Fluorescent lamps: A review on environmental concerns and current recycling perspectives highlighting Hg and rare earth elements

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ABSTRACT

The presence of Hg and the high consumption of fluorescent lamps (FL) have increased environmental concerns. However, other toxic and potentially toxic elements, like rare earth elements (REE), are neglected in FL. The problems FL cause are currently less in focus since they are being replaced with light emitter diodes (LED). However, replacement is lagging mainly in developing countries, which are usually more affected by electronic wastes. Therefore, it can take many years for FL to be completely phased-out and no longer be a matter of environmental concern. It is essential to keep the FL issue under our attention, considering current consumption and disposal scenarios and the lamps' composition, as demonstrated through life cycle assessment (LCA) works. This revision summarizes the information available in the literature, focusing on 2010–2020, on the current scenario of FL regarding its replacement, LCA, contamination risks, disposal, and recycling, highlighting the areas that still require attention. The Hg in lamps can reach the order of mg per lamp. However, Cd, As, Pb, and other potentially toxic metals are also found in FL. In fact, REE have high concentrations in FL, representing more than 23 % of the phosphor, and these emergent contaminants are unregulated and have poorly understood environmental effects. Recycling of FL is often low or even inexistent among countries. However, authors have proposed efficient methods for recovering metals (>90 % efficiency) and using end-of-life FL in manufacturing new materials. This might be a silver lining to the critical contamination problem arising from FL.

1. Introduction

After the energy crisis in the early 2000 s, the replacement of incandescent lamps (IL) with fluorescent lamps (FL) has intensified due to the greater energy efficiency of FL. IL are known for their higher energy consumption, generation of a significant amount of heat, and shorter life span, which motivated their replacement. FL have now been suppressed by light emitter diodes (LED) in terms of energy efficiency. Nevertheless, FL are still highly used, especially in developing countries, and are responsible for large amounts of waste [7,40,84,95,121].

FL are filled with inert gas and contain up to 15 mg of mercury, partially in vapor form. When an electric current is applied to the

electrodes, the mercury vapor is excited and, upon return to the ground state, emits ultraviolet radiation (UV). The high efficiency of FL is possible thanks to the fluorescence emitted by the phosphor material that coats the glass and is responsible for converting the UV into visible light [40,55,92,148]. The phosphor material was developed in the 1970 s, consisting of phosphors doped with rare earth elements (REE), emitting light in a specific wavelength of the visible spectrum. They were named according to the color emitted (red: Y_2O_3:Eu^{3+}; blue: $BaMgAl_{10}O_{27}$:Eu^{2+} and $(Sr,Ca,Ba)_5(PO_4)_3Cl$:Eu^{2+}; and green: $CeMgAl_{11}O_{19}$: Tb^{3+} and $LaPO_4$:Ce^{3+}, Tb^{3+}) and when mixed in the correct quantities, produce high-quality, powerful white light. They are used in patented compositions mixed with halophosphate and correspond to 3–5% of the lamp weight [181]. Lead, nickel, arsenic, chromium, and gadolinium can also be found in FL in lower amounts [55,97,128,155,165,179].

The low energy consumption of FL is an advantage relative to a few other electrical power supplies; nevertheless, they can be a source of environmental contamination if inappropriately disposed of [44,73]. In 2007, 397 million compact fluorescent lamps (CFL) were sold in the United States and about 288 million units in Europe [150,162]. In Brazil, around 206 million units of FL were discarded every year around 2014 [13]. In 2011, the production of FL in China reached 7 billion units, but the annual production fluctuated between 3 and 4 billion around 2014 [131,158]. In South Korea, around 2.16 million units of FL were produced in 2017 [182]. A decrease in the production of FL has been observed in recent years: 35 million units entering the Brazilian market in 2019 as opposed to 112.1 million in 2018 [136]; 524 million units being exported from China in the first quarter of 2020, a decrease of approximately 35 % compared to the previous year [69]; estimation of roughly 300 million units being sell in Europe in 2021 (among CFL without integrated ballast and linear fluorescent lamps (LFL)) [49], with 61 % of the exports from France, The Netherlands, Poland, Germany, and Hungary being within European countries and 39 % for non-European countries [35]. Nevertheless, FL contributed the largest market share (52.03 %) in 2019 [112]. According to the International Energy Agency lighting report [76], FL accounted for 33 % of global lighting sales in 2020. Being Europe and the United States responsible for the rapid LED expansion. However, LED penetration is uneven across markets. In India, IL and FL still account for 54 % of the lighting market [27]. FL accounted for 51.03 % of the market in China in 2019 [58]. Despite being replaced by LED, the global production of FL is estimated to be approximately 14 million units by 2025, according to the Global Fluorescent Lighting Market Research Report by Market Research Future [110].

Most spent FL are not disposed of safely, even in developed countries [115]. According to the Report to the Committee on the Regulation of Hazardous Substances by the Swedish Energy Agency and Collaborative Labeling and Appliance Standards Program (CLASP) [18], more than half of the FL sold in Europe are discarded in regular municipal waste. The high reported recycling rates in Europe are not based on the total number of lamps removed from service but only on the ones delivered to the correct waste treatment facility. Brazil had a recycling target of 60 million units by 2021 (corresponding to 20 % of the units put on the market in 2012). By 2019, 7.1 million units (11 % of the target value) were recycled, and most spent FL are still disposed of in municipal landfills and open dumps [31,136]. In Beijing (China), 70 % of spent FL are sent to landfills or incinerators [31,131]. In the Philippines, around 50 million pieces of FL are discarded annually, and 84 % are disposed of in landfills and open dumpsites in private or public vacant lots [78]. In Sri Lanka, waste collection is still not very efficient, and the country has only one recycling company registered for recycling CFL [114]. Recycling rates are still being reported as 23 % for the United States, 7 % for Canada, and < 10 % for Japan [31,47,93].

Contamination of environmental compartments with metal pollutants is a current matter of concern. Human activities are one of the sources of metal pollutants entering the biosphere [37]. Since heavy metals are not biodegradable, bioaccumulation is observed with latency

periods of decades [113]. As mentioned, Hg and REE are metals found in high concentrations in FL. Mercury is one of the most toxic pollutants in the environment. Although it can occur naturally, anthropogenic emissions are responsible for most environmental Hg pollution [91]. Mercury can reach remote areas of the planet; contaminate water, soil, and atmosphere; and bioaccumulate and biomagnify in food chains, posing a threat to animals and humans due to its toxicity even at low concentrations [34,64,88,171]. REE are naturally occurring metals that include lanthanides, yttrium, and scandium [14,67]. Due to their unique magnetic properties, industrial application of REE has significantly improved, being recognized as essential to the high-tech and renewable energy industries [6,41,149,173,177]. REE are considered emerging pollutants. Their high application in technology has resulted in considerable local emissions related to mining and enrichment, which contributes to an increase in diffuse pollution in industrial areas, raising concerns about chronic toxicity [177]. Studies on their ecotoxicological behavior are increasing; however, there is still much to understand about their bioavailability, ecotoxicity, and mechanisms of leaching and mobility between environmental compartments [14,51]. Prolonged exposure to REE can cause adverse health effects and bioaccumulation [63,140]. Studies with aquatic organisms revealed that exposure to lanthanides could affect skeletal growth, reproductive performance, and fertilization [111].

The environmental impacts associated with the production, usage, and discard of FL can be determined with the life cycle assessment (LCA), which is an analytical method that allows evaluating the environmental profile of a product, process, or activity. This tool enables the identification and estimation of materials and energy employed in a production process and the waste released into the environment during the product's life cycle. Therefore, LCA can be a powerful ally in product optimization and environmental studies, as it allows the identification of weaknesses and improvement of the processes for economic and environmental purposes [11,87,89]. Regarding FL, it allows the evaluation, for example, of the reduction of environmental impacts by recycling them. Spent FL can be used in metal recovery processes, preventing metals from being released into the environment, and for economic purposes, allowing the reuse of resources of industrial interest [11,28,80,82,159]. Currently, different methodologies are developed for recovering elements from FL [43,100,164]. For instance, there are methods in place for recovering Hg from FL, a practice required by legislation in many countries [93]. Although applied on an insufficient scale for global demand, such procedures reduce the release of Hg into the environment. However, the recovery of REE from FL have been attracting more interest. They are considered critical metals by different countries since they are mostly irreplaceable for the manufacture of high technology products and green energy advances such as displays, fiber optics, wind turbines, and batteries for electric cars [20,42,48,53,149]. Therefore, the recovery of these metals from secondary sources is a critical matter considering their global monopoly by China and the consequent lack of REE supply that non-producer countries can face in case the demand for REE increases in the future [20,80,127]. The chemical company Solvay-Rhodia already practiced the recovery of REE from end-of-life FL by opening two industrial-scale plants in 2011. However, it had to be closed following the drop in the prices of REE [134]. Considering the fluctuations of the market of REE, prices might rise again, as is currently observed according to Patil et al. [127]. New recovery plants may emerge, and the knowledge acquired by recovery investigations of REE could help enforce more efficient recovery pathways.

2. Why still discuss fluorescent lamps?

In recent decades, there has been an undeniable growth in the LED market, which has directly affected the production, commercialization, and sale of FL. Despite the reduction in the market of FL and the policies already in place for the phasing out of these lamps in several countries,

FL are still produced, and it will take years to stop their waste generation (complete replacement of FL with LED). It is important to emphasize that recycling is far behind in developing countries compared to developed countries, and environmental issues are a global problem, as we are not isolated on the planet.

The primary concern related to FL is the presence of Hg; however, other toxic elements present, such as As, Pb, and Cd, also require attention. In addition, the presence of REE in lamps, which are emerging and potentially toxic contaminants, is neglected regarding environmental and human health risks. Some effects of these metals have been reported in studies. However, little is known about the long-term effects and possible interactions, raising concerns regarding the disposal of fluorescent lamps. There is still a high annual discard of FL, and the disposal of this waste is not always done correctly. Recycling rates are still low in many countries, disposal in open dumps still occurs, and many people are unaware of the dangers of this material. Some recycling methodologies are only concerned with eliminating Hg and neglect other metals. Furthermore, FL are excellent secondary sources for obtaining REE, and this potential is under-exploited. These matters should not be overlooked due to LED lamps' gradual replacement of FL. Fluorescent lamps represent both an environmental problem and a source of desired raw material; therefore, they demand our attention.

This review aims to highlight the contamination risks and environmental and human impacts that can arise from the inadequate discard of the lamps through life cycle assessment and metal quantification studies. We emphasize the dangers of FL even today, and we demonstrate that FL are far from ceasing to be an environmental problem and that improvements still need to be made in public policies for handling and recycling FL, which are issues that not everyone realizes. Our focus goes beyond Hg to other toxic, potentially toxic, and unregulated elements. The discrepancy between the volume of works related to Hg in FL in relation to other metals is evident in the literature, especially from an environmental point of view. Elements such as Cd, Pb, As, Mn, and REE, which are also present in the phosphor, are neglected. The dangers related to the presence of these elements in FL demand further studies. And the behavior and effects of REE, in particular, is an area of research that needs growth.

Furthermore, we review methodologies employed in recycling and recovering REE and Hg from FL, which are areas of interest from both environmental and economic points of view. The recovery of metals of economic interest from hazardous waste through methods of viable application has the potential to transform what would be a sanitation problem into a solution for the industry and technology sectors. Further research in this area, aiming to improve recovery methodologies and allow a large-scale application, are also essential.

The research parameters employed in the review process are described in Topic S1 in the supplementary material.

3. Metal contamination associated with fluorescent lamps

The high metal content in FL is a cause of concern due to its potential toxicity. In acknowledgment of this, many studies have been conducted over the years to exploit environmental contamination and threat to human health. Due to a relatively recent recognition of REE as emerging contaminants and the lower concentration of other metals in FL, most works regarding the hazards associated with metals in FL focus on mercury.

3.1. Mercury

3.1.1. Human exposure

Fluorescent lamps can be responsible for emitting mercury into the atmosphere. In China, FL are responsible for 18 % of atmospheric Hg emissions [147]. Luo et al. [106] evidenced higher levels of total gaseous mercury in industrial areas of China during working hours, although no significant non-carcinogenic risk was found for residents.

However, consumers of FL are still susceptible to Hg exposure in case of indoor lamp breakage, as discussed in the work of Sarigiannis et al. [145], Li and Jin [96], Nance et al. [120], and Salthammer et al. [142]. Sarigiannis et al. [145] reported that most Hg intake occurs in the first hours after lamp breaking. Infants are significantly more exposed because of their breathing area, and higher indoor air temperature increases Hg release. According to Nance et al. [120], most releases do not produce levels of concern (hazard quotients were less than 1), but in some scenarios, risk targets were exceeded, and further studies are needed. Li and Jin [96] observed that broken lamps continuously release Hg vapors for over ten weeks, which can exceed the human exposure limit if the room is poorly ventilated. However, immediate removal of Hg and ventilation could prevent acute health effects [142].

Mercury contamination poses a greater threat to workers in sectors of production, transportation, and recycling of FL. Workers are in constant and close contact with sources of Hg, increasing their exposure risk, and this matter was addressed in multiple works in literature. Zimmermann et al. [187] indicate that workers in the recycling sector in France are exposed to worrying levels of pollutants in all process steps. Individual exposure to mercury in the facility among the processes ranged from 0.1 to 86.7 $\mu g\ m^{-3}$. This can also be observed in the work of Darvishi et al. [29], which assessed the Hg levels in personal, biological, and air samples in a factory of CFL in Iran. The average Hg vapor levels in personal and air samples were 14.78 ± 5.76 and $67.10 \pm 59.37\ \mu g\ m^{-3}$, respectively. The mean urinary mercury level (UML) was 13.85 ± 13.14 $\mu g\ g^{-1}$ creatinine, and 13.6% of workers were above the 20 $\mu g\ g^{-1}$ creatinine level recommended by the Centre of Environmental and Occupational Health in Iran, demonstrating the contamination potential. Al-Batanony et al. [2] also evaluated the Hg occupational exposure in workers of a factory of FL, this time in Egypt. Workers presented higher UML ($44.1 \pm 17.5\ \mu g\ g^{-1}$) than the control group ($6.1 \pm 4.9\ \mu g$ g^{-1}). Some workers had manifestations of Hg toxicity, and deterioration of pulmonary function was observed with the increase of UML and employment duration.

Wilson et al. [176] found Hg levels in the air of a recycling facility of FL exceeding the recommended threshold, reaching a maximum of 207.4 $\mu g\ m^{-3}$ on the floor level and 99.7 $\mu g\ m^{-3}$ at the breathing level. Five of seven workers had elevated Hg levels, and two presented signs of Hg toxicity. Do et al. [36] published a case report on acute Hg poisoning in workers dismantling a factory of FL in Korea. Eighteen of twenty-one workers presented symptoms of Hg poisoning, and ten had persistent symptoms 18 months after exposure.

Gul et al. [60] investigated the inorganic (I-Hg) and methylmercury (Me-Hg) presence in biological samples - red blood cells (RBC), plasma, hair, urine, nails - of workers of fluorescent lamp industries. Results showed that Hg concentrations among these workers were significantly higher than in the control groups. In RBC, plasma, and urine of exposed workers, Me-Hg and I-Hg concentrations were, respectively, up to 27.7 and 137.50 $\mu g\ L^{-1}$. In hair and nails, Me-Hg concentration was, respectively, 3.21 and 5.52 $\mu g\ g^{-1}$, while I-Hg was 0.31 and 0.60 $\mu g\ g^{-1}$.

The surrounding areas of FL and electronic waste (e-waste) production and recycling industries are also at imminent contamination risk. Ni et al. [122] analyzed hair samples of volunteers living near an e-waste recycling industry. Hg concentrations in the hair of exposed volunteers (average 0.99 $\mu g\ g^{-1}$) were higher than those in the control group (average 0.59 $\mu g\ g^{-1}$). In the work of Liang et al. [99], the total Hg (T-Hg) and Me-Hg in the hair of workers and individuals living in the manufacturing area of FL ranged from 143 to 22,800 mg kg^{-1} and from 4.0 to 1190 mg kg^{-1}, respectively. Workers had significantly higher T-Hg concentration in hair than non-workers.

The studies' results corroborate the danger of human contamination by mercury associated with direct contact with FL or areas close to their production and recycling. As mentioned in the work of Calabrese et al. [24], functionality and performance recovery of workers who experienced acute and chronic occupational exposure to Hg over time varies considerably with the individuals. This evokes the necessity of further

improvements in the security of workers and residents of fluorescent lamp handling areas and Hg containment procedures. Many of the works discussed present data on human exposure in both developing and developed countries, indicating a systematic problem. Furthermore, considering that fluorescent lamp recycling plants and facilities presumably have some safety measures in place to prevent the release of Hg and the consequent contamination of their premises, we can imagine the potential risk of exposure from lamps that are unsafely disposed of and handled by people with no knowledge on the matter.

3.1.2. Mercury in FL and environmental samples

Although FL account for only 1 % of total mercury emissions in the United States, these emissions often occur in lamp breakage in households and transportation [96]. The release of mercury vapor from FL, the total concentration of the metal, and sequential extraction of Hg were addressed in the works, among others, of Li and Jin [96], Hobohm et al. [70], De Farias et al. [31], Dos Santos et al. [39], Lecler et al. [92], Rhee et al. [139], Rey-Raap and Gallardo [138], and Viererbl et al. [170]. The analytical details and mean results reported by these studies are summarized in Table S1 in the supplementary material. Only the weak acid fraction of the sequential extraction method (SEM) employed by De Farias et al. [31] was mentioned due to their representation of more immediate forms of environmental contamination. Hg vapor release was estimated with the time approaching infinity in the work of Li and Jin [96]. Reports of acid and neutral extraction performed by Rey-Raap and Gallardo [138] were for 24 h stirring and a liquid/solid ratio of 10.

As presented in Table S1, the reported concentrations for Hg reached 4.83 mg kg^{-1} in glass samples and about 8 mg kg^{-1} in end caps. In the phosphor material and a mixture of glass and phosphor, the concentrations of Hg reached, respectively, more than 1.5 g kg^{-1} and 11.3 mg per lamp. Differences in Hg concentrations among similar fractions are observed in the reviewed studies and can be justified in many ways. The amount of mercury per lamp varies with the brand, country, and year of production due to national regulations aiming the Hg reduction, for example. Darwish et al. [30] observed higher Hg content in Chinese CFL that, in most cases, exceeded the maximum limit recommended by US Environmental Protection Agency (US EPA). Even within a single lamp, variations in results for the replicates can be significant, as observed by Li and Jin [96]. Differences in the sample preparation could affect Hg recovery, also influencing the results. In addition, several factors can influence the distribution of Hg within the lamps. For example, the interactions between Hg and the matrices can increase as a function of the lamp usage time [92]. The amount of Hg in the solid phase increases in spent FL compared to new FL [92]. In the vapor phase, mercury is mostly in Hg^0 form in new FL, while in spent ones, the oxidized forms Hg$^+$ and Hg^{2+} are predominant [92,96]. The majority of Hg in the solid phase is usually found in the phosphor material, as reviewed by Hobohm et al. [70] and shown in the results of Rhee et al. [139] and Lecler et al. [92] for new FL. However, the results Lecler et al. [92] found for spent FL showed higher Hg concentration in end caps instead of phosphor material, indicating once again the variability of Hg distribution inside the lamps. Thermal treatment appears to effectively remove Hg from the phosphor when comparing the results of De Farias et al. [31] with others. De Farias et al. [31] also demonstrated the possible Hg released into the environment due to natural acidic conditions. The more favorable extraction of Hg by acidic conditions was also shown in the results of Ray-Raap and Gallardo [138].

The determination of Hg in areas surrounding landfills and factories of FL has also gained attention. Landfills are still the traditional form of disposal of FL [68,73,98]. In addition, according to Li et al. [98], part of the spent FL is even discarded into the environment. According to the Brazil National Confederation of Municipalities - CNM - approximately 2518 open dumps are functioning in the country and 2257 landfills [22]. It shows how the discard of FL is still a matter of concern. The mercury contained in the lamps (and other solid wastes) can be released into the environment through leaching and gas emissions, contaminating soils

and invertebrates [68]. Shao et al. [148] and Liang et al. [99] observed the presence of Hg in soil and food sources in the surroundings of a factory of FL.

In 2011, mercury contained in fluorescent lamp production in China, which is partially exported, was 29.31 tons, with 20.45 tons being domestically used. Around 3.89 tons of Hg were incinerated, and 15.54 tons were sent to landfills [186]. Facilities and techniques for Hg disposal and recycling are still underdeveloped, and it is necessary to encourage companies' engagement to improve environmental protection [97]. Researchers have been developing techniques to help deal with Hg emissions from FL, and some interesting propositions are summarized in Topic S2 in the supplementary material. Such studies should be incentivized and further investigated for large-scale use.

3.2. Rare earth elements

The extensive application of REE can be an anthropic source of environmental contamination due to e-waste generation and industrial effluents, causing an enrichment in their concentration in waters, soil, and vegetables in upcoming years [63,117]. Morf et al. [118] indicate that the major source of REE is electronic equipment waste and estimate an annual input of REE in solid waste of an incineration plant in Switzerland at 1.6 ± 0.7 t/year for Y, 1.5 ± 0.6 t/year for Nd, 380 ± 170 kg/year for Pr, and 150 ± 35 kg/year for Gd. The REE (Eu, Y, Ce, La, and Tb) are important constituents of lamps, and the high rate of disposal of FL raises concern regarding the REE release into the environment [33]. It is estimated that about 4167 tons of REE were present in scraps of FL in 2020 [83]. This high value serves as an alert for the risk of contaminations resulting from fluorescent lamp wastes, in addition to demonstrating the potential of these wastes as secondary sources of REE. Tunsu et al. [162] found an average concentration of REE in waste of FL after removing large impurities of 105.7 ± 5.3 g kg^{-1}, with Y being the most abundant REE (94.8 ± 4.6 g kg^{-1}). Mishra, Devi, and Sarangi [116] observed that REE represented about 23.13% of the phosphor material, with Y responsible for 20.1%. Pavón et al. [129] found a total concentration of REE in fluorescent lamp waste of 93.2 ± 4.6 g kg^{-1}, with a Y concentration of 84.0 ± 8 g kg^{-1}. The revised works found varying concentrations of REE; however, consistent results regarding the greater abundance of Y among the REE were observed.

Many studies address the leaching of REE from FL, focusing on the recovery of the metals. Authors frequently apply extreme conditions, such as strong acids, pressure, and heat, to achieve extraction [32,71, 172]. However, the use of extreme conditions is not always necessary. Some phosphors can be easily leached under more mild conditions, allowing good recoveries for Y and Eu, as evidenced in the works of Mishra, Devi, and Sarangi [116] and Botelho Junior, Espinosa, and Tenório [21]. REE can be leached from spent FL even under naturally occurring acidic conditions, increasing the possibilities of soil and water contamination due to the inappropriate discard of the lamps. Landfills can act as reservoirs for REE due to the high discard of electronics (including FL). This motivated a few studies of quantification of REE in landfills and surrounding areas from both economic and environmental points of view. Kamura et al. [86] found high concentrations of REE in sections of a landfill in Japan, indicating the potential for contamination with REE through waste but also raising the possibility of using landfills as urban mines to recover rare metals. Nigro et al. [123] observed enrichment in medium REE in the groundwater of a landfill site in Italy. Barbieri et al. [16] found concentrations of pseudo-total REE up to 27.68 mg kg^{-1} and exchangeable REE up to 3.07 mg kg^{-1} in a landfill in Italy.

Despite gaining attention in the last few years, the environmental and health effects related to REE still need to be studied in greater depth, and the studies do not usually focus on a specific source of REE. Therefore, the following studies are not directly linked to FL. It is known that REE are more mobile in solutions rich in F$^-$, Cl$^-$, HCO$_3^-$, CO$_3^{2-}$, HPO$_4^{2-}$, SO$_4^{2-}$, and PO$_4^{3-}$ ions, and, under natural conditions, only small amounts

become available through the atmosphere and groundwater. The expanding industrial use and mining of REE have increased their transport routes into the environment, along with the amount available for bioaccumulation in plants, animals, and humans. Nevertheless, there is still no regulation for the accepted limit of REE in the environment, drinking water, or human bodies [14,15,63,90,173].

Once in the environment, REE can be partitioned among solid phases [63], bioaccumulated in plants, and cause adverse effects in living biological systems [85,109]. According to González et al. [56], toxicity increases for heavier lanthanides. Hanana et al. [66] demonstrate in their study that La can bioaccumulate in muscles and affects the excretory and nervous systems of freshwater invertebrates. In the study of MacMillan et al. [108], the authors found the highest concentrations of REE in low trophic levels. They also concluded that, for some species, the uptake of REE is faster than the elimination, and the bioaccumulation patterns of REE appear to be tissue- and species-specific.

Regarding the human health effects, a few studies investigate the toxic effects of REE; others focus primarily on monitoring exposure to REE of residents close to mining activities. A discussion of human exposure and health effects linked to FL has not yet gained attention. It is known that REE can enter the bloodstream and cells, affecting cell function and protein production, interfering with the metabolism of sugar, fat, drugs, and proteins, and influencing the respiratory tract and liver function [14,25]. In addition, excessive exposure can harm the reproductive system in humans [25]. Wei et al. [173] evidenced higher concentrations of REE in human hair of residents near a mining area in China. They also observed that the accumulation of light REE increased with age, suggesting a low excretion rate and potential for bioaccumulation. Considering these studies, we believe that occupational exposure could also affect workers from factories and recycling facilities of FL, as well as residents. Therefore, we feel that studies assessing human exposure to REE in such environments should be encouraged, especially if there is an intent of increasing recycling rates of FL and possible recovery of REE. Additionally, environmental studies linking REE and FL are also lacking. It is crucial to understand better the contamination risk of soils and waters with REE from discarded lamps, as well as their behavior and fate in the environment.

3.3. Other potentially toxic elements

Other elements that constitute FL can represent a potential environmental and human health risk [162]. Although usually neglected in studies about FL, potentially toxic elements were approached in the works of Dos Santos et al. [39], Taghipour et al. [155], Tunsu et al. [162], Pavón et al. [129], and De Farias et al. [31]. Dos Santos et al. [39] determined Pb by analyzing a slurry of glass and phosphor using ICP OES and found an average concentration of 0.19 ± 0.01 mg per lamp. Taghipour et al. [155] determined the Pb, Ni, As, and Cr content in 20 spent CFL (glass and phosphor) from Iran, finding mean concentrations of, respectively, 2.33, 0.064, 0.056, and 0.012 mg per lamp. Tunsu et al. [162] found concentrations of Cd, Pb, Al, Ba, and Mn in fluorescent lamp waste, finding concentrations of, respectively, 0.3 ± 0.02, 0.7 ± 0.04, 14.3 ± 0.7, 10.5 ± 0.4, and 3.1 ± 0.2 g kg^{-1} of waste after removal of large impurities. Pavón et al. [129] determined the concentration of several metals in fluorescent lamp waste, including Al, Ba, and Mn, with concentrations in g kg^{-1} of, respectively, 11.0 ± 0.1, 4.0 ± 0.2, and 2.0 ± 0.1. De Farias et al. [31] evaluated the mobility of Cd, Cu, Mn, Ni, Pb, and Zn after a sequential extraction method and their concentrations after total digestion using FAAS. In the exchangeable fraction of the SEM, which simulates a water-soluble scenario, Cd, Cu, and Mn became available, being Cd the most mobile metal, with approximately 5% being mobilized. This result is concerning due to the known toxicity of Cd, which causes cancer and targets cardiovascular, renal, gastrointestinal, neurological, reproductive, and respiratory systems [125]. An increase in acidity allows more metals to be extracted, with 30 % of Zn, 21 % of Ni, and 0.39 % of Pb also becoming available in the weak acid

fraction. Mn and Pb have elevated concentrations in the phosphor material; however, despite being mobilized in the exchangeable or weak acid fractions, they need more extreme conditions to be mobilized in higher quantities. Nevertheless, due to the high toxicity of lead and its capability of bioconcentrate, the possibility of natural leaching of Pb from FL into the environment is a reason for caution. For total digestion, concentrations ranged from 34.9 (Ni) to 3134 mg L^{-1} (Mn). The amount of Mn and Al found in FL is also concerning. Exposure to high levels of Mn is associated with Parkinson's and attention deficit hyperactivity disorders [169], and Al can result in the development of neurodegenerative and neurodevelopmental disorders [151]. Concentrations of the quantified elements in the revised works were usually low. Nevertheless, they can still result in impacts considering the amounts of fluorescent lamp waste generated annually and the toxicity potential of some of the discussed elements.

The studies regarding metal contamination that are presented in Section 3 demonstrate how the presence of toxic metals in FL and e-waste, in general, is a substantial risk for the environment and human health if the wastes are not adequately discarded and efficient safety measures are not in place for people dealing with transport, disposal, and recycling. E-waste disposal is a public health challenge. Many developing and undeveloped countries still have inappropriate forms of disposal of FL, and it is estimated that FL will still be available for recycling for the next 20–30 years [22,59,98]. In 2019, around 53.6 million metric tons (Mt) of e-waste were generated worldwide, with only 17.4 % being correctly collected and recycled [52]. The high costs for e-waste recycling and disposal have led to the exportation of e-waste from developed countries (United States, Europe, Australia, Japan, and South Korea) to low and medium-income countries, such as China, Nigeria, and Ghana, where the labor costs are lower, and regulations and resources for recycling and safely disposing of the waste are ineffective or non-existent [23,74,124]. The informal recycling scheme that comes from the export of e-waste by developed countries puts the population and the environment of more vulnerable countries at imminent danger. Non-formal workers are susceptible to injuries, exposure to noise, and toxic chemicals that can cause numerous health effects (even long-term) [23,50,77,122,153,154]. E-waste sites can affect even children who live in surrounding areas [105,152]. Additionally, the potential for environmental contamination is increased in areas near e-waste centers because part of the waste is periodically burned to reduce volume, allowing toxic substances to be released into the environment. Contaminants leached by rains and floods can also be a path of environmental contamination of soils, water bodies, and organisms, as observed in Accra, Ghana [74]. This current global issue regarding e-waste raises awareness of the dangers the population of least developed countries still faces concerning hazardous wastes (including FL).

4. Life cycle assessment

Worldwide, there has been growing attention and interest in the LCA of FL, especially regarding the environmental impacts of their disposal. Although FL have been used for many years, there are few studies in the literature about their LCA. For the time range selected, only 15 LCA studies having FL as the primary or one of the primary materials were found. Most of them analyzed only FL [9,26,79,141,156,160,180,184], others compared FL, IL, and LED [5,19,46,101,104,133,143], or other types of lamps, such as tungsten and halogen lamps [174]. The studies were also reviewed in terms of boundary systems, functional units, software, impact assessment methodology, and end-of-life scenarios. Table S2 in the supplementary material summarizes the methods used. Most authors considered "cradle-to-grave" as system boundaries, which include all life cycle stages from the acquisition of raw materials and manufacture, to use and end-of-life. Particular interest was given to LCA studies that considered "gate-to-grave", which is only the final disposal (end-of-life) of the lamp, focusing on the recycling and waste treatment of the phosphor material. A discussion on functional units, software,

impact assessment methodology, and analyzed impact categories is provided in Topic S3 in the supplementary material.

Regarding the end-of-life of the lamps, the studies of Amato et al. [9], Yang et al. [180], Liu and Keoleian [104], and Ippolito et al. [79] focus on the metal and recovery of REE from waste phosphor material. Amato et al. [9] evaluated waste treatment for REE and metal recovery. They concluded that the processes involving the phosphor material cause the most significant impact in the categories of climate change, ionizing radiation, and resources. Yang et al. [180] compared three scenarios: Recycling with or without final disposal and landfill. The authors concluded that recycling REE without final disposal was the highest contributor to the environmental impact in the categories of non-carcinogens, freshwater ecotoxicity, and respiratory inorganics. Chemicals and energy production were the main contributors to the overall environmental impact of the recycling scenario with final disposal due to the high emissions of particulate matter, sulfur dioxide, heavy metals, carbon dioxide, and fossil fuel use. In comparison, the landfill scenario contributed less to the overall environmental impact (considering REE), with energy savings and low direct emissions to the environment.

Ippolito et al. [79] compared the sustainability of two recycling processes for REE recovery from the phosphor material. The processes differ only in the pretreatment step, with one using heat treatment, and the other using mechanical activation. The authors concluded that, although the thermal pretreatment requires greater energy demand than mechanical activation, the higher rare earth recovery yields give the thermal option greater environmental credit, showing an advantage of around 20 %. Furthermore, the authors noticed that the potential benefit of rare earth recovery from secondary sources compared to the primary production was mainly in the categories of climate change, and resource use and energy carriers. They also observed that the leaching phase was the main environmental burden in most of the environmental categories analyzed. The set of analyzes employed by the authors allowed to conclude that the recovery of rare earth oxides from FL is advantageous both environmentally and economically, with profitability being affected by market price fluctuations, which is a very important result for the area.

Liu and Keoleian [104] examined the environmental impacts of recovering REE from FL. They concluded that it resulted in more environmental impacts than landfill disposal across all impact indicators, except fresh water and marine ecotoxicity, due to the consumption of large quantities of chemicals, which contributed to 71–100 % of impacts. This indicates that improvements in recovery procedures for REE should be encouraged. Nevertheless, the acquisition of REE from a secondary source, such as lamps, implicates diminishing mining of REE, which is also responsible for considerable environmental impacts [79, 107]. Therefore, studies comparing the mining and recovery of REE from e-wastes, such as the work by Ippolito et al. [79], are interesting for economic and environmental evaluations. The assessment of the environmental impact of the processes used for the extraction and recovery of metals from the phosphor material and the associated wastewater treatment are also important, as presented in a study by Rocchetti et al. [141]. Results showed that the metal extraction from e-wastes, such as the residue from physical processes for recycling fluorescent lamps, represented the highest load on the environment; however, treating phosphor material appears beneficial to the environment regarding CO_2 emissions.

Authors that focused only on FL also compared types, such as linear and compact lamps. Tan et al. [160] concluded that linear fluorescent lamps have less environmental impact than compact ones. The use stage contributed to more than 94 % of total impacts due to electricity consumption, which could be reduced using a different electric source. In the manufacturing stage, the ballast component contributed the most to the environmental impact of the CFL. In the end-of-life stage, the lamps were sent to licensed waste treatment companies to treat mercury, which provided a net environmental benefit. Chen et al. [26] compared

greenhouse gas (GHG) emissions of CFL and LFL from the manufacture to the final disposal phase. The results also indicated that the greatest negative impact of both kinds of lamps is in the use phase due to electricity consumption. In general, the conclusion is that LFL are a better source of light than CFL concerning GHG emissions.

The same conclusion about the use stage was obtained in the study by Tahkamo et al. [156], in which the energy consumption in the use phase of FL had the most significant global environmental impacts (93 %), followed by manufacturing, with 7 % and end-of-life, with less than 1 %. The authors suggested that changing the energy source to a less polluting type, such as hydro power, could reduce the environmental impacts of the use stage by approximately 90 %. Recycling and disposal of lamps are not responsible for the most significant environmental impact; however, it is part of the lamp cycle that still has room for substantial improvements to potentially reduce the impacts, which are not so easy for other segments.

On the other hand, the use phase had less impact on the entire lamp life cycle in the study by Bertin et al. [19], who compared the potential environmental impacts of CFL, LED, and LFL using an electricity mix for indoor workplace lighting. The lower impact of the use phase can be explained by the electricity used, which is generated mainly from nuclear plants.

Principi and Fioretti [133] conducted a comparative LCA study between CFL and LED luminaires at all life cycle stages, showing that the use stage is responsible for 82–99 % of the environmental impact, mainly due to the consumption of electricity. The use of LED allows a 41–50 % reduction in the emission of greenhouse gases and the accumulated demand for energy. However, considering the production phase, LED have a higher environmental impact than CFL due to the greater weight of the luminaire and its electrical components. Also, the use of LED reduces mercury emissions by 41–50%. Recycling was the disposal scenario that had the lowest environmental impact.

Many authors have also made comparisons involving IL and CFL to highlight the advantages and disadvantages caused by replacing the former with the latter. Incandescent lighting is much less efficient because most of the consumed energy becomes heat. However, based on the studies reviewed, despite CFL being harmful to the environment due to their mercury content, the impact of other lamps could be worse depending on the energy source used, as explained by Allione et al. [5]. The authors concluded that the main environmental gains of a CFL come from its environmental-friendly behavior during the use phase, with lower CO_2 emissions. However, CFL's embodied energy and global warming potential (GWP) are higher than IL in the production and end-of-life phases. In general, the low environmental loads of the lifetime of CFL compensate for the worst performances of CFL that occur in other stages of the life cycle. Corroborating the previous studies, Elijošiutė et al. [46] compared CFL and IL. They concluded that the negative environmental impact was greater in the use phase and that CFL are more appropriate than IL, mainly for the more efficient use of electricity and the longer exploration time.

Sangwan et al. [143] compared the environmental impacts of IL, LFL, CFL, and LED and found that LED consume more resources and have greater effects on human health than CFL and LFL. The CFL presented more significant eutrophication potential and ionizing radiation categories in the end-of-life phase. Energy consumption of IL was high, as well as contributing to human health and resource consumption. In the study by Lim et al. [101], resource depletion and toxicity potentials were analyzed in IL, CFL, and LED. CFL and LED had higher resource depletion and toxicity potentials than the IL due mainly to the high content of aluminum, copper, gold, lead, silver, and zinc, also causing more impact on human health and ecosystems in the disposal stage.

Although the types of lamps compared to FL were primarily LED or incandescent, different lighting technologies were observed in the study by Welz et al. [174], who presented a LCA comparison of the IL, the halogen lamp, the LFL, and the CFL. The results showed that FL had less environmental impact than tungsten and halogen lamps, and the use

phase was the main contributor to the impact due to the electricity mix used, independent of the actual lamp type examined. An appropriate disposal process, such as recycling and incineration, is important, but the differences between the two remain small compared to the impacts associated with the use.

In general, the literature review demonstrated the researchers' preference for LCA studies that evaluated only FL, with established system boundary from cradle-to-gate, assessed by SimaPro software using the CML and Eco Indicator 99 impact assessment methodologies.

Although not common among the studies reviewed, the analysis of reverse logistics can be determined by LCA, as in the study of Zappe et al. [184]. The authors evaluated the reverse logistics of FL, considering the environmental impact of landfill and recycling, and concluded that recycling the bulbs can bring environmental benefits when compared to disposal of lamps without any kind of treatment since it prevents the release of mercury into the environment. However, the burning of fuels in the processes was the main source of atmospheric depreciation and generation of pollutants, such as CO_2, CO, NOx, SO_2, volatile compounds, and particulates, among others.

Then, the intention of this work by reviewing LCA studies about fluorescent lamps is to help identify the influence of different factors on the environmental burdens of each life cycle activity, considering materials, energy, and pollutants or waste released. The studies reviewed in this section showed that the use phase of the lamps had the highest global environmental impacts due to the energy source used, which can be reduced by changing it to a cleaner energy source, like hydropower. Regarding end-of-life, some authors concluded that the disposal in landfills could have a lower environmental impact than recycling [104, 180]. Nevertheless, most results indicate that recycling still is the best alternative to treat spent FL [19,26,101,133,160,174,184], especially if applied on a large scale, resulting in less environmental impact, and allowing the recovery, treatment, and reuse of metals and other components, thus, providing the circular economy.

Evaluating all life cycle stages is important because sometimes a process or product is considered ecologically correct but has a high environmental impact at some stage. For example, it is known that replacing FL with LED lamps has many advantages, as the use of LED is beneficial in almost all stages. However, studies reviewed in this section found that LED consume more resources due to the greater weight of the luminaire and its electrical components and have higher effects on human health than FL in the manufacturing stage.

5. Recycling and metal recovery from fluorescent lamps

5.1. Recycling of fluorescent lamps

The presence of Hg and phosphor is the main environmental concern regarding FL, which is why they are classified as hazardous waste [81, 137]. US EPA recommends segregating FL from general waste for recycling or safe disposal. Laws in several countries control their collection for the treatment and removal of mercury.

Currently, different technologies are used for recycling FL, as described in Topic S4 and summarized in Fig. S1, both in the supplementary material. The dismantled components of FL obtained in the recycling process are contaminated with mercury, needing thermal or chemical treatments for its decontamination to safe concentrations, being then distilled or stabilized, which will be discussed in Section 5.2.2 [93]. After the dismantling and decontamination, the lamp components can be further recycled. The glass is mainly employed in glass and glass wool factories and the metallic caps in foundry companies, while the phosphor can be treated chemically for recovery of REE, as reviewed in the next section, or employed in the ceramic, paint, and pigments industries [13]. Even though it is possible to reuse the phosphor material directly for manufacturing new lamps, this is not a recommended approach since the quality of the phosphor deteriorates over its lifetime, and the illumination intensity can be impaired. In addition,

this practice would only be possible if the reuse was made by the same producer of the end-of-life lamp where the phosphor was withdrawn since different manufacturers use different phosphor combinations [20, 107]. The removed Hg can be reused by industries of FL to be employed in chemical industries, such as in chloro-alkali production. Although forbidden in most countries, Hg is still used in gold mining [13,93]. A few authors have also approached the use of fluorescent lamp waste in the production of construction materials, such as Red-Clay Bricks [12], roof tiles [38], and recycled glass powder as cement replacement [119, 130,132,157]. Gedik, Selcuk, and Lav [54] investigated using fluorescent lamp waste as mineral filler in asphalt pavements finding promising results, while Ling and Poon [102] studied the use of glass from spent FL as a substitute for fine aggregate in cement mortar.

5.2. Recovery of metals from fluorescent lamps

5.2.1. Rare earth elements

Obtaining REE from secondary sources, such as FL, has gained much attention in recent years, motivated mainly by geopolitical tensions related to China's monopoly on production and supply over REE and the increase in their industrial demand in the last 20 years, which grew from 75,500 t in 2000–123,100 t in 2016 [57]. In the last ten years, a decrease in the price of REE was observed, making the recovery of these metals economically unfeasible for large-scale applications [134]. Nevertheless, considering the market volatility, it is important to develop and improve methods for recovering metals from e-waste so that countries, where REE are considered critical, can have more autonomy over metal supply, mainly for political and economic reasons.

Many authors have developed methods for recovery of REE into rare earth element compounds from the phosphor of FL. The first step is phosphor dissolution, which can be achieved through acid leaching [4, 10,116,129,144,162,179], alkaline fusion [80,100,185], or with the use of ionic liquids [43,146]. However, other authors used organic acid-producing microbes, like tea fungus Kombucha, for the leaching of the phosphor, being able to solubilize the phosphor $Y_2O_3:Eu^{2+}$ [72]. Fig. 1 summarizes some of the main paths for the full recovery of REE from FL, with examples of employed reagents and the resulting product, in each approach used.

5.2.1.1. Acid leaching.
Several mineral acids can be used to dissolve the phosphor material, such as H_2SO_4, HNO_3, and HCl. The different phosphors that compose the phosphor material have different chemical resistances. Mishra, Devi, and Sarangi [116] used HCl 2 mol L^{-1} to leach the phosphor, which resulted in good recoveries for Y and Eu but not for Tb and La (Table S3 in the supplementary material). Leaching tests performed by Anand et al. [10] also showed that ~2 mol L^{-1} acid solutions (H_2SO_4 1.62 mol L^{-1}) provided good recoveries for Y and Eu but not for Tb and La, which needed a second acid leaching step with HCl (54 % v/v) to be efficiently dissolved (Table S3 in the supplementary material).

Tunsu et al. [162] studied the leaching of the phosphor of FL with different solutions. Pure water and 1 mol L^{-1} NH_4Cl could not leach the REE; however, different percentages of REE were leached with other acid solutions (Table S3 in the supplementary material). Even using strong acid solutions (4 mol L^{-1} HCl or HNO_3), Tb, Ce, and La were poorly extracted, showing the great chemical resistance of the structurally complex compounds in which these elements are present. Regarding the leaching kinetics, faster dissolution was observed for Y and Eu with HNO_3 2 mol L^{-1}, while Ce and Tb leaching took place slowly, not reaching the equilibrium even after 96 h (50 %). La leaching reached a maximum of approximately 20 % extraction after 24 h, showing no further dissolution.

Yang et al. [179] also showed the difference in leaching behavior of the different REE. Fig. S2 in the supplementary material presents the leaching and extraction process applied (the extraction will be discussed
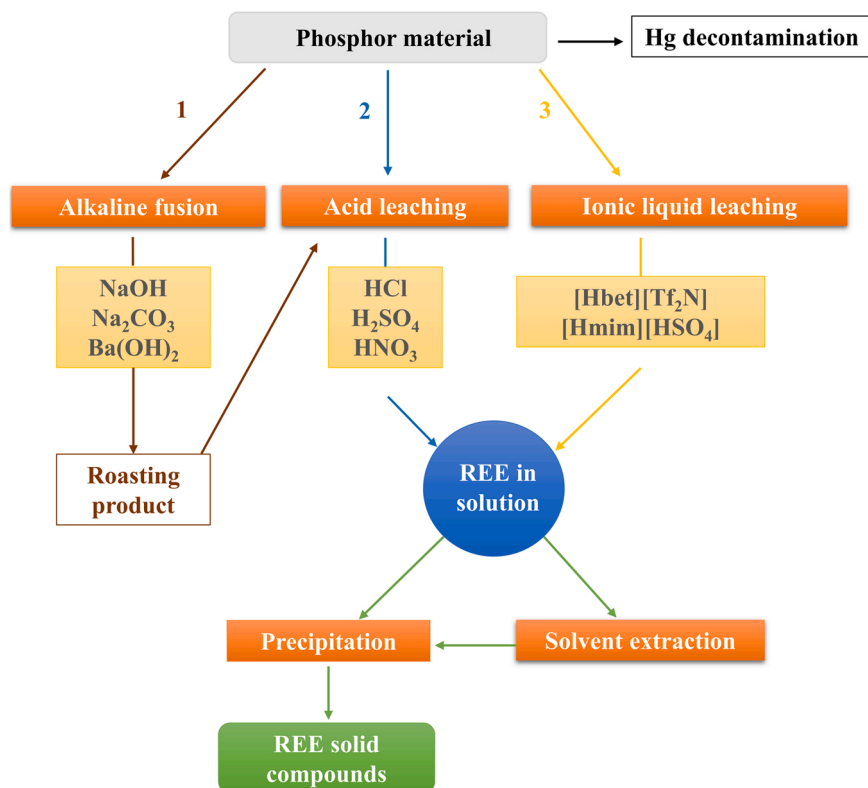
Fig. 1. Flowsheet of the main possible ways to recover REE from the phosphor material.

later on). The authors used HCl, HNO$_3$, and H$_2$SO$_4$ 5 mol L^{-1} to leach the phosphor, being able to dissolve Y and Eu with efficiency higher and close to 90 %, respectively, with all three acids. However, a second leaching step was needed with HNO$_3$ 5 mol L^{-1} to leach Tb (98 %), Ce (85.9 %), and La (91.1 %) from the green phosphor.

So far, it has been noticed that mineral acids, such as HCl, H$_2$SO$_4$, and HNO$_3$, present similar leaching capacities for each phosphor that compose the phosphor material of FL, which has different leaching resistance, as presented in Table S4 in the supplementary material.

The red phosphor is the component with the highest intrinsic value on the waste phosphor since it is the phosphor that contains REE present in greater quantity, is easily solubilized by diluted acids, and contains 87 % in weight of Y and Eu, which are in the list of critical materials for both the United States [53] and Europe [48]. Therefore, its extraction and recovery from FL are attractive and desired, having great potential since, according to European Commission [48], respectively, 31 % and 38 % of the overall demand of Y and Eu in the EU can be satisfied through secondary raw materials.

The low solubility of the green and blue phosphors (La, Ce, and Tb) can be explained by the fact that, while Y and Eu exist as simple oxides, La, Ce, and Tb exist as complex crystalline structures, which are hard to dissolve. Besides, the complex structures of the blue and green phosphors consume a higher quantity of protons [162], meaning that the protons left after the dissolution of halophosphates and red phosphors [164] cannot be enough to dissolve them completely, making necessary higher acid concentrations and heating, and limiting the solid/liquid (S/L) ratio (% w/v) of the acid leaching to 20 %.

Since Tb is the most valuable rare earth element among those in the phosphor [166], finding sustainable ways to recover it is a significant challenge. Tan et al. [161] employed mechanical activation of the phosphor by destroying the crystalline structure of the green and blue phosphor in a ball mill, making acid leaching easier. Increasing the rotational speed of the mill and the activation time had a positive impact on the leaching rates of the REE. The leaching obtained with HCl

6 mol L^{-1}, S/L ratio of 0.0167 g mL^{-1}, and for 15 min in magnetic stirring were 89.4 % (Tb), 93.7 % (Ce), and 91.4 % (La) from the activated phosphor, against less than 1 % of each one from the inactivated phosphor. In order to investigate the activation mechanism, the activated phosphor was analyzed by X-ray diffraction (XRD), which showed the destruction of the crystalline structure of the green phosphor and the formation of rare earth element oxides, such as La$_2$O$_3$ and Tb$_2$O$_3$. A decrease in the bond strength between La, Tb, Ce, and PO$_4^{3-}$ was also observed.

5.2.1.2. Alkaline fusion. Alkaline fusion is an alternative to help dissolve the phosphors that are more resistant to acid leaching. Ippolito et al. [80] proposed alkaline fusion of the phosphor with Ba(OH)$_2$, which transforms the phosphates of REE into their respective oxides that are more easily dissolved. Zhang et al. [185] proposed applying two acid leaching stages, interspersed with alkaline fusion, for the recovery of REE present in the phosphors of FL. In the first HCl leaching stage, the red phosphor (Y and Eu) was mainly dissolved (more than 95 %), the residue was submitted to alkaline fusion with NaOH, and then the most resistant REE, Ce, and Tb were about 90 % leached on the second HCl leaching stage.

According to Liang et al. [100], the trivalent form that Ce and Tb are present in the phosphor can be oxidized to form tetravalent oxides during the alkaline fusion, which are more difficult to decompose in comparison with trivalent oxides. Therefore, they developed a process by mixing the phosphor material with a reductive ferrous powder for the alkaline fusion process. This reducing agent would keep the REE in the trivalent state during the roasting. The subsequent leaching with HCl leads to the dissolution of more than 98 % of the REE.

As seen in the revised works, alkaline fusion can be done before any acid leaching, i.e., in the phosphor mixture containing all REE or in the solid residue resulting from the acid leaching stage (after the dissolution of the red phosphor), to transform the undissolved REE present in the structurally complex oxides of the green and blue phosphors into their

respective simple oxides that are more easily dissolved by a second acid leaching stage. As shown in Table S3 in the supplementary material, both procedures can provide good recoveries of REE from the phosphor (>80 %). The latter strategy enables the formation of more purified leachates, which can facilitate the recovery process of REE from it; however, at least two acid steps are needed to dissolve the different phosphors. Even though the alkaline fusion enables a better leaching efficiency of Tb, La, and Ce, high temperature and energy-consuming conditions are necessary to melt the phosphor.

5.2.1.3. Ionic liquid. The REE present in the phosphor mixture of FL can also be dissolved using a treatment with ionic liquids (ILiq) called ionometallurgy. This is considered an environmentally friendly option compared to acid leaching and alkaline fusion since it uses nonaggressive conditions and solvents with low vapor pressure, flammability, and toxicity [43], which can be recovered and reused by the end of the process. Also, depending on the ILiq used, it is possible to extract certain REE from the phosphor selectively.

Dupont and Binnemans [43] and Schaeffer et al. [146] studied the possibility of using the ILiq bis(trifluoromethylsulfonyl)imide ([Hbet] [Tf$_2$N]) and 1-methylimidazolium hydrogen sulfate ([Hmim][HSO$_4$]), respectively, for the selective dissolution of Y and Eu from the phosphor material of FL. The first step was to stir and heat the phosphor mixture with the ILiq and pure water in a container. After separating the undissolved phosphor, Y and Eu were recovered from the ILiq solution by precipitation with oxalic acid. The Y and Eu oxalate obtained was filtrated and calcined, regenerating the red phosphor. After the calcination, Dupont and Binnemans [43] obtained yttrium oxide doped with Eu^{3+}, just like the original red phosphor (Y$_2$O$_3$:Eu^{3+}), with 99 % purity, while Schaeffer et al. [146] obtained Y and Eu mixed oxides (Y$_{0.95}$Eu$_{0.05}$)$_2$O$_3$ with 86 % purity, being Ca the main impurity in the product. The leaching efficiency of both ILiq used in these works is presented in Table S3 in the supplementary material. In both cases, the luminescence spectrum of the final product was similar to that of the pure red phosphor, having the same characteristic emission peaks and showing that the recovered phosphor could be reused.

As observed in the results summarized in Table S3 in the supplementary material, any phosphor dissolution method described in the last sections can provide good leaching efficiencies for the REE when the experimental conditions are optimized. The acid leaching of the phosphor is a simple process, and any acid concentration easily dissolves Y and Eu from the red phosphor. Still, aggressive conditions such as strong acidic solutions and high temperatures are necessary if Tb, Ce, and La recovery are desired. Strategies such as the mechanical activation or alkaline fusion of the phosphor can be applied to increase the dissolution of the green and blue phosphor with more moderate acidic conditions by a second leaching step. However, these processes come with energy-consuming aspects like temperatures such as 800 °C for the fusion and electricity for the rotating mill in the mechanical activation. The use of ILiq is a novel and environmentally friendly option for phosphor dissolution. It does not need heating and enables an efficient selective dissolution of the red phosphor, keeping the other phosphors in the solid phase, which facilitates the recovery of the REE from the leachate. However, the high cost of some ILiq is a drawback, and it was not possible to find works in the literature concerning the dissolution of other REE.

Regarding recovery and reuse of REE, which is an important matter but a procedure still not adopted in every county, it is important to be done by sustainable processes to be viable on an industrial scale. Since the dissolution characteristics of each phosphor present in FL are already known, we recommend that studies concerning the dissolution of the phosphor should focus on developing sustainable methods for the recovery of REE from phosphor material of FL, with consumption of reagents and energy as low as possible and with low cost. Studies about scaling up the developed methods with the assessment of price and quantity of reagents and energy necessary should also be done, as well as the assessment of the environmental impacts of the recovery methods.

5.2.1.4. Metal recovery from leachates. After solubilizing the REE present in the phosphor, it is necessary to recover them from the leachate, which is usually performed by precipitation with oxalic acid or liquid/liquid extraction (solvent extraction). The precipitation with oxalic acid and subsequent calcination of the rare earth element oxalates generates rare earth element oxides [164]. However, the chemical complexity of the phosphor makes the final acid leachate contaminated with varying amounts of undesirable ions, especially those related to the halophosphate (Sr, Ca, F, and Mn) that also precipitate as oxalates and form the respective oxides by the calcination. Furthermore, in solution, fluoride from halophosphates can complex with REE to form REEF$_3$, as Han and Han [65] stated.

To improve the purity of the rare earth element oxides, De Michelis et al. [32] chose H$_2$SO$_4$ to leach the phosphor material since it forms insoluble sulfates of Ca, Pb, and Ba, which can be filtered before the addition of the oxalic acid, serving as a purification of the leachate. The solid characterization by XRD showed a 90–95 % purity of the resulting oxalate, with low contamination of other elements, such as Fe and Ca (less than 1.5 % w/w). However, the H$_2$SO$_4$ can also form rare earth element sulfates, which can precipitate on the surface of the phosphors and inhibit further leaching, as stated by Zhang et al. [185].

An alternative is to use solvent extraction for the separation of the REE. This procedure is divided into extraction and stripping steps. In the first step, the REE are selectively captured from the phosphor leachate by an extractant dissolved in an organic liquid, keeping the undesirable ions in the aqueous phase. Then, in the stripping step, the REE are stripped from the organic solution by mixing it with an acidic solution, regenerating the extractant in the organic solution. The acidic aqueous stripping solution has only rare earth ions, which can be precipitated with oxalic acid without major problems.

Besides the type of extractant to be employed, whose efficiency and selectivity for separating REE from the leachate varies, it is also important to optimize the kind of acid used in leaching and stripping, its concentration, the ratio between the volume of aqueous and organic phases, and the number of steps of extraction and stripping.

Tunsu et al. [164] developed a route to recover Y and Eu from fluorescent lamp waste using a three-step leaching process, followed by solvent extraction with Cyanex 923, a commercial mix of trialkylphosphine oxides. A flowsheet of the extraction process (adapted from Tunsu et al. [164]) and the efficiencies obtained in each step are present in Fig. S3 in the supplementary material. The authors also proposed the decontamination of Hg from the solution obtained in the first leaching step by ion exchange, reduction, or solvent extraction, as will be discussed later. In Pavón et al. [129], Cyanex 923 was also effective when used as a carrier in a supported liquid membrane to recover REE leached from phosphor material. Batchu et al. [17] showed that the Cyanex 923 extractant could also separate Y and Eu present in non-aqueous media such as ethylene glycol.

ILiq can also be used as a solvent for the extractant in the recovery process of REE by solvent extraction. The advantages of ILiq to conventional organic solvents are less toxicity and volatility, being favorable for use in processes undergoing high temperature, as the case in phosphor leachate. Yang et al. [179] used the extractant N,N-dioctyldiglycol amic acid (DODGAA) diluted in the ILiq (1-butyl-3--methylimidazolium bis(trifluoromethylsulfonyl)imide) ([C$_4$min] [Tf$_2$N]) for the extraction step (Fig. S2 in the supplementary material). The extraction system exhibited high affinity and selectivity to rare earth ions, keeping metallic impurities in the solution. Furthermore, it was observed that the extraction capacity of DODGAA dissolved in the ionic liquid was greater than dissolved with n-dodecane. Quantitative stripping was also noticed with the above conditions; however, a loss of 1.4 % of the ILiq in the stripping product was observed. The ability to

reuse the ILiq as the solvent for the extractant after stripping the REE was evaluated, with the same process repeated five times. The extraction ability decreased slightly, but the authors considered recycling the extracting phase possible.

5.2.2. Mercury

The Minamata Convention stipulated that the manufacture, import, and export of mercury-added products such as pesticides, biocides, and non-electronic measuring devices such as barometers and thermometers should not be allowed after 2020 [94]. Regarding FL, the Convention states that CFL of < 30 W and LFL containing phosphor of REE of < 60 W must have less than 5 mg of mercury by the same date. Despite the tendency to ban mercury in several products, the metal is still allowed in FL since it is essential for the lamp's functioning [135]. However, the Hg present in FL can be recovered and reused by the industry after thermal or chemical treatments [175].

5.2.2.1. Thermal treatments. The most common way used by lamp recyclers to decontaminate FL is through thermal desorption processes [28]. After being broken, the lamps are submitted to high temperatures, and the mercury is vaporized and distilled. Although elementary Hg is volatile and has a boiling point relatively low at 357 °C [175], during the use of the lamps, the elemental mercury vapor can react and deposit on the phosphor, glass matrix, and end caps in oxidized forms [135], requiring temperatures up to 800 °C to ensure its total removal from the lamps, and allow safe disposal or recycling. This makes Hg thermal desorption a very energy-consuming process [163].

Pyrolysis at anaerobic conditions can decrease the risk of oxidation of toxic metals in the material, being effective for the thermal treatment of several wastes. Wijesekara et al. [175] applied a laboratory-scale pyrolyzer to remove mercury from the glass of used FL. Three traps were used sequentially to collect the vaporized Hg: A cold trap to condense the purged Hg vapor, a particular type of coal with additives to adsorb the metal, and a 2.5 % HNO_3 solution, capable of oxidizing and trapping the Hg to soluble salts. About 89.7 % of the total Hg was removed at 400 °C, and 600 °C was necessary for complete decontamination (99.8 %). Also, it was seen that the cold trap, additive coal, and acid trap collected 19.04 %, 77.62 %, and 3.3 % of vaporized mercury, respectively, showing the good capacity of additive coal to capture mercury.

Gupta et al. [62] synthesized and tested three types of functionalized multi-wallet carbon nanotubes (CNT) for the capture of mercury from FL: Oxidized CNT (CNT-OX), iodine incorporated CNT (CNT-I), and sulfur incorporated CNT (CNT-S). CNT are characterized by their nanostructure and high surface area, having a great capacity to adsorb metallic pollutants such as Pb, Cd, and Hg. The idea of functionalizing the nanotube with iodine and sulfur has the objective of improving the selectivity and adsorption capacity of the sorbent by the high stability of S-Hg and I-Hg complexes formed on its surface. Nitrogen adsorption isotherms showed that the maximum adsorption capacity of the synthesized CNT was in the order of CNT-S> CNT-I> CNT-OX> CNT, with CNT-S being 27.5 times more efficient than the virgin CNT. The authors evaluated the capacity of the synthesized CNT to adsorb Hg^0 present in FL. They placed 40 g of broken CFL in a U-tube, where nitrogen gas was purged for five hours, and the steam generated bubbled into a 4 % solution of $KMnO_4$ to oxidize and capture the gaseous mercury. After, the same procedure was performed with the broken lamps placed next to 1 g of each synthesized adsorbent. The oxidizing solution was analyzed by cold vapor atomic absorption spectrometry (CV AAS). It showed that, without the presence of the synthesized CNT, 2.5 mg L^{-1} Hg was determined in the solution, while in the presence of 1 g of CNT, CNT-OX, CNT-I, or CNT-S, only 0.170, 0.062, 0.005 and 0.002 mg L^{-1} of Hg were determined in the trap, respectively, corroborating the preferential Hg absorption by the CNT.

5.2.2.2. Chemical treatments. The high energy consumption required in thermal treatments to make the complete desorption of mercury makes a wet decontamination process (chemical treatment) sounds more viable for removing the toxic metal from the waste. Similarly to the REE, the mercury in the phosphor of FL can also be leached with acidic solutions. *Aqua regia* is usually employed by various authors to determine the total amount of mercury in the wastes since the strongly acidic and oxidizing characteristic of this mixture can oxidize and capture the mercury in solution in the Hg^{2+} divalent form [137]. However, besides the environmental impact, the use of strong acids to leach Hg can also leach the phosphor mixture. Therefore, if the recovery of REE were intended, the toxic metal would exist as an impurity in the leachate and vice versa. This would hinder the selective recovery process of REE, in addition to contaminating the aqueous waste streams produced. Thus, some studies focused on developing forms of wet decontamination of fluorescent lamp waste using oxidizing solutions that selectively leach Hg and not the phosphor mixture. Tunsu et al. [163] studied molecular iodine and potassium iodide (I_2/KI) solutions for the Hg decontamination from fluorescent lamp waste and its recovery from the resulting leachate by the three methods. According to the authors, this type of oxidizing solution has already been used successfully to remove Hg from soils and other contaminated materials. The mercury leaching mechanism occurs as follows:

(i) Oxidation of mercury and its compounds to Hg^{2+}.
(ii) Reaction with iodide ions to form the highly water-soluble HgI_4^{2-} complex.

The influence of some experimental parameters on mercury dissolution was investigated. Increasing the concentration of the leachate solution generally increased the Hg dissolution, which presented no further leaching after two hours of process, while increasing the S/L ratio to more than 10 % (v/v) decreased the extraction capacity. During the tests, the leaching of REE and metals from the halophosphate phosphor was low (<2 %), showing the selectiveness of the leachate to dissolve mercury. Fig. S4 in the supplementary material shows the whole process and the percentage of mercury recovered by each method. The optimal leaching conditions removed 97.3 % of the Hg present in the material.

Coskun and Civelekoglu [28] also optimized the use of an oxidative leaching process with a NaOCl/NaCl mixture for the selective dissolution of Hg from FL. The leaching mechanism is similar to that of the I_2/KI solution, causing the formation of a high soluble tetrahalogenated complex ($HgCl_4^{2-}$) (Eq. 1):

$$Hg_{(s)} + HOCl + 3Cl^- \rightarrow HgCl_4^{2-} + OH^- \qquad (1)$$

The authors used cementation to recover mercury from the leachate. In this technique, Hg soluble species are selectively reduced and precipitated into a solid phase using a reducing agent. Neutral powdered metals, such as Zn, Cu, and Fe, were tested as the reducing agents and mixed with the leachate obtained in the leaching step. The optimal leaching conditions (0.5/0.2 mol L^{-1} NaOCl/NaCl, pH 7.5, 1/2 S/L w/v, and two hours) enable a 96 % decontamination of Hg, while the optimal cementation conditions (5 g/L of Zn in the leachate, pH 1, and 5 min) cemented 99 % of the dissolved Hg. The solid formed by cementation was analyzed by XRD, showing that the precipitated mercury was presented as kleinite ($Hg_6Cl_3N_3H_2O$).

In order to recover the mercury extracted into the leachate, it is necessary to carry out its reduction, mainly of the Hg^{2+} ions, to the elemental state, which precipitates and can be separated. Also, other strategies can be applied to recover Hg from the leachate of FL.

Ozgur et al. [126] used electro-winning to recover the Hg leached from fluorescent lamp waste with the NaOCl/NaCl solution. In this process, an electrolytic cell is immersed in a solution, where electrons are passed from the metallic impurity present as the anode (IrO_2) to the

cathode (graphite), oxidizing and diluting the first and reducing the Hg in the solution, which deposits on the graphite cathode. Mercury reduction can be facilitated by adding citric acid to the solution before the electro-winning process. It forms a complex with Hg^{2+} ions, which reacts with hydroxyl radicals (Eq. 2), helping to reduce the metal, in addition to reducing the formation of oxygen gas (Eq. 3). In the optimization tests, it was shown that the increase in current and concentration of citric acid in the leachate corresponds to an increase in the rate of Hg reduction.

$$[Hg^{2+}nCit^{3n-}] + (3n-2)H^+ + (18n-2)OH^- \rightarrow 6nCO_2 + (13n-2)H_2O + Hg^0 \quad (2)$$

$$2\ OH^- \rightarrow H_2O + 1/2\ O_2 \quad (3)$$

The optimal leaching conditions recovered 95 % of the Hg from the waste. In contrast, in the optimal electro-winning conditions, 81 % of the Hg in the solution was reduced and recovered in the graphite cathode, whose analysis by scanning electron microscopy with energy dispersive X-ray (SEM/EDX) proved to be equivalent to 2.83 % of its weight.

The studies reviewed in this section show that the selective leaching of mercury with oxidative solutions is the best and easier strategy to decontaminate FL, considering that most toxic metals in the product end up on the phosphor material. By using these solutions, the REE are not leached and can be later recovered by the processes described in section 5.2.1 without contamination of the aqueous streams generated. Also, it was shown that the resulting Hg in the solution could be efficiently recovered by precipitation methods, allowing the leaching solution to be regenerated and reused. As stated, even though thermal decontamination processes are energy-consuming, they are still the most common way used by lamp recyclers to remove Hg from FL. Also, using carbonaceous materials (CNT, activated carbon) shows a great capacity to capture the vaporized mercury.

6. Current efforts and future perspectives

As demonstrated and discussed in previous sections, environmental and health impacts could arise from discarding spent FL and e-wastes due to toxic metals. Moreover, the impacts on the environment and human health related to REE are not yet fully understood, and the proportions that this can reach over the years, with the increase in their industrial applications, are unknown. The impacts related to FL depend on how the lamps are handled in the end-of-life, the policies in place to guarantee their safe disposal, and population commitment. Despite affecting countries unequally, this remains an ongoing issue of worldwide concern. As discussed in the United Nations Environment Programme - UNEP [168] regarding the sustainable development goals (SDG) on sustainable consumption and production, it is essential to promote the circular economy, meaning the reuse, recycling, remanufacture, and recovery of products in end-of-life, to reduce the use of virgin materials and minimize the generation of hazardous waste and greenhouse emissions.

Countries like The United States, Germany, Austria, Switzerland, and others have measures to appropriately handle spent FL (and other e-wastes) already in place, with growing recycling rates [93]. However, this does not represent a worldwide scenario. Most countries in Africa still have no legislation regarding the management of e-waste, and the recycling rate in the continent is 0.9 % [52]. In Brazil, initiatives for appropriately handling spent FL are expanding but were far below the expectations for 2020. National collection measures still depend heavily on population awareness and their willingness to go to collection points (which cover just a little more than 7 % of the country's municipalities), hindering the advancement of recycling in the country [136].

It is evident that more government resources and action must be directed towards a circular economy for hazardous materials, and regulatory measures for the disposal and recycling of hazardous waste (including FL), especially in developing countries. It is still necessary to make the population aware of the dangers of the metals in FL and of the

importance of disposing of spent FL in appropriate places to reduce the impacts of such materials. Special attention should be given to procedures regarding the recovery and reuse of metals present in the phosphor material in a sustainable way. Studies on applying fluorescent lamp recycled contents in other segments of the industry should be encouraged and put into practice when feasible. The replacement of FL with LED can positively contribute to minimizing environmental contamination by the energy sector, but the replacement rate is not equal in all countries, and it can take years for FL to be completely phased out. Furthermore, this replacement needs to be accompanied by effective recycling of the discarded FL (considering all potentially toxic metals present).

On a broader issue, the generation and consumption of energy are responsible for more environmental impacts than the end-of-life of products, as seen in LCA studies discussed in Section 4, and are the dominant contributor to the greenhouse effect [167]. Considering the SDG on affordable and clean energy, a way to mitigate environmental impacts and slow the depletion of many natural resources is to increase energy efficiency and the use of renewable energy (solar, wind, hydropower, biofuels, and geothermal) [167,168]. The goal by 2030 is to substantially increase the share of renewable energy in the global energy matrix and double the global rate of improvement in energy efficiency [167]. Considering the lighting scenario, replacing lamps such as incandescent, halogen, and fluorescent lamps with more energy-efficient ones like LED reduces energy consumption. And if robust growth is maintained, LED can cover up to 90 % of sales by 2030 [75].

7. Conclusion

Despite gradually being replaced by LED, FL are still used in commercial and domestic lighting in many countries and are responsible for generating significant amounts of hazardous wastes and environmental impacts. For this reason, it is still vital to give attention to the environmental risks arising from FL and the measures that can be taken to minimize them.

According to the reviewed LCA studies, the use stage of FL contributed to the greatest environmental impact due to the electricity consumption that, in most studies, did not come from clean energy. It highlights the importance of measures to encourage the use of renewable and clean sources for energy generation, as discussed in the United Nations' sustainable development goals. Nevertheless, the environmental impacts associated with the end-of-life of FL still need attention.

The toxicity of elements such as Hg, Pb, As, and Cd is well known and debated in a variety of samples, but, regarding FL, Hg is the metal that receives most of the attention. Additionally, REE are important components of the phosphor material of FL and other electronic equipment that has been discarded more and more, yet little is known about their toxicity in humans or its behavior and impacts on the environment. Considering its composition, FL can contaminate soils and waters if not properly discarded, resulting in human and animal exposure to toxic metals whose effects may not yet be fully known and can continue for years after disposal.

Many countries have taken measures to prevent the environmental impacts that may result from the disposal of spent FL. However, there is still a long road ahead considering the global scenario. Most bulbs are not safely disposed of even in developed countries, and it becomes particularly worrisome when we consider the situation in less developed countries, which lack regulations on e-waste treatments and deal not only with their e-waste but also with the wastes received from developed countries. Establishing procedures and regulations on handling spent FL and e-waste is extremely necessary for many countries to minimize the current contamination risk. Raising the population's awareness of the dangers of fluorescent lamps and providing incentives for the correct disposal of FL and replacement with LED lamps are also ways to minimize impacts associated not only with the lamps' end-of-life but also with energy consumption. The higher energy efficiency of LED and the

absence of Hg makes these lamps a great alternative for FL in terms of environmental safety, but it might still take a few years to replace most of the FL. Nevertheless, replacing FL with LED needs to be accompanied by effective recycling of the discarded FL. Many countries have no formal established program to recycle FL or have low rates of recycling; therefore, establishing such programs and increasing their coverage and capacity is also essential to reduce the environmental impacts of FL.

In addition, phosphor treatment processes could be beneficial to the environment. Efficient procedures for recovering metals from FL allow not only to minimize their release into the environment but also the use of these metals in other production processes, reducing the need for obtaining them from natural sources. Many studies achieved high recovery rates for REE and Hg through different procedures. Methods for resource recovery from FL should be further explored, and their large-scale application should be investigated. Nevertheless, it is still important to be concerned with the recovery procedures applied in order to avoid an increase in environmental impact due to the chemicals used in the process.

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

No data was used for the research described in the article.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jece.2022.108915.

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