs with the highest contribution to the environmental impact.

The next step was the characterization of the system with the two alternative air emissions abatement technologies. In order to simplify the analysis, this step was conducted exclusively for the topcoat process since the application of the abatement systems was only considered for this sub-process. After the characterization, the results for the impact categories were normalized for the system in the initial situation and with the two abatement technologies to allow a better comparison.

For the comparison of the normalized data, in a first analysis, all 18 impact categories included in the methodology were taken into account. In subsequent stages, the most relevant impact categories were selected on the basis of these two criteria:

- All impact categories whose relevant contribution to the total environmental impact was higher than 2%. Eight categories could be identified by evaluating the normalization results. These eight relevant impact categories contribute to 95.5% to the total environmental impact.
- Human toxicity (HT) exclusively, since the reduction of human toxicity through the reduction of formaldehyde emissions due to its cancer-causing characteristic is the objective of the abatement equipment in this study.

Finally, for the purpose of comparing the two proposed scenarios with the initial situation and identify the most environmentally favorable solution, the normalized index considering only the HT impact category and the sum of the normalization values for the selected impact categories as described above were represented for each scenario.

2.4. Methodology for economic evaluation and eco-efficiency analysis

As a prerequisite for the eco-efficiency analysis, an initial study of the two abatement systems was performed, including an economic evaluation to obtain all necessary economic data. The capital costs for the installation of the two systems were requested from equipment manufacturers for the paint shop studied. The operational and maintenance costs data were available from other company plants with similar equipment. These costs were calculated for the whole lifespan of the installations (RTO = 25 years; Recup. Oxidizers = 15 years) and the annual increase in the cost of natural gas and spare parts for equipment was taken into account using the average inflation rates of recent years of the country where the paint shop is located. Finally, the total costs were normalized to the functional unit to allow the comparison between the two systems.

Ultimately, to bring the economic perspective into the comparison, an eco-efficiency analysis was conducted based on the criteria prescribed in the ISO 14045:2012.

The eco-efficiency analysis was performed with the focus set on the topcoat process, for which the abatement equipment was planned. The normalization results previously obtained in the LCA were used for the eco-efficiency evaluation. Two different indicators were selected and represented against the total cost per hour:

- Variation of the normalized index, expressed in percentage, considering the sum of the impact categories with a relevant contribution to the total environmental impact greater than 2% in the normalization results.
- Variation of the normalized index, expressed in percentage, of the HT impact category.

It must be highlighted that, even though the LCA did not consider the construction of the painting installations but only the operation of the equipment, the costs for the installation of the two planned abatement systems could not be excluded. They are the major cost factor in the total calculated costs of the two systems and are necessary to achieve a representative comparison. Besides, all other painting equipment and installations were already available in the paint shop at the time of this study and no additional investment would be necessary.

3. Results and discussion

3.1. Initial evaluation of the abatement systems

An initial evaluation of the two different abatement systems was performed prior to the LCA. This information is required to better understand the differences between the two systems and, along with the results obtained in the LCA and eco-efficiency analysis, support the decision-making process. The advantages and drawbacks of both technologies can be summarized as follows:

- As far as natural gas consumption is concerned, the installation of eight recuperative thermal oxidizers was considered much more convenient since the excess heat can be reused to heat the topcoat drying ovens, with the consequent elimination of existing burners and the concomitant savings of natural gas.
- Initial investment costs are much higher for the eight recuperative thermal oxidizers with their connected heat exchangers than for the RTO. Maintenance costs are higher for the RTO, mainly due to the regular exchange of the ceramic packaging. Generally, the installation of the RTO is more advantageous from an economic point of view.
- For the installation of recuperative oxidizers, longer production shutdown periods are needed, which would have an impact on the production volumes of the plant. An RTO can be built in parallel to the production and only several weeks of production shutdown are needed to conclude the installation. Therefore, the timeframe for the installation of an RTO is considerably shorter than the time required for the installation of the recuperative oxidizers.
- The RTO is independent of the topcoat painting lines and ovens, so that in the event of breakdowns, production can continue without vehicle losses and production delays. Since the excess heat from the recuperative thermal oxidizers would be used to heat the drying ovens, drying and painting shall be stopped in case of malfunctions and necessary repairs.

A simplified representation of the main differences, advantages and drawbacks of the two alternative systems is shown in Table 1.

3.2. Life Cycle Inventory

The inventory data presented in Table 2 correspond to the stepwise painting process that takes place in the paint shop analyzed. Data were collected for one year of production and all data refer to the functional unit of 1 h of operation. Even though the reduction of formaldehyde emissions constitutes the focus of this work, the emissions of all other air and water pollutants that occur in the vehicle paint shop were considered in order to elaborate the life cycle inventories.

To compile the life cycle inventories, data were grouped into three subsystems corresponding to the coating and painting steps included in the scope of this work. The three subsystems together constitute an overall painting process system as shown in Table 2. Additionally, all inputs and outputs that could not be assigned to the different subsystems were included in the global paint process system for the LCA.

The installation of the two different abatement technologies compared in this work for the waste gas stream led to significant variations in the inventory data. The differences are shown in Table 3.

The data were collected from the continuous sampling and monitoring campaigns of two sister plants with similar characteristics and in which both types of abatement technology are currently in operation. At the same time as the overall formaldehyde emissions from the paint facility were reduced by the abatement equipment in the topcoat process, a significant increase was observed in the concentration of the co-products from the oxidation process, i.e. NOx and CO. It was noted that the emission concentrations of formaldehyde, NOx and CO from the abatement system were very similar for both technologies. Thus, the absolute emissions of the three pollutants in terms of kilogram per hour were considered to be the same in the Life Cycle Inventory (LCI). The major difference found in developing the inventory data for the two different systems is due to the use of natural gas. All other inventory parameters for input materials, water consumption, quantities of waste and wastewater generated, and wastewater emissions were not affected due to the installation of the waste gas abatement equipment.

3.3. Life Cycle Assessment

Characterization. In the characterization step, the hot spots were detected by analyzing the contribution of the different subsystems to the

Table 1
Comparison of the main characteristics of the two alternative abatement systems identified in the initial evaluation for the paint shop of study. Authors' own elaboration.

Main Characteristics	5-bed RTO	8 Recuperative Thermal Oxidizers
Abatement efficiency	95–99%	≈100%
Initial investment costs	Lower	Higher
Maintenance costs	Higher	Lower
Natural gas consumption	Higher	Lower due to heat recovery to oven
Energy recovery	Not possible in this paint shop	Excess heat to ovens
Ability to Run Oven Without Abatement System	Yes	No
Space Utilization	Lower	Higher due to high number of devices
Accessibility for Repair or Parts Replacement	Higher	Lower
Time to install	During production	Outside production
Lifespan	Long Life Equipment	It will have to be replaced during life of oven

Table 2
Life Cycle Inventory (LCI) of the initial situation of the paint shop under evaluation. All quantities are referred to the functional unit, i.e. 1 h of operation for each sub-process.

	S1 Painting Process	SS1 Electrocoating	SS2 Primer	SS3 Topcoat	Unit	Method/Source of data
INPUTS: from Technosphere						
Materials						
Water DI	8.39	2.39			m^3	Paint shop consumption records
Purge solvent	73.1				kg	Paint shop consumption records
Industrial water	16.07				m^3	Paint shop consumption records
Binder:						Paint shop consumption records
Organic solvent		12.18			kg	MSDS[a]
Epoxy resins		203			kg	Supplier information[b]
Water		190.82			kg	Calculation
Cationic paste:						Paint shop consumption records
Organic Solvent		139.09			kg	MSDS
Epoxy resins		794.8			kg	Supplier information
Pigment		397.4			kg	Supplier information
Water		655.71			kg	Calculation
Primer Paints:						Paint shop consumption records
Organic solvent			4.82		kg	MSDS
Polyester resins			15.41		kg	Bibliographic research[c]
Amino resins			6.74		kg	Supplier information
Free formaldehyde			0.1		kg	Supplier information
Polyurethane			1.93		kg	Supplier information
Water			38.52		kg	Supplier information
Pigments			28.89		kg	Calculated
Basecoat Paints:						Paint shop consumption records
Organic solvent				85.4	kg	MSDS
Polyester resins				32.2	kg	Bibliographic research
Amino resins				14	kg	Supplier information
Free formaldehyde				0.14	kg	Supplier information
Pigment white				0.14	kg	Calculated
Pigment Black/grey				4.2	kg	Calculated
Pigment Blue/Green				0.84	kg	Calculated
Additives				1.4	kg	Supplier information
Basecoat White Paint:						Paint shop consumption records
Organic Solvents				19.32	kg	MSDS
Polyester Resins				7.14	kg	Supplier information
Amino resins				2.94	kg	Supplier information
Free formaldehyde				0.04	kg	Supplier information
Pigment white				11.76	kg	Supplier information
Additives				0.84	kg	Supplier information
Clearcoat Lacquer:						Paint shop consumption records
Organic solvent				82.81	kg	MSDS
Amino resins				28.73	kg	Supplier information
Hydroxyl Resin/Acrylic				52.39	kg	Supplier information
Free formaldehyde				0.17	kg	Supplier information
Additives				5.07	kg	Supplier information
Solvents Basecoat (viscosity adjustment):						Paint shop consumption records
Butanol				7.28	kg	MSDS
Butylacetate				25.48	kg	MSDS
Xylene				53.69	kg	MSDS
Solvent organic				5.46	kg	MSDS
Energy						
Natural Gas		4.81	6.23	15.2	MWh	Paint shop energy records
Electricity		2.04	2.35	7.22	MWh	Paint shop energy records
Cogeneration		1.38	1.59	4.88	MWh	Paint shop energy records
OUTPUTS: emissions to the environment						
Emissions to air[d]						
Dust	3.51	0.05	0.27	10.71	kg	Measured, Calculated
NOx	24.62	4.15	8.09	35.02	kg	Measured, Calculated
CO	7.4	20.6	2	4.3	kg	Measured, Calculated
CO_2	433	1193	n.a.[e]	n.a.	kg	Measured, Calculated
VOCs	73.13	4.42	1.3	200.57	kg	Calculated
Formaldehyde	0.12	0.01	0.01	1.36	kg	Measured, Calculated
Emissions to water[f]						
COD	3.25	222			g O_2	Measured, Calculated
COT	0.99	59.8			g	Measured, Calculated
Zinc	0.03	1.7			g	Measured, Calculated
Iron	0.03	2.4			g	Measured, Calculated
Phosphorus	0.11	6.86			g	Measured, Calculated
Oil & greases	0.11	7.45			g	Measured, Calculated
Suspended solids	0.59	41			g	Measured, Calculated
Fluorides	n.a.	15.8			g	Measured, Calculated

(*continued on next page*)

Table 2 (*continued*)

	S1 Painting Process	SS1 Electrocoating	SS2 Primer	SS3 Topcoat	Unit	Method/Source of data
Waste						
Emulsion paints to incineration	4.37				kg	Paint shop waste records
Paint to be separated	68.6				kg	Paint shop waste records
Solid hazardous to incineration	1.78				kg	Paint shop waste records

[a] MSDS: Material Safety Datasheet.

[b] Supplier Information refers to indicative percentages communicated by suppliers of the constituents of each paint and material. The percentages were used to calculate the absolute amounts of each constituent in the input materials.

[c] Stoye and Freitag (1998). Bibliographic research refers to indicative percentages of the constituent in paints and materials.

[d] The "emissions to air" values were calculated as the sum of the emission loads measured at all emission sources of the different sub-processes of the paint shop. The measurement uncertainties of the method are as follows: Dust: n.a.; NOx: 15%; CO: 9%; Formaldehyde: 15%.

[e] n.a.: not available.

[f] The "emissions to water" values were calculated by means of the annual average concentration values for two different wastewater streams and considering the total volume flows and operating hours.

Table 3

Differences in the LCI for the topcoat process due to the abatement installation. All quantities are referred to the functional unit, i.e. 1 h of operation for each sub-process.

	Initial Situation	5-bed RTO	8 Recuperative Oxidizers	Unit
INPUTS: from Technosphere				
Energy				
Natural Gas	15.2	17.9	12.7	MWh
OUTPUTS: emissions to the environment				
Emissions to air				
NOx	35.0	56.74	56.74	kg
CO	4.27	9.63	9.63	kg
VOCs	201	172	172	kg
Formaldehyde	1.36	0.48	0.48	kg

impact categories. An initial assessment of the overall painting process (Fig. 2 (a)), comprising the inputs and outputs of the electrocoating, primer and topcoat processes, as well as all other inputs and outputs that could not be assigned to the three subsystems, shows that the topcoat process has the largest contribution for most impact categories. The smallest effect of the topcoat process can be noticed in the WD impact category with 40.1 m^3 in a total of 246.6 m^3 (16.3%), whilst the highest share of this process in an impact category can be observed in POF with 251.4 kg NMVOC in a total of 422.2 kg NMVOC (59.5%). Furthermore, the impact categories MD = 242.1 kg Fe eq. for the topcoat process (a contribution of 59.4% to the total of the impact category), FET = 164.4 kg 1,4-DB eq. (57.3%), MET = 137.0 kg 1,4-DB eq. (57.3%), NLT = 1.48 m^2 (55.3%), OD = 0.001 kg CFC-11 eq. (54.1%), ALO = 106.9 m^{2}a (53.6%), TET = 0.65 kg 1,4-DB eq. (53.5%) and IR = 230.0 kBq U235 eq. (53.2%) are remarkably affected by the topcoat process as the given percentages indicate.

These results can be explained by the fact that the topcoat is the process with the highest consumption of materials and producer of emissions, in particular the use of solvent-borne paints, higher consumption of natural gas due to the use of more burners to heat the drying ovens than in the other sub-processes, and the direct emissions into the atmosphere without installed abatement systems. A similar outcome was observed by Bianco et al. (2020), specifically for the POF impact category, who identified the provision of heating and the direct emissions of VOCs as hot spots of the vehicle painting process.

Additionally, it can also be observed that the electrocoating process plays an important role in all impact categories, with the greatest contribution in the categories of WD = 172.9 m^3 (70.2% contribution to the total of the impact category), PMF = 25.2 kg PM10 eq. (51 %), CC = 10224.5 kg CO$_2$ eq (47.3%), FD = 3587.1 kg oil eq. (45.4%) and TA = 49.1 kg SO$_2$ eq. (42 %), primarily due to the materials used in this process. Subsequently, the three subsystems were analyzed separately:

Electrocoating process: Fig. 2 (b) shows the results of the characterization of the electrocoating process. For most of the impact categories, the materials used in the electrocoating baths, i.e. the binder, consisting of organic solvents, epoxy resins and water, and the cationic paste, which is made of organic solvents, epoxy resins, pigments and water, make the greatest contribution. In particular, the cationic paste presents the largest impacts. For instance, in the NLT and the FE impact categories, the cationic paste represents 43.4% (0.24 m^2) and 59.1% (0.31 kg P eq.) of the total impact category, respectively. In order to identify which component of the cationic paste is responsible for this result, a characterization of the cationic paste based on its constituents was carried out. In this analysis, it was observed that the epoxy resin in the formulation of the cationic paste is the main contributor to the majority of the impact categories, being POF = 0.02 kg NMVOC (97.3% of the total of the impact category), FD = 1.07 kg oil eq. (97.2%), PMF = 0.009 kg PM10 eq. (96 %), CC = 2.71 kg CO$_2$ eq. (95.8%) and TA = 0.016 kg SO$_2$ eq. (93 %) the most affected categories. This result can be mainly explained due to the direct emissions of air pollutants associated with the resin production process, such as CO$_2$ from combustion processes (Wilson, 2009).

Primer process: As can be seen in Fig. 2 (c), most of the impact categories are highly affected by natural gas consumption, given its background processes related to its extraction and production, each with a high impact. The direct emissions of the primer process are more relevant than in the electrocoating and have their main contribution to the impact categories of POF = 9.5 kg NMVOC (67.1%), ME = 0.32 kg N eq. (61.8%), PMF = 1.78 kg PM10 eq. (42.9%) and TA = 4.53 kg SO$_2$ eq. (36.4%). With regard to the acidification and eutrophication impact categories, the high relevance of the direct emissions can be assigned to the NOx emissions coming from the oxidation process of the recuperative thermal oxidizers installed in the primer ovens, what confirms observations made by Banar and Çokaygil (2010).

Topcoat process: The characterization results of the topcoat process are shown in Fig. 2 (d). To simplify the interpretation of the results, all created paint clusters were finally grouped into an individual system namely “Total paints”. These results present a similar behavior as for the primer process. Direct emissions considerably increase their relevance in the following impact categories: POF = 236.9 kg NMVOC (94.3%), ME = 1.37 kg N eq. (70.3%) and PMF = 7.7 kg PM10 eq. (53.4%). Moreover, the share of the direct emissions in the TA impact category raises with 19.6 kg SO$_2$ eq. to 47.8%, 11.4% higher than in the primer process. This trait can be attributed to two reasons. Firstly, the increment of the NOx emissions in the exhaust air of the recuperative thermal oxidizers installed in the basecoat spray booths. In particular, the high contribution of the direct emissions to the TA and ME categories is manifestly connected to the NOx emissions from the oxidation process (Banar and Çokaygil, 2010). Secondly, the increase of the VOC emissions from the topcoat ovens, where the solvent-borne paints are being cured without abatement equipment, unlike the primer process. The high impact of the direct emissions to the POF impact category is reasonable since VOCs are well-known precursors of photochemical smog and tropospheric ozone,

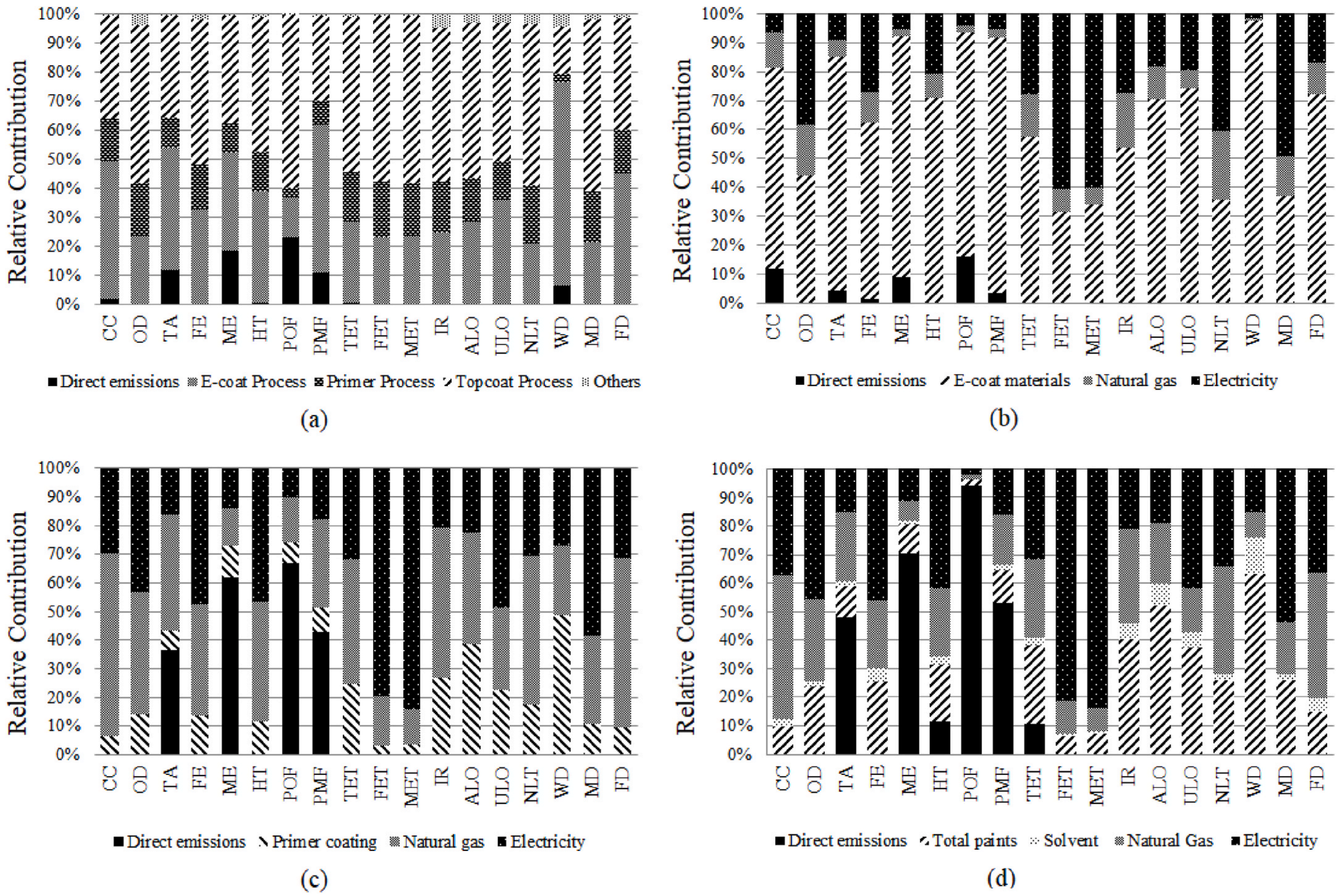


Fig. 2. Results of the LCI characterization. (a) S1: Painting process; (b) SS1: Electrocoating process; (c) SS2: Primer process; (d) SS3: Topcoat process. Acronyms used in this figure: CC: Climate Change; OD: Ozone Depletion; TA: Terrestrial Acidification; FE: Freshwater Eutrophication; ME: Marine Eutrophication; HT: Human Toxicity; POF: Photochemical Oxidant Formation; PMF: Particulate Matter Formation; TET: Terrestrial Ecotoxicity; FET: Freshwater Ecotoxicity; MET: Marine Ecotoxicity; IR: Ionizing Radiation; ALO: Agricultural Land Occupation; ULO: Urban Land Occupation; NLT: Natural Land Transformation; WD: Water Depletion; MD: Metal Depletion; FD: Fossil Depletion.

as also discussed by Finlayson-Pitts and Pitts (2000), Xu et al. (2015) and Xu et al. (2016).

As shown in Fig. 2 (d), the relevance of the paints in most impact categories notably increased in comparison to the primer process. The highest contributions of the topcoat paints can be observed in WD = 25.3 m^3 (63.1%), ALO = 56.1 m^2a (52.4%) and IR = 93.0 kBq U235 eq. (40.4%), whose relevance can presumably be traced back to the manufacturing process of the paints and to their constituents.

As can be observed in the characterization results for all the sub-processes, natural gas consumption has a relevant influence on almost all impact categories and therefore on the total environmental impact of the painting process. For instance, in the topcoat process, CC = 3897.4 kg CO_2 eq. (50.5%), FD = 1338.2 kg oil eq. (43.8%) and NLT = 0.55 m^2 (37.5%) are the most affected impact categories. Moreover, natural gas plays an important role in the OD impact category with a relevant contribution of 29.0%. The contributions of the natural gas to the CC, FD and OD impact categories were already reported in other studies (Skone et al., 2016; Tomatis et al., 2019).

Proposed abatement scenarios: A characterization of the two proposed scenarios considering the topcoat process was performed and the results compared to the initial situation. For the majority of the impact categories, an increase can be observed when the RTO scenario is compared to the initial situation. On the contrary, most of the categories decrease their values when the scenario with the recuperative oxidizers is compared to the initial situation. The highest differences could be found for CC (initial situation = 7715 kg CO_2 eq; RTO = 8407 kg CO_2 eq; Recup. Oxidizers = 7082 kg CO_2 eq), FD (initial situation = 3056 kg oil eq.; RTO = 3294 kg oil eq.; Recup. Oxidizers = 2839 kg oil eq.), NLT (initial situation = 1.48 m^2; RTO = 1.58 m^2; Recup. Oxidizers = 1.39 m^2), IR (initial situation = 230.0 kBq U235 eq.; RTO = 243.6 kBq U235 eq.; Recup. Oxidizers = 217.6 kBq U235 eq.), OD (initial situation = 1.036×10^{-3} kg CFC-11 eq.; RTO = 1.089×10^{-3} kg CFC-11 eq.; Recup. Oxidizers = 0.987×10^{-3} kg CFC-11 eq.), and TET (initial situation = 0.647 kg 1,4-DB eq.; RTO = 0.633 kg 1,4-DB eq.; Recup. Oxidizers = 0.572 kg 1,4-DB eq.). Regarding the reduction of the HT, the main objective of the installation of the abatement system, the results obtained for the initial situation and the two scenarios are: initial situation = 1329 kg 1,4-DB eq.; RTO = 1284 kg 1,4-DB eq.; Recup. Oxidizers = 1176 kg 1,4-DB eq. A reduction in the HT category was achieved by both scenarios, with a decrease being significantly higher for the recuperative thermal oxidizers compared to the RTO. Furthermore, in the impact categories ME (initial situation = 1.94 kg N eq.; RTO = 2.81 kg N eq.; Recup. Oxidizers = 2.77 kg N eq.), PMF (initial situation = 14.46 kg PM10 eq.; RTO = 19.69 kg PM10 eq.; Recup. Oxidizers = 18.83 kg PM10 eq.) and TA (initial situation = 41.0 kg SO_2 eq.; RTO = 54.9 kg SO_2 eq.; Recup. Oxidizers = 51.6 kg SO_2 eq.), an increment was observed for both proposed systems but a higher increase is caused by the RTO.

All in all, the characterization results indicate that, from the two proposed scenarios, the installation of an RTO is the less beneficial from the environmental impact perspective. This result can be explained by the fact that the installation of eight recuperative oxidizers implies an overall reduction in natural gas consumption due to the energy recovery in the heating of the drying ovens with the excess heat from the combustion process. The installation of the RTO implies an overall increase in the natural gas consumption of the plant, which leads to an increase of the total environmental impact. This observation is in line with the results obtained by Tomatis et al. (2019), who compared two abatement systems, i.e. an RTO and a catalytic thermal oxidizer, to an outdated thermal oxidizer. In this study, the auxiliary fuel used to run the equipment was also identified as the major contributor to the overall environmental impact. This study also highlighted the significant role of natural gas to the HT impact category and explains the higher decrease in HT obtained by the recuperative thermal oxidizers due to the lower natural gas needs.

In order to better illustrate these results, a normalization of the data was carried out.

Normalization. Fig. 3 shows the results of the normalization represented to allow the comparison between the initial situation and the two technical alternatives.

Fig. 3 (a) represents the normalized index for the topcoat process considering the eight relevant impact categories selected as explained in the *Materials and methods* section. An increase of the normalized index from 51.5 in the initial situation to 53.1 for the installation of the RTO is observed, while a reduction from 51.5 to 50.1 is achieved with the installation of the eight recuperative thermal oxidizers. This corresponds to a raise in the normalized index of 3.1% for the RTO against a decrease of 2.6% for the recuperative thermal oxidizers. A similar effect was observed when considering the 18 impact categories.

In this context, the ultimate objective of the waste gas abatement facility is the reduction of formaldehyde emissions and therefore the reduction of the human toxicity impact category. Consequently, the normalization data for the HT impact category was represented for the initial situation and the two scenarios to better illustrate the reduction in human toxicity achieved by the installation of the two abatement systems (Fig. 3 (b)). The RTO installation achieves a reduction of 3.4% in the normalized index with regard to the initial situation without abatement equipment (initial situation = 2.11; RTO = 2.04), while in the case of the installation of the recuperative thermal oxidizers a decrease of 11.5% was achieved (initial situation = 2.11; Recup. Oxidizers = 1.87). The difference in the reduction on human toxicity can be explained by the difference in natural gas consumption of both alternatives. The reduction in formaldehyde emissions was achieved by both technologies; however, the increase in natural gas consumption due to the installation of the RTO implies a smaller decrease in the HT impact category than for the recuperative oxidizers, as observed by the characterization results.

3.4. *Eco-efficiency analysis*

The results of the eco-efficiency analysis are represented in Fig. 4. The percentage variation of the normalized index has been assessed with the costs in € per hour of operation considering installation costs as well as maintenance and operational costs over the lifetime period of both types of technology. It was calculated that 1 h of operation of the RTO implies a cost of 137 € while for the eight recuperative thermal oxidizers, the costs reach 305 €. Considering all selected relevant impact categories (Fig. 4 (a)), it can be recognized that the installation of the recuperative thermal oxidizers to reduce formaldehyde emissions is the most environmentally sustainable alternative, although the costs are 2.2 times those of the RTO. On the other hand, the most economically favorable abatement technology is the installation of a regenerative thermal oxidizer.

Regarding the reduction of the HT impact category, the two technologies can also be compared from the perspective of eco-efficiency in Fig. 4 (b). This graph clearly illustrates the difference in the reduction of the human toxicity achieved by both technologies considering their implementation costs. Even though a reduction is obtained by both technologies, the costs to gain a reduction of more than 10% of the HT impact category for the topcoat process are much higher for the implementation of the eight recuperative thermal oxidizers (305 €/h and 137 €/h, respectively). From these results, it can be concluded that the implementation of the recuperative thermal oxidizers is a better solution if a higher reduction of the human toxicity is pursued and the higher costs are not a relevant factor.

4. Limitations of the study and future research

The distinctive characteristic of this work is the fact that all data was collected under the operating conditions of a real vehicle paint shop. This has by nature several limitations that are intrinsic to this kind of studies. On the other hand, the scarcity of existing publications on LCA and eco-efficiency analysis for vehicle paint shops and their air emissions abatement equipment hinders the comparison and validation of the results obtained in this work with the set up and outcomes of other studies.

Even though the percentage of paint shops in the EU using solvent-based paints is still high, efforts from vehicle manufacturers have been observed in the last years to reduce the usage of solvent-based coatings and thus reduce their VOC emissions (ACEA, 2022). Consequently, the number of paint shops using water-based paints is constantly increasing. However, the complete retrofit of a paint shop from solvent to water-based paints is usually not possible due to the bigger spaces needed for painting lines with waterborne materials. Therefore, the change from a solvent to a water-based paint shop is usually only executed when the vehicle manufacturing site has sufficient space to install a completely new paint shop. This study was carried out in a paint shop using solvent-based paints. A similar study might be conducted for a water-based paint shop in future research to evaluate the differences

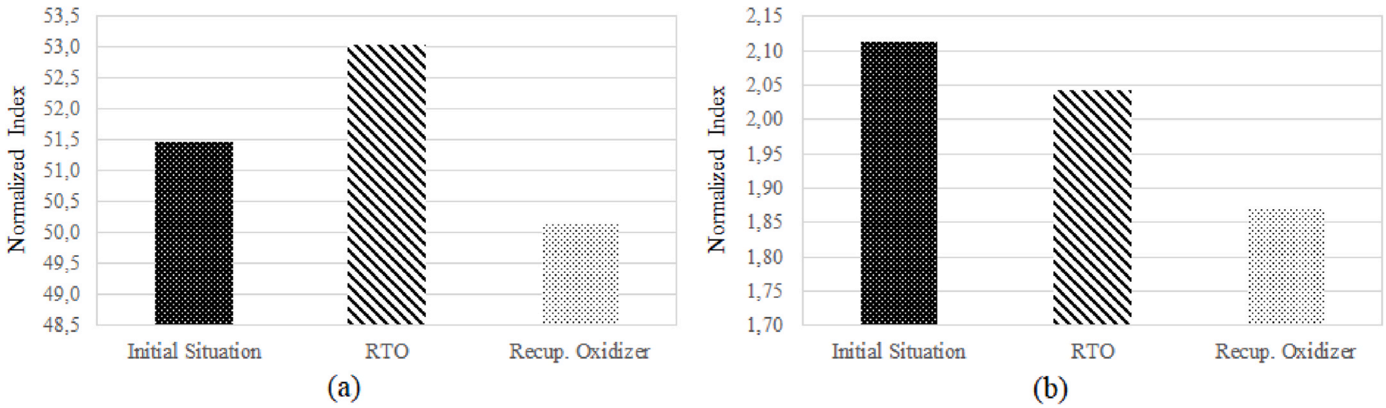


Fig. 3. Results of the normalization. (a) Topcoat process: selected relevant impact categories; (b) Topcoat process: Human Toxicity.

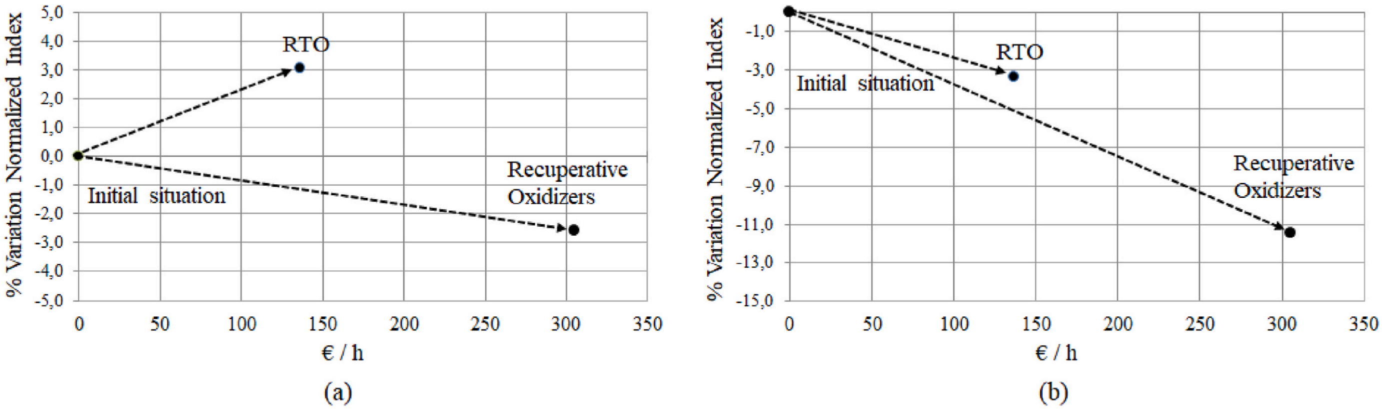


Fig. 4. Results of the eco-efficiency analysis. (a) Topcoat process: selected relevant impact categories; (b) Topcoat process: Human Toxicity.

and confirm the results of this work in another paint shop.

Furthermore, a growing number of paint shops are starting to install new paint shop concepts that considerably contribute to energy savings. Several plants have eliminated in the last 10–15 years the primer oven by applying the three-wet concept (Silva Cavalcante et al., 2020). In this process, the application of primer, basecoat and clearcoat is performed without intermediate drying oven, with the consequent energy saving (STS BREF, 2020). In this study, the paint shop analyzed applied the conventional painting steps with an intermediate curing step of the primer surfacer before the application of the basecoat paint. A future investigation could also be undertaken in a paint shop that applies the three-wet concept.

Finally, one additional improvement that can be examined in a future study is the introduction of heat recovery at the stacks of the RTO in this paint shop. The energy surplus of the eight recuperative thermal oxidizers considered was recovered to heat the drying ovens. However, with regard to the RTO, the energy excess of the hot cleaned gases from this abatement system was not recovered to be used in other paint shop processes. This possibility was disregarded by the paint operator due to space, timing and monetary limitations. Nevertheless, future research could include the addition of heat exchangers to recover the energy excess from the stacks of the RTO, decreasing the overall energy consumption of the paint shop.

5. Conclusions

Life Cycle Assessment (LCA) was applied to compare two different abatement technologies currently used in vehicle paint shops across the world for the elimination of VOCs and, particularly, formaldehyde emissions. While both abatement technologies achieved a decrease in the human toxicity due to the reduction of formaldehyde emissions, the implementation of recuperative thermal oxidizers reached a more significant human toxicity reduction than the regenerative thermal oxidation technology (11.5% and 3.4% reduction of the normalized index in the HT impact category, respectively). Moreover, the recuperative thermal oxidizers achieved an overall improvement when considering the most relevant impact categories selected in this study, unlike the regenerative thermal oxidizer (2.6% reduction of the normalized index observed for the recuperative thermal oxidizers against 3.1% increase for the regenerative thermal oxidizer). The differences can be mainly explained by the higher overall energy consumption of the latter technology. Thus, the installation of the recuperative thermal oxidizers is the most environmentally sustainable alternative.

The eco-efficiency analysis shows that in cases where only the reduction of the formaldehyde emissions along with an overall improvement of the human toxicity are pursued and the costs are the main factor in the decision-making process, the regenerative thermal oxidizer is the most economically favorable abatement option.

This is the first study comparing different air emissions abatement technologies for vehicle paint shops based on life cycle and eco-efficiency assessments. The results provide new information to vehicle manufacturers and paint shop operators, facilitating the decision-making process in the selection of the most suitable abatement technology for the reduction of formaldehyde emissions. LCA and eco-efficiency analysis have been confirmed as powerful tools that can be applied during the planning and selection activities to evaluate and identify the most environmentally sustainable technology.

CRediT authorship contribution statement

Daniel Granadero: Methodology, Formal analysis, Investigation, Writing – Original Draft, Visualization. Aida Garcia-Muñoz: Formal analysis, Investigation. Renate Adam: Resources, Supervision, Writing – Review & Editing. Francisco Omil: Supervision, Validation. Gumersindo Feijoo: Conceptualization, Supervision, Validation, Writing – Review & Editing.

Declaration of competing interest

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

Data availability

Data will be made available on request.

Acknowledgements

The authors Daniel Granadero, Gumersindo Feijoo and Francisco Omil belong to the Galician Competitive Research Groups (GRC) ED431C-2021/37.

References

ACEA Automobile Industry Pocket Guide 2022-2023 European automobile manufacturers' association (ACEA). https://www.acea.auto/publication/automobile-industry-pocket-guide-2022-2023/.

Akafuah, N.K., Poozesh, S., Salaimeh, A., Patrick, G., Lawler, K., Saito, K., 2016. Evolution of the automotive body coating process—a review. Coatings 2016 6 (2), 24. https://doi.org/10.3390/COATINGS6020024.

Banar, M., Çokaygil, Z., 2010. Life Cycle Assessment of electricity production from natural gas combined cycle. Chemical Engineering Transactions 21, 157–162. https://doi.org/10.3303/cet1021027.

Berenjian, A., Chan, N., Malmiri, H.J., 2012. Volatile organic compounds removal methods: a review. Am. J. Biochem. Biotechnol. 8, 220–229. https://doi.org/10.3844/ajbbsp.2012.220.229.

Bianco, I., Panepinto, D., Blengini, G.A., Onofrio, M., Zanetti, M., 2020. Inventory and life cycle assessment of an Italian automotive painting process. Clean Technol. Environ. Policy 22, 247–258. https://doi.org/10.1007/s10098-019-01780-3.

Borwankar, D.S., Anderson, W.A., Fowler, M., 2012. A technology assessment tool for evaluation of VOC abatement technologies from solvent based industrial coating operations. Air Qual. - New Perspect. https://doi.org/10.5772/45745.

Bouter, A., Guichet, X., 2022. The greenhouse gas emissions of automotive lithium-ion batteries: a statistical review of life cycle assessment studies. J. Clean. Prod. 344, 130994 https://doi.org/10.1016/j.jclepro.2022.130994.

Brummer, V., Yong Teng, S., Jecha, D., Skryja, P., Vavrcikova, V., Stehlik, P., 2022. Contribution to cleaner production from the point of view of VOC emissions abatement: a review. J. Clean. Prod. 361, 132112 https://doi.org/10.1016/j.jclepro.2022.132112.

Bysko, S., Krystek, J., Bysko, S., 2020. Automotive paint shop 4.0. Comput. Ind. Eng. 139, 105546 https://doi.org/10.1016/j.cie.2018.11.056.

STS BREF. European Commission, Joint Research Centre (JRC), Chronopoulos, G., Cakmak, G.-E., Tempany, P., Klein, G., Brinkmann, T., Zerger, B., Roudier, S., 2020. Best Available Techniques (BAT) Reference Document on Surface Treatment Using Organic Solvents Including Preservation of Wood and Wood Products with Chemicals: Industrial Emissions Directive 2010/75/EU (Integrated Pollution Prevention and Control). Publications Office. https://data.europa.eu/doi/10.2760/857.

da Silva, L.F., Barbosa, A.D., de Paula, H.M., Romualdo, L.L., Andrade, L.S., 2016. Treatment of paint manufacturing wastewater by coagulation/electrochemical methods: proposals for disposal and/or reuse of treated water. Water Res. 101, 467–475. https://doi.org/10.1016/j.watres.2016.05.006.

Debnath, N.C., 2013. Importance of surface preparation for corrosion protection of automobiles. J. Surf. Eng. Mater. Adv. Technol. 94–105. https://doi.org/10.4236/jsemat.2013.31A014.

Desli, E., Gkoulgkoutsika, A., Sdrolia, E., Zarotiadis, G., 2021. Eco-efficiency: A methodological framework and assessment. Cleaner Environmental Systems 3, 100049. https://doi.org/10.1016/j.cesys.2021.100049.

Directive 2010/75/EU of the European Parliament and of the Council of 24 November 2010 on industrial emissions (integrated Pollution Prevention and Control), 2010. Official Journal of the European Union. http://data.europa.eu/eli/dir/2010/75/oj.

Finlayson-Pitts, B.J., Pitts, J.N., 2000. Chapter 1 - overview of the chemistry of polluted and remote atmospheres. In: Chemistry of the Upper and Lower Atmosphere. Academic Press, pp. 1–14. https://doi.org/10.1016/B978-012257060-5/50003-4.

Gebler, M., Cerdas, J.F., Thiede, S., Herrmann, C., 2020. Life cycle assessment of an automotive factory: identifying challenges for the decarbonization of automotive production – a case study. J. Clean. Prod. 270, 122330 https://doi.org/10.1016/j.jclepro.2020.122330.

Geldermann, J., Rentz, O., 2005. Multi-criteria analysis for technique assessment case study from industrial coating. J. Ind. Ecol. 9 (3), 127–142. https://doi.org/10.1162/1088198054821591.

German Environment Agency, 2023. Umweltbundesamt, Nationale Trendtabellen für die deutsche Berichterstattung atmosphärischer Emissionen seit 1990, Emissionsentwicklung 1990 bis 2021. Stand 03/2023). https://www.umweltbundesamt.de/daten/luft/luftschadstoff-emissionen-in-deutschland/emission-fluechtiger-organischer-verbindungen-ohne#entwicklung-seit-1990.

Giampieri, A., Ling-Chin, J., Ma, Z., Smallbone, A., Roskilly, A.P., 2020. A review of the current automotive manufacturing practice from an energy perspective. Appl. Energy 261, 114074. https://doi.org/10.1016/J.APENERGY.2019.114074.

Giampieri, A., Ma, Z., Ling-Chin, J., Smallbone, A.J., Roskilly, A.P., 2022. A techno-economic evaluation of low-grade excess heat recovery and liquid desiccant-based temperature and humidity control in automotive paint shops. Energy Convers. Manag. 261, 115654 https://doi.org/10.1016/j.enconman.2022.115654.

Goedkoop, M., Heijungs, R., Huijbregts, M., De Schryver, A., Struijs, J., van Zelm, R., 2013. ReCiPe 2008, A Life Cycle Impact Assessment Method Which Comprises Harmonised Category Indicators at the Midpoint and the Endpoint Level. Ministerie van VROM Rijnstraat.

Guo, W., Feng, T., Li, W., Hua, L., Meng, Z., Li, K., 2023. Comparative life cycle assessment of sodium-ion and lithium iron phosphate batteries in the context of carbon neutrality. J. Energy Storage 72, 108589. https://doi.org/10.1016/j.est.2023.108589. D.

Hernandez, M., Messagie, M., Hegazy, O., Marengo, L., Winter, O., Van Mierlo, J., 2017. Environmental impact of traction electric motors for electric vehicles applications. Int. J. Life Cycle Assess. 22, 54–65. https://doi.org/10.1007/s11367-015-0973-9.

ILCD, 2010a. Analysis of Existing Environmental Impact Assessment Methodologies for Use in Life Cycle Assessment. Background Document. European Commission. Joint Research Centre. Institute for Environment and Sustainability.

ILCD, 2010 b. Framework and Requirements for Life Cycle Impact Assessment Models and Indicators. Joint Research Centre. Institute for Environment and Sustainability. European Commission.

ISO, 2006a. 2006 Environmental Management. Life Cycle Assessment, Principles and Framework. International Organisation Standardisation, 14040.

ISO, 2006b. 2006 Environmental Management. Life Cycle Assessment, Requirements and Guidelines. International Organisation Standardisation, 14044.

ISO, 2012. 2012 Environmental Management. Eco-Efficiency Assessment of Product Systems. Principles, Requirements and Guidelines. International Organisation Standardisation, 14045.

Jurgetz, A., 1995. Automotive paint performance. Met. Finish 93, 53–55. https://doi.org/10.1016/0026-0576(96)80432-6.

Kim, J.-A., Kim, S., Kim, H.-J., Kim, Y.-S., 2011. Evaluation of formaldehyde and VOCs emission factors from paints in a small chamber: the effects of preconditioning time and coating weight. J. Hazard Mater. 187 (1–3), 52–57. https://doi.org/10.1016/j.jhazmat.2010.10.094.

Klöpffer, W., Grahl, B., 2014. Life cycle assessment (LCA): a guide to best practice. Life cycle assess. A Guid. to Best Pract. 1–396. https://doi.org/10.1002/9783527655625.

Li, D., Wang, L., Lu, Y., Deng, H., Zhang, Z., Wang, Y., Ma, Y., Pan, T., Zhao, Q., Shan, Y., Shi, X., Ma, J., He, H., 2023. New insights into the catalytic mechanism of VOCs abatement over Pt/Beta with active sites regulated by zeolite acidity. Appl. Catal. B Environ. 334, 122811 https://doi.org/10.1016/j.apcatb.2023.122811.

Lopes Silva, D.A., De Oliveira, J.A., Padovezi Filleti, R.A., Gomes de Oliveira, J.F., Da Silva, E.J., Ometto, A.R., 2018. Life Cycle Assessment in automotive sector: a case study for engine valves towards cleaner production. J. Clean. Prod. 184, 286–300. https://doi.org/10.1016/j.jclepro.2018.02.252.

Marder, A.R., Goodwin, F.E., 2023. Chapter 12 - Zn-coated steel product properties. In: Woodhead Publishing Series in Metals and Surface Engineering, the Metallurgy of Zinc Coated Steels. Elsevier, pp. 397–455. https://doi.org/10.1016/B978-0-323-99984-7.00014-2.

McMahon, M.E., Kammrath, B.W., Bender, L., B-C, 2023. Automotive paint. In: Encyclopedia of Forensic Sciences, third ed. Elsevier, pp. 240–252. https://doi.org/10.1016/B978-0-12-823677-2.00221-X. 1.

Mulholland, K.L., Dyer, J.A., 1999. Pollution Prevention: Methodology, Technologies, and Practices. American Institute of Chemical Engineers.

Nair K, S., Manu, B., Azhoni, A., 2021. Sustainable treatment of paint industry wastewater: current techniques and challenges. J. Environ. Manag. 296, 113105 https://doi.org/10.1016/j.jenvman.2021.113105.

Nayane de Queiroz, M., de Oliveira Lima, A.M., Gomez Winkler, M.E., Hafemann Fragal, V., Forti Rubira, A., Sequinel, T., da Silva Ribeiro, L., de Souza Neto, F.N., Rodrigues Camargo, E., Ferreira Arlindo, M.Z., Saraiva Ogrodowski, C., Gorup, L.Z., 2022. Chapter 20 - conductive nanopaints: a remarkable coating. In: Micro and Nano Technologies, Nanotechnology in the Automotive Industry. Elsevier, pp. 429–449. https://doi.org/10.1016/B978-0-323-90524-4.00020-7.

Onofre, A., Godina, R., Carvalho, H., Catarino, I., 2020. Eco-innovation in the cleaning process: an application of dry ice blasting in automotive painting industry. J. Clean. Prod. 272, 122987 https://doi.org/10.1016/j.jclepro.2020.122987.

Ou, R., Chang, C., Zeng, Y., Zhang, X., Fu, M., Fan, L., Chen, P., Ye, D., 2022. Emission characteristics and ozone formation potentials of VOCs from ultra-low-emission waterborne automotive painting. Chemosphere 305, 135469. https://doi.org/10.1016/j.chemosphere.2022.135469.

Papasavva, S., Kia, S., Claya, J., Gunther, R., 2001. Characterization of automotive paints: an environmental impact analysis. Prog. Org. Coating 43, 193–206. https://doi.org/10.1016/S0300-9440(01)00182-5.

Pizzi, A., Ibeh, C.C., 2022. Chapter 4 – aminoresins. In: Handbook of Thermoset Plastics, fourth ed. William Andrew Publishing, pp. 65–82. https://doi.org/10.1016/B978-0-12-821632-3.00003-8.

Poth, U., 2008. Automotive coatings formulation: chemistry. Physics und Practices.. Vincentz Network, Hannover, pp. 86–90. ISBN 9783866309043, 386630904X.

Ramírez-Melgarejo, M., Güereca, L.P., Gassó-Domingo, S., Salgado, C.D., Reyes-Figueroa, A.D., 2021. Eco-efficiency evaluation in wastewater treatment plants considering greenhouse gas emissions through the data envelopment analysis-tolerance model. Environ. Monit. Assess. 193, 301. https://doi.org/10.1007/s10661-021-09063-5.

Regulation (EC) No 1272/2008 of the European Parliament and of the Council of 16 December 2008 on classification, labelling and packaging of substances and mixtures (EC) No 1907/2006 Amending and Repealing Directives 67/548/EEC and 1999/45/EC, and Amending Regulation, 2008. Official Journal of the European Union. http://data.europa.eu/eli/reg/2008/1272/oj.

Regulation (EC) No 1907/2006 of the European Parliament and of the council of 18 December 2006 concerning the Registration Evaluation, Authorisation and Restriction of Chemicals (REACH), Establishing a European Chemicals Agency, Amending Directive 1999/45/EC and Repealing Council Regulation (EEC) No 793/93 and Commission Regulation (EC) No 1488/94 as Well as Council Directive 76/769/EEC and Commission Directives 91/155/EEC, 93/67/EEC, 93/105/EC and 2000/21/EC, 2006. Official Journal of the European Union.

Regulation (EU) No 2015/491 of the European Parliament and of the Council of 23 March de 2015 amending Regulation (EU) No 605/2014 amending, for the purposes of introducing hazard and precautionary statements in the Croatian language and its adaptation to technical and scientific progress, Regulation (EC) No 1272/2008 of the European Parliament and of the Council on classification, labelling and packaging of substances and mixtures, 2015. Official Journal of the European Union. http://data.europa.eu/eli/reg/2015/491/oj.

Regulation (EC) No 1272/2008 of the European Parliament and of the Council on classification, labelling and packaging of substances and mixtures Regulation (EU) No 605/2014 of 5 June 2014 ("6. ATP") Amending, for the Purposes of Introducing Hazard and Precautionary Statements in the Croatian Language and its Adaptation to Technical and Scientific Progress, 2014. Official Journal of the European Union. http://data.europa.eu/eli/reg/2014/605/oj.

Rivera, J.L., Reyes-Carrillo, T., 2014. A framework for environmental and energy analysis of the automobile painting process. Procedia CIRP 15, 171–175. https://doi.org/10.1016/j.procir.2014.06.022.

Rivera, J.L., Reyes-Carrillo, T., 2016. A life cycle assessment framework for the evaluation of automobile paint shops. J. Clean. Prod. 115, 75–87. https://doi.org/10.1016/j.jclepro.2015.12.027.

Sala-Garrido, R., Maziotis, A., Mocholi-Arce, M., Molinos-Senante, M., 2023. Assessing Eco-Efficiency of Wastewater Treatment Plants: A Cross-Evaluation Strategy, vol. 900. Science of The Total Environment, 165839. https://doi.org/10.1016/j.scitotenv.2023.165839.

Salthammer, T., 2019. Formaldehyde sources, formaldehyde concentrations and air exchange rates in European housings. Build. Environ. 150, 219–232. https://doi.org/10.1016/j.buildenv.2018.12.042.

Salthammer, T., Mentese, S., Marutzky, R., 2010. Formaldehyde in the indoor environment. Chem. Rev. 110, 2536. https://doi.org/10.1021/CR800399G.

Shamoon, A., Haleem, A., Bahl, S., Javaid, M., Garg, S.B., Sharma, R.C., Garg, J., 2022. Environmental impact of energy production and extraction of materials - a review. Mater. Today: Proc. 57–2, 936–941. https://doi.org/10.1016/j.matpr.2022.03.159.

Shi, Y., Xi, Y., Lv, D., Simayi, M., Liang, Y., Ren, J., Xie, S., 2023. Sector-based volatile organic compound emission characteristics and reduction perspectives for coating materials manufacturing in China. J. Clean. Prod. 394, 136407 https://doi.org/10.1016/j.jclepro.2023.136407.

Silva Cavalcante, E., Sales Vasconcelos, L.G., de Farias Neto, G.W., Brandão Ramos, W., Pereira Brito, R., 2020. Automotive painting process: minimizing energy consumption by using adjusted convective heat transfer coefficients. Prog. Org. Coating 140, 105479. https://doi.org/10.1016/j.porgcoat.2019.105479.

Skone, T.J., Littlefield, J., Marriott, J., Cooney, G., Demetrion, L., Jamieson, M., Jones, C., Mutchek, M., Shih, C.Y., Schivley, G., Krynock, M., 2016. Life Cycle Analysis of Natural Gas Extraction and Power Generation. U.S. Department of Energy, pp. 1–79. https://doi.org/10.2172/1480993.

Sorrels, J.L., Baynham, A., Randall, D., Hancy, C., 2017. Incinerators and Oxidizers. U.S. Environmental Protection Agency (EPA), Research Triangle Park, NC 27711, pp. 1–70.

Stoye, D., Freitag, W., 1998. Paints, coatings and solvents. In: 2. Completely Rev. Wiley-VCH.

Streitberger, H.-J., Dössel, K.-F., 2008. Automotive Paints and Coatings. Second, Completely Revised and Extended Edition. Wiley-VCH.

Tomatis, M., Moreira, M.T., Xu, H., Deng, W., He, J., Parvez, A.M., 2019. Removal of VOCs from waste gases using various thermal oxidizers: a comparative study based on life cycle assessment and cost analysis in China. J. Clean. Prod. 233, 808–818. https://doi.org/10.1016/j.jclepro.2019.06.131.

Torregrossa, D., Leopold, U., Hernández-Sancho, F., Hansen, J., 2018. Machine learning for energy cost modelling in wastewater treatment plants. J. Environ. Manag. 223, 1061–1067. https://doi.org/10.1016/j.jenvman.2018.06.092.

VDI 3862 Part 2:2000-12. VDI/DIN-Kommission Reinhaltung der Luft (KRdL) – Normenausschuss, 2000. Gaseous emission measurement - Measurement of aliphatic and aromatic aldeydes and ketones by DNPH method - Impinger method.

VDI guideline 3455:2013-08. VDI/DIN-Kommission Reinhaltung der Luft (KRdL) – Normenausschuss, 2013. Emission control. High-volume car body painting plants.

Wilson, J.B., 2009. Resins: A Life-Cycle Inventory of Manufacturing Resins Used in the Wood Composites Industry. Department of Wood Science and Engineering, Oregon State University, Corvallis. OR 97331-5751.

Winjobi, O., Kelly, J.C., Dai, Q., 2022. Life-cycle analysis, by global region, of automotive lithium-ion nickel manganese cobalt batteries of varying nickel content. Sustainable Materials and Technologies 32, e00415. https://doi.org/10.1016/j.susmat.2022.e00415.

Xu, J., Tai, X., Betha, R., He, J., Balasubramanian, R., 2015. Comparison of physical and chemical properties of ambient aerosols during the 2009 haze and non-haze periods in Southeast Asia. Environ. Geochem. Health 37, 831–841. https://doi.org/10.1007/s10653-014-9667-7.

Xu, J., Xu, H., Xiao, H., Tong, L., Snape, E., Wang, C., He, J., 2016. Aerosol composition and sources during high and low pollution periods in Ningbo, China. Atmos. Res. 178–179, 559–569. https://doi.org/10.1016/j.atmosres.2016.05.006.

Yang, C., Miao, G., Pi, Y., Xia, Q., Wu, J., Li, Y., Xiao, J., 2019. Abatement of various types of VOCs by adsorption/catalytic oxidation: a review. Chem. Eng. J. 370, 1128–1153. https://doi.org/10.1016/j.cej.2019.03.232.