

Article



The LCA Methodology for Ceramic Tiles Production by Addition of MSWI BA

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Abstract: Integrated waste management and sustainable use of natural resources are the basis of the Green Economy. In this context, the management of the Municipal Solid Waste Incineration Bottom Ashes (MSWI BA) is one of the current issues worldwide. This paper presents an application of the Life Cycle Assessment (LCA) procedure to the industrial production of ceramic tiles using bottom ashes in the mixture together with feldspathic sands and clays. The comparison between ashes and traditional mixture showed a similar mineralogical and rheological composition. In the reported procedure the MSWI BA, after storage, were treated to separate and recover metals. The residual ashes were added to the mixture and then they followed the traditional industrial production cycle. Samples of the different materials were taken during the experimental industrial activity and leaching tests were carried out to verify the environmental compatibility of MSWI BA use to produce ceramic tiles. The results of the LCA show large environmental and energy benefits related to the proposed reuse of BA. Metal recovery and lower use of clay in traditional mixture avoids emission of substances with a negative potential impact for environment. This study provides a sustainable alternative to the MSWI BA final disposal in landfill as MSWI BA are hazardous wastes that present complicated management and high disposal costs.

Keywords: bottom ashes; ceramic tiles; green economy; waste management; LCA

1. Introduction

In waste management, the measures used for waste recovery must be adopted with priority. The goal of disposal is to allocate material that cannot be reused in some other way. Therefore, a continuous search for innovative methods that allow the reuse of waste and not its disposal, is the task of an advanced society aiming towards sustainable development.

As reported by the Italian law (Legislative Decree n° 152/2006, art. 179), "Recycling" and "Recovery of the other types" must be preferred to "Landfill disposal". Therefore, companies can include reuse and recovery of waste products in their production cycle, promoting the development of the Green Economy. Experimental industrial activity, i.e., the focus of this study, is in accordance with the Legislative Decree n° 152/2006 and is also in agreement with the Regional Smart Specialization Strategy. The Strategy proposes experimentation and commercialization of "bio-building" materials. The use of Municipal Solid Waste Incineration Bottom Ashes (MSWI BA) in the production of ceramic tiles is consistent with the previsions for their treatment from chapter 4.6.9 ("Bottom ash treatment using

thermal systems") of the European Commission document "Reference Document on the Best Available Techniques for waste incineration". In Italy, Ministerial Decree 05/02/98 allows the recovery of bottom ashes from incineration in cement products (R5), however, there are no specific recommendations about the use of MSWI BA in the production of ceramic tiles. In this context, the authors drew up a procedural Protocol to achieve the technical compliance of floor tiles. In this Protocol, authorized by Regional Government Institution, there is a description of the tests to be carried out on materials involved in the reported study.

Under the framework of integrated waste management, the main goal is to use society's waste in a sustainable way: Reuse of waste is an environmental and economic virtuous process. First, it reduces the amount of waste to be landfilled and second it allows the recycling of materials. The reuse of waste is also important for the economy because it is a way to relieve companies from difficulties by economic savings.

In this context, incineration represents a valid solution to avoid landfilling for different materials considered as solid wastes (i.e., Automotive Shredder Residual, ASR) [1–5] and, at the same time, it can be a source of energy. In Italy, according to the Superior Institute for Environmental Protection and Research, the active incinerators are 41 for a production of 1.4 million tons of Bottom Ashes [6]. Italy is the third nation in Europe with the highest number of incinerators, after France and Germany, which had respectively 128 and 80 active incinerators [6]. The European average of incinerated municipal solid waste is about 125 (kg/person*year), whereas in Italy it is approximately 90 (kg/person*year). Due to the considerable MSWI BA mass produced, different technological proposals are born for their recovery.

The recycling of MSWI BA could be transformed in an important resource, in fact this matter has been the argument of several scientific papers for more than 15 years [7–10]. In the literature there are reported different experiences showing the possibility to use MSWI BA as a component in concrete production [8,11], as an upgrading material in concrete aggregates production, or as part of the mixture for concrete products preparation [12]. There are also many examples of MSWI BA use in road construction [13,14].

Ceramic-based materials, which are manufactured at high temperatures into non-metallic and inorganic solid products (e.g., tiles, bricks, refractories), are used in several fields of engineering, due to their specific properties (i.e., good insulation and chemical resistance, high hardness, and high melting points). Ceramic-based products could be considered as ideal candidates for the incorporation of recycling materials, like MSWI BA, as a substitute of valuable and finite natural resources [15] exactly for the above-mentioned properties.

The present work deals with an experimental project in the south part of Latium Region, where an industrial plant was modified and adapted to produce ceramic tiles. The modified procedure is based on the addition of bottom ashes to the traditional mixture [16,17].

In the first part of the paper, a broad description of the experimental industrial process is reported. In the second part, the Life Cycle Assessment (LCA) procedure is used to evaluate the environmental impacts due to the production of ceramics materials using BA deriving from MSWI.

The LCA analysis aims to provide preliminary indications on the potentialities of the hypothesized MSWI BA recovery, evaluation of cumulative energy demand, and indicators of environmental impact.

The environmental benefit of a project, like the one presented, is not only connected to waste valorization, but it is also related to the consequent minor use of natural raw material and, obviously, to a more stable final disposal of these materials [18]. Using a given percentage (20–30%) of ashes to replace clay in the traditional mixture, it is possible to reduce exploitation of clay quarries. To use MSWI BA as replacement for clay it is necessary a treatment, allowing recovery of ferrous (Fe) and non-ferrous metals (NFe). The treatment allows for the preservation of the natural environment and to save energy. Specifically, the recovery of aluminum permits 95% of energy to be saved [19] compared to primary production and it also avoids extraction of primary non-renewable materials.

2. Case Study: Production Plant of Ceramic Tiles with Recovery of MSWI BA

The Bottom Ashes (BA) quantity utilized by the industrial plant to produce ceramic tiles can be estimated in around 32,000 tons per year. At the beginning, the material is stored for two months to allow the process of carbonation to occur. At the end of this period, treatments are applied to the stabilized ashes to remove metals. Fe metals are recovered using a magnetic separator and NFe metals are divided from the BA using an eddy current technology. Metals recovery is an important step, because the absence of this treatment can affect the BA use. In particular, the presence of iron can cause discoloration, while the presence of aluminum can induce the generation of hydrogen leading to swelling and a decrease in the mechanical properties of the structure [20]. After recovery of metals, residual BA are mixed with clay and then cooked to obtain ceramic tiles. In each production step samples were taken and then they were submitted to leaching and ecotoxicological tests to ensure compliance of environmental rules.

MSWI BA Composition

In order to determine the composition of BA, laboratory tests were carried out on a sample of the ashes collected after the removal of ferrous metals (Figure 1).



Figure 1. The chemical composition of Municipal Solid Waste Incineration Bottom Ashes (MSWI BA) obtained by laboratory test.

As shown in Table 1, the experimental result is comparable to the MSWI BA composition reported in many literature studies [15,21,22]. One of the most abundant elements in the ashes is silicon, which comes from glass and ceramic materials contained in the waste. The BA composition can present variations due to different waste management of countries. However, the majority elements found in BA composition are the same in all countries (Table 2).

Table 1.	MSWI	BA elements	[21]
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Composition of MSWI BA				
Quantity	Elements			
>10,000 mg/kg 1000 mg/kg > x > 10,000 mg/kg <1000 mg/kg	Si, Fe, Ca, Al, Na, K, C Mg, Ti, Cl, Mn, Ba, Zn, Cu, Pb, Cr Sn, Sb, V, Mo, As, Se, Sr, Ni, Co, Ag, Mg, B, Br, F, and I			

Oxide	Amount (% wt)							
	Spain	Italy	Germany	Netherlands	Japan	China	Taiwan	USA
SiO ₂	43.30	33.70	55.70	54.23	31.93	59.59	50.30	23.64
CaO	16.90	35.00	11.90	13.45	33.40	7.58	15.27	23.82
Fe ₂ O ₃	14.10	5.37	8.80	13.83	5.97	5.50	7.72	17.05
Na ₂ O	7.58	2.27	1.40	2.81	2.53	1.32	1.30	1.70
Al_2O_3	5.80	13.31	14.10	7.86	16.65	18.61	16.43	14.25
MgO	2.22	4.62	2.70	1.81	3.33	1.32	n.d.	1.85
K ₂ O	1.11	1.66	1.20	0.88	2.22	2.29	2.14	0.42

Table 2. Chemical composition of MSWI BA [22].

The main mineralogical phases that are generally found in the MSWI BA are: Anhydrite, Calcite, Ettringite, Portlandite, Quartz, Feldspars, Dolomite, Galenite, and Magnetite. The mineralogical and rheological compatibility between the ashes and the traditional mixture can be affirmed by comparing their composition.

Leaching tests were performed on the collected samples using the standard procedures reported in UNI 10802 and in UNI EN 12457-2 (Compliance Test for Leaching of granular waste materials). Figure 2 and the Figure 3 show the measured concentrations of the substances before and after the duration tests i.e., freeze-thaw cycles (respectively "Tiles" and "Aged Tiles").



Figure 2. Concentrations of the substances measured in the leaching test before the duration test on the "Tiles".



Figure 3. Concentrations of the substances measured in the leaching test after the duration test on the "Aged Tiles".

In Figure 3 it can be noted that the concentrations of all the analytes, except for sulphates, decrease after the aging process of the durability tests.

The obtained results from the leaching tests on the tiles show the suitability of the MSWI BA to be reused to produce ceramic tiles.

3. LCA Procedure

LCA is a procedure to evaluate the environmental burdens associated with a product, a process, or an activity by quantifying used energy and materials and wastes released to the environment. LCA is also applied to assess the impact of used and released energy and materials, and to evaluate opportunities to enhance the environment. The assessment includes the entire life cycle of the product, process, or activity and thus it encompasses extraction and processing of raw materials, manufacturing, transport, distribution, use, reuse, maintenance, recycling, and final disposal [23]. ISO 14040 sets out the principles and frameworks for life cycle assessment described in different literatures [24–26] and it includes: definition of the LCA goal and scope, the Life Cycle Inventory (LCI) analysis phase, the Life Cycle Impact Assessment (LCIA) phase, the life cycle interpretation phase, a critical review of the LCA, limitations of the LCA, the relationship between the LCA phases, conditions for use of the chosen values, and optional elements [27,28].

3.1. Goal and Scope Definition

The goal of the applied LCA procedure is to evaluate the environmental impacts (avoided and produced) due to reuse of inert materials from bottom ashes, in the production of ceramics tiles.

The LCA procedure was performed for two scenarios (1 and 2) and their results were compared. To evaluate the LCA steps, the environmental indicators were calculated based on a time of 100 years, thus to avoid uncertainties due to eventual considerations regarding long exposure time.

Scenario **1** describes the hypothesis of BA disposal in landfill (Figure 4). It is assumed to transport BA to the closest landfill site for special and hazardous waste whose distance from the incinerator is 32 km. In this scenario the impacts caused by transport emission and by leachate were regarded. To evaluate the composition of leachate we used the results of the previously described test (Figure 1) and the data directly available from the industrial plant considered.

Scenario **2** describes the processes to recover materials from BA and the subsequent activities of their recycling (Figure 4). It is assumed that recycled NFe and Fe are respectively reused for secondary production of aluminum and steel, avoiding manufacture of the same quantity of primary materials. The LCA was applied considering the impacts produced by BA transport from incinerator to the industrial plant and by the activities to recover Fe and NFe (Figure 4). The avoided impacts caused by BA disposal in landfill and by the activities related to the primary production of metals and of inertial materials were also regarded (Figure 4).

Specifically, recovery of NFe avoids primary extraction of 4 kg of bauxite for each kg of aluminum [29] and recovery of Fe avoids primary extraction of 1.37 kg of magnetite for each kg of iron scraps [30]. In this study extraction of primary minerals was assumed to be carried out in the closest quarry to the industrial plant. The quarries for bauxite and magnetite extraction are located respectively 262 and 3000 km from the ceramic tiles plant. Moreover, while the secondary production of aluminum involves energy saving, for secondary iron it is assumed that Fe scraps are ready to be sent to foundry involving the same energy spending of primary production. The treatment of Fe minerals consists of different milling processes [31] that present similarities with the treatment of BA. For the metal's recovery, it was considered that the oxidation of them during the incineration [32] and the quantitation of impurities during the re-melting of scrap can cause both lower recycling returns between the primary product and the secondary product [33]. Thus, a foundry performance of 90% is assumed. The recycled inert material is reused as a substitution for the clay at a quantity of 30% in the mixture to obtain ceramic tiles, thus it was assumed to represent the missed extraction of clay for the same quantity. The quarry for clay extraction is located at 1660 km from the production plant.



Figure 4. Comparison between the activities of Scenario 1 (BA disposal in landfill) and Scenario 2 (materials recovery and BA reuse).

3.2. Modeling Framework and Life Cycle Inventory

The software used for the LCA application was derived from the implementation of the Solid Wastes Flows Optimization program for the integrated management of Municipal Solid Waste according to the LCA method, developed by "La Sapienza" University of Rome. The software provides air, soil, water emissions (related to fuel and energy consumption) and environmental impact indicators. Additional data refer to activities due to treatment of BA with material recovery. The data used in this paper are a combination of literature and experimental values (Table 3). The comparison between them and the values range proposed in literature shows the suitability of the used data in our LCA application (Table 4).

Table 3. Input data used	for the Life Cycle Assessme	nt (LCA) process.
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Scenario 1—Landfill Disposal of BA								
Process	Resource	Value	Unit	Impact				
BA transport from incinerator to landfill [34]	Fuel	2.8-machine capacity 28 t	Km/l	Direct				
Leachate (data obtained from the experimental test)	-	10% lost humidity	l/year	Direct				

Scenario 2—Treatment of BA for Production of Ceramic Tiles for Fe and NFe Recycling							
Process	Resource	Value	Unit	Impact			
BA transport from incinerator to industrial plant [34]	Fuel	2.8-machine capacity 28 t	Km/l	Direct			
BA treatment [35]	Electric energy	4	kWh/t	Indirect			
Transport of recovered NFe from industrial plant to foundry [34]	Fuel	2.8-machine capacity 28 t	Km/l	Direct			
Transport of recovered Fe from industrial plant to foundry [34]	Fuel	2.8-machine capacity 28 t	Km/l	Direct			
Aluminum secondary production [19]	Electric energy	0.75	kWh/kg	Indirect			
Clay transport [34]	Fuel	2.8-machine capacity 28 t	Km/l	Avoided			
Clay extraction [36]	Fuel	17.3-machine potential 80 kW	l/h	Avoided			
Bauxite extraction [36]	Fuel	17.3-machine potential 80 kW	l/h	Avoided			
Bauxite transport to foundry [34]	Fuel	2.8-machine capacity 28 t	Km/l	Avoided			
Bayer treatment [19]	Electric energy	4	kWh/kg	Avoided			
Electrolytic process [19]	Electric energy	20	kWh/kg	Avoided			
Ferrous mineral extraction [36]	Fuel	30.3-machine potential 140 kW	l/h	Avoided			
Ferrous mineral transport from quarry to foundry [34]	Fuel	2.8-machine capacity 28 t	Km/l	Avoided			
Ferrous mineral treatment [31]	Electric energy	10	kWh/t	Avoided			

Table 3. Cont.

Table 4. Comparison between the used LCA input values and values proposed in literature.

Process	Resource	Value	Unit	References
		4 *		[35]
BA treatment	Electric energy	10	kWh/t	[37]
		8		[38]
Looghato	_	10% lost humidity *	1/woor	**
Leachate		7% lost humidity	i/year	[35]
Aluminum secondary	Electric operation	0.75 *	kWh/ka	[19]
production	Electric energy	2	KVVII/Kgaluminum	[39]
		20 *		[19]
Electrolytic process	-	13–17	kWh/kg _{aluminum}	[39]
		12.9–15.5		[40]

* Value used in the described LCA application. ** Value obtained from the reported experimental industrial activity.

Based on the information acquired from the considered industrial plant, the material balance can be summarized as follows (Figure 5):

- BA arriving at the industrial plant contain 13% of humidity. During storage, they lose 7% of initial humidity by evaporation and leaching. The produced leachate represents 10% of humidity loss and it is collected through a sewage system.
- After BA storage, metals are separated by the ashes. A mass of Fe corresponding to 21% of the total ashes is recovered using a magnetic separator belt and a magnetic separator drum. Recovery of NFe is about 3% of the total ashes and was obtained by an Eddy Current Separator (ECS).
- The residual BA is utilized to produce the ceramic tiles.

The choice of the technologies and of raw material supply used in the study is based on data provided by the industrial plant.

By comparing information sheets from different treatment plants [35], it follows that the recovery of Fe and NFe are respectively higher at 11% and 2% compared to other treatment plants in Italy.



Figure 5. The material balance of ashes during the recovery treatment.

3.3. Life Cycle Impact Assessment

The main task of this phase is to convert LCI data in aggregated indexes able to measure the environmental impacts of the system. Due to the lack of a generally accepted way of assessing the value of the damage to ecosystems, it is possible to refer to the guidelines expressed by the ISO regulations 14041 and 14042—"Environmental management Life Cycle Assessment" [27,28]. They are based on the following procedure:

- 1. selection of impact categories;
- 2. classification;
- 3. characterization.

Six categories of potential impacts have been identified:

- 1. Global Warming Potential (GWP);
- 2. Photochemical Ozone Creation Potential (POCP);
- 3. Acidification Potential (AP);
- 4. Eutrophication Potential (EP);
- 5. Human Toxicity Potential (HTP);
- 6. Resources Depletion Potential (RDP).

The above listed impact categories are the more commonly used for assessment of the impact on the global as well as the regional scale. This is because they have been more widely studied and introduced in many international protocols for Environmental Quality Control, signed by many nations worldwide.

In the classification step, all substances are sorted into classes according to the effect they cause on the environment. Certain substances can be included in more than one class.

Once substances are aggregated within each class, it is necessary to produce an effect potential (EfP). This cannot be done by simply adding up the quantities of substances involved, as some

substances may have a more intense effect than others. To overcome this problem weighting factors, generally referred to as Equivalence Factors (EFs), are applied to the different substances, according to the Environmental Design of Industrial Products (EDIP) characterization method [41,42].

The EF of a given substance represents the potential impact of a unit mass of that substance relative to some reference substances, referred to as the common unit of the category indicator.

For example, referring to GWP, the common unit is carbon dioxide (CO_2) and consequently the EF of a given greenhouse gas i (EFi) is defined as:

$$EF^{GW_i} = \frac{intensity \ of \ the \ effects \ on \ GW \ due \ to \ gas_i}{intensity \ of \ the \ effects \ on \ GW \ due \ to \ CO_2}$$
(1)

The list of EFs assumed in this study are reported in Table 5 [43]. As a consequence, the global potential effect for each impact category can be calculated as the amount of emissions multiplied by their specific EF:

$$EfP_j = \sum_i (Q_i \times EF_{ij}) \tag{2}$$

where Qi is the total of the i-th substance produced (t/year) and EFij is the Equivalence Factor of the i-th substance referred to the j-th impact class.

	Global Warming (Time = 100 years) "EDIP"					
Formula	Substance	Equi	valence Factor			
Torniulu	Substance _	Value	U.M. (100 years)			
CO ₂	Carbon dioxide	1	g CO ₂ /g substance			
N ₂ O	Nitrous oxide	320	$g CO_2/g$ substance			
CH_4	Methane	25	$g CO_2/g$ substance			
CFCl ₃	CFC-11	4000	$g CO_2/g$ substance			
CF_2Cl_2	CFC-12	8500	$g CO_2/g$ substance			
CF ₃ Cl	CFC-13	11,700	$g CO_2/g$ substance			
CF ₂ ClCF ₂ Cl	CFC-114	9300	g CO ₂ /g substance			
	CFC-116	12,500	g CO ₂ /g substance			
CCl_4	Tetrachloromethane	1400	g CO ₂ /g substance			
HCFC22	HCFC22	1700	$g CO_2/g$ substance			
HCFC141b	HCFC141b	630	$g CO_2/g$ substance			
HCFC142b	HCFC142b	2000	$g CO_2/g$ substance			
CO	Carbon Monoxide	2	$g CO_2/g$ substance			
CF ₃ Br	Halon 1301	6200	$g CO_2 / g substance$			
	Acidification ("EDIP")				
Formula	Substance	Equivalence Factor				
Torntula	Substance	Value	U.M. (100 years)			
SO ₂	Sulfur dioxide	1	$g SO_2/g substance$			
SO_3	Sulfur trioxide	0.8	$g SO_2/g$ substance			
NO ₂	Nitrogen dioxide	0.7	g SO ₂ /g substance			
NO	Nitrogen monoxide	1.07	g SO ₂ /g substance			
HNO ₃	Nitric acid	0.51	g SO ₂ /g substance			
H_2SO_4	Sulfuric acid	0.65	g SO ₂ /g substance			
H_3PO_4	Phosphoric acid	0.98	$g SO_2/g$ substance			
H_2S	Hydrogen sulfide	1.88	g SO ₂ /g substance			
HF	Hydrofluoric acid	1.6	g SO ₂ /g substance			
HCl	Hydrochloric acid	0.88	g SO ₂ /g substance			
NH ₃	Ammonia	1.88	g SO ₂ /g substance			

Table 5. Equivalence Factors (EFs) [39].

	Eutrophication ("EDIP")							
Formula	Substance	Value	Equivalence Factor U.M. (100 years)					
cvanide	Cvanide	2.38	g NO ₃ /g substance					
Ntot	Total nitrogen	4.43	g NO ₃ /g substance					
N ₂ O	Nitrous oxide	2.82	g NO ₂ /g substance					
NH ₂	Ammonia	3.64	$g NO_2/g$ substance					
Ptot	Total phosphorus	32.03	$g NO_2/g$ substance					
1 tot	Photochemical Ozone (Treation ("FI						
			Environter of Footon					
Formula	Substance	Value	U.M. (100 years)					
CH_4	Methane	0.03	g Ethylene/g substance					
	Ethane	0.3	g Ethylene/g substance					
	Propane	1.2	g Ethylene/g substance					
	Butane	1.2	g Ethylene/g substance					
	Hexane	1.5	g Ethylene/g substance					
	Heptane	1.7	g Ethylene/g substance					
	Alkane	1.2	g Ethylene/g substance					
	CHarom.	0.048	g Ethylene/g substance					
	Methanol	0.21	g Ethylene/g substance					
	Acetone	0.27	g Ethylene/g substance					
	Butene	1.2	g Ethylene/g substance					
	Benzene	0.45	g Ethylene/g substance					
	Taluana	0.45	g Ethylene/g substance					
	EthilBonzono	0.05	g Ethylono/g substance					
	Eunidenzene	1.1	g Ethylene/g substance					
	Formaldenyde	0.36	g Ethylene/g substance					
	Acetaldenyde	1.2	g Ethylene/g substance					
	Aldehyde	1.3	g Ethylene/g substance					
	VOC	0.808	g Ethylene/g substance					
	Human toxicity	(USES 2.0)						
Formula	Substance	Value	Equivalence Factor					
		value	U.M. (100 years)					
air	Antino ant	6200	a 14 dishlarahangana					
30	Amminony	270.000	g _{eq} 1-4-dictionoberizene					
AS	Arsenic	370,000	g_{eq} 1-4-dichlorobenzene					
Ba	Barium	/10	g _{eq} 1-4-dichlorobenzene					
Ca	Cadmium	160,000	g _{eq} 1-4-dichlorobenzene					
Co	Cobalt	19,000	g _{eq} 1-4-dichlorobenzene					
Cu	Copper	4700	g _{eq} 1-4-dichlorobenzene					
Pb	Lead	360	g _{eq} 1-4-dichlorobenzene					
Hg	Mercury	1200	g _{eq} 1-4-dichlorobenzene					
Mo	Molybdenum	4900	g _{eq} 1-4-dichlorobenzene					
Ni	Nickel	38,000	g _{eq} 1-4-dichlorobenzene					
Se	Selenium	43,000	g _{eq} 1-4-dichlorobenzene					
Sn	Tin	1.2	g _{eq} 1-4-dichlorobenzene					
V	Vanadium	6000	g _{eq} 1-4-dichlorobenzene					
Zn	Zinc	110	g _{eq} 1-4-dichlorobenzene					
NH_3	Ammonia	1	g _{eq} 1-4-dichlorobenzene					
H_2S	Hydrogen sulfide	0.77	g _{eq} 1-4-dichlorobenzene					
HCI	Hydrogen chloride	2.40	g _{eq} 1-4-dichlorobenzene					
C ₂ H ₄	Ethvlene	0.69	g _{eq} 1-4-dichlorobenzene					
CH ₂ O	Formaldehyde	0.91	g _{og} 1-4-dichlorobenzene					
C _c H _c	Benzene	2000	g _{eq} 1-4-dichlorobenzene					
C ₆ H ₋ CH ₋	Toluono	0.26	σ_{-1} - dichlorobonzono					
C.H_OU	Phonols	0.50	$g_{eq} = 4$ disblorshop zero					
	Chloreform	0.37	g _{eq} 1-4-dichlorobenzene					
	Dishlaraath	12	g_{eq} 1-4-ulchiorobenzene					
$1,2CH_2CICH_2CI$	Dichloroethane	7	g _{eq} 1-4-aichlorobenzene					

Table 5. Cont.

	Human toxicity (USES 2.0)						
Formula	Substance	Equi	valence Factor				
roimula	Substance	Value	U.M. (100 years)				
water							
As	Arsenic	880	g _{eq} 1-4-dichlorobenzene				
Ва	Barium	570	g _{eq} 1-4-dichlorobenzene				
Cd	Cadmium	23	g _{eq} 1-4-dichlorobenzene				
Cr	Chrome	2.1	g _{eq} 1-4-dichlorobenzene				
Со	Cobalt	99	g _{eq} 1-4-dichlorobenzene				
Cu	Copper	1.3	g _{eq} 1-4-dichlorobenzene				
Pb	Lead	12	g _{eq} 1-4-dichlorobenzene				
Hg	Mercury	250	g _{eq} 1-4-dichlorobenzene				
Mo	Molybdenum	5000	g _{ea} 1-4-dichlorobenzene				
Ni	Nickel	310	g _{og} 1-4-dichlorobenzene				
Se	Selenium	51,000	g _{eq} 1-4-dichlorobenzene				
Sn	Tin	0.017	g _{og} 1-4-dichlorobenzene				
V	Vanadium	2900	g _{eq} 1-4-dichlorobenzene				
Zn	Zinc	0.57	g _{eq} 1-4-dichlorobenzene				
CH ₂ O	Formaldehyde	0.04	g_1-4-dichlorobenzene				
C/H/	Benzene	1900	g_1-4-dichlorobenzene				
C H-CH	Toluene	0.33	g 1-4-dichlorobenzene				
C ₄ H ₋ OH	Phenols	0.054	g 1-4-dichlorobenzene				
coil	1 Henois	0.034	geq1-4-dicitiorobenzene				
5011	Arconic	400	a 14 dichlarabanzona				
AS Cd	Cadmium	490	g 1.4 dichlorobonzono				
Cu	Cabalt	90	geq1-4-dichlorobenzene				
Co	Coball	01	g _{eq} 1-4-dichlorobenzene				
Cu	Copper	3.Z	g _{eq} 1-4-dichlorobenzene				
PD	Lead	180	g _{eq} 1-4-dichlorobenzene				
Hg	Mercury	200	g _{eq} 1-4-dichlorobenzene				
MO	Molybdenum	2800	g _{eq} 1-4-dichlorobenzene				
N1	Nickel	160	g _{eq} 1-4-dichlorobenzene				
Se	Selenium	25,000	g _{eq} 1-4-dichlorobenzene				
11	Thallium	100,000	g _{eq} 1-4-dichlorobenzene				
Sn	Tin	32	g _{eq} 1-4-dichlorobenzene				
V	Vanadium	1600	g _{eq} 1-4-dichlorobenzene				
Zn	Zinc	0.35	g _{eq} 1-4-dichlorobenzene				
C ₆ H ₅ Cl	Chlorobenzene	7.1	g _{eq} 1-4-dichlorobenzene				
	Depletion of non-ren	ewable resources					
Formula	Substance	Res	erve available				
Formula	Substance	value	U.M. (100 years)				
		R(i)	F(i): frac of reserve				
	Oil (in ground)	2.4×10^{14}	4.17×10^{-15}				
	Natural gas	1.3×10^{14}	7.69×10^{-15}				
U	Uranium (hours)	1.3×10^{10}	7.69×10^{-11}				
Cu	Copper (hours)	6.1×10^{11}	1.64×10^{-12}				
Ph	Lead (hours)	1.2×10^{11}	8.33×10^{-12}				
Ni	Nickel	1.2×10^{11}	9.09×10^{-12}				
Zn	Zinc	3.3×10^{11}	3.03×10^{-12}				
	Rauvito	3.5×10^{13}	3.00×10^{-14}				
F203	Iron	2.0×10^{-1}	5.57×10^{-14}				
ге Ма	Manganasa	1.0×10^{12}	1.00×10^{-13}				
	Cileran	5.0×10^{-2}	2.00×10^{-9}				
Ag	Suver	$4.2 \times 10^{\circ}$	2.38×10^{-16}				
	Coal (in ground)	3.0×10^{13}	3.33×10^{-10}				

Table 5. Cont.

The potential impacts can be classified as reported below:

- Direct impacts, which account for effects directly related to a given impact class;
- **Indirect impacts**, which account for possible effects associated to a given impact category due to a transformation after primary emission;
- Avoided impacts, which consider the saved impacts due to the presence of profitable outputs. They are equivalent to the impacts that would have occurred in actual production of the same amount of recovered energy and they need to be deducted from the impacts caused by other processes.

The considered impacts in the two described scenarios are reported in Table 3 and shown in Figure 4. In the first scenario the direct impacts produced by BA transport from incinerator to landfill and by leachate were regarded. In the second scenario the direct impacts related to the transport of BA from incinerator to industrial plant and of recovered metals to foundry were accounted. In scenario 2 we considered also indirect and avoided impacts. Specifically, the indirect impacts produced by energy consumption for BA treatment and for Al secondary production were taken into account. The avoided impacts related to the first scenario and to the primary production of NFe (Bauxite extraction, its transport from quarry to foundry, Bayer treatment, and Electrolytic process), Fe (Ferrous minerals extraction, their transport from quarry to foundry, and their treatment) and inert material (clay extraction and transport from quarry to the industrial plant) were regarded.

GWP

Global Warming is a phenomenon caused by greenhouse gases. Natural greenhouse gases are water vapor (H_2O), methane (CH_4), carbon dioxide (CO_2), nitrogen peroxide (N_2O), and ozone (O_3).

GWP is calculated for each greenhouse gas (GWP_i), considering their radiation absorption capacity and time spent in the atmosphere. For its quantification standardization factors are used, which refer to the kg of equivalent CO_2 .

AP

Air acidification is an environmental problem involving air, water, and soil. A substance causes a contribution to acidification if it is able to increase the concentration of hydrogen ions in the environment. Standardization factors that refer to the kg of equivalent SO_2 are used for its estimation.

EP

Eutrophication is an impact on the ecosystem caused by an excessive supply of substances (nutrients) containing nitrogen (N) or phosphorus (P).

The standardization of eutrophication takes place by reporting the quantities of the inventoried substances to the kg of NO_3 or to the kg of PO_4 equivalents.

POCP

Photochemical ozone creation is a phenomenon due to the presence of unburnt hydrocarbons and nitrogen oxides that contribute to ozone formation in presence of solar radiation. Ethylene is used for the standardization of POCP, therefore, evaluation takes place expressing the gases capable of producing ozone in kg of equivalent ethylene.

HTP

The potential of human toxicity is an equivalent factor of toxic potential that is introduced by the definition of potential harm of a released chemical unit. Standardization factors that refer to the kg of equivalent dichlorobenzene are utilized for the HTP evaluation.

RDP

Resource depletion is defined as the decrease in the availability of natural resources. In this study the method proposed by the Centre of Environmental Science of Leiden University [44] was

used to express the reduction of non-renewable resources. This method expresses the consumption of non-renewable resources in terms of the fraction of known reserves, adopting an appropriate weight factor (Fi):

$$F_i = \frac{1}{R_i} \tag{3}$$

where R_i (kg) indicates the reserve still available referred to the analyzed resource.

The total value of the R_i index, which quantifies the overall impact on the category in question, is therefore calculated as follows:

$$RDP = \sum_{i} F_i * m_i \tag{4}$$

where Fi is the weight factor of the i-th consumed resourced and mi is the mass (kg) of consumed resource.

4. Results and Discussion

The LCA results, reported in Table 6, are obtained using the whole BA quantity utilized by the industrial plant (32,000 t/year).

	Unit	Scen. 1-BA	Scen. 2-Inert	Scen. 2-NFe	Scen. 2-FE
		Landfill	Recovery	Recovery	Recovery
GWP					
Direct impacts	t CO _{2eq} /year	28.500	241.000	29.500	107.000
Indirect impacts	t CO _{2eq} /year	0.000	69.100	-6880.000	-42.900
Avoided impacts	t CO _{2eq} /year	0.000	-1680.000	-12.100	-1190.000
Total impacts	t CO _{2eq} /year	28.500	-1370.000	-6860.000	-1120.000
AP	-				
Direct impacts	t SO _{2eq} /year	0.460	0.838	0.062	0.613
Indirect impacts	t SO _{2eq} /year	0.000	0.527	-52.400	-0.327
Avoided impacts	t SO _{2eq} /year	0.000	-16.000	-0.297	-11.200
Total impacts	t SO _{2eq} /year	0.460	-14.600	-52.600	-10.900
EP	*				
Direct impacts	t NO _{3eq} /year	0.022	0.000	0.062	0.000
Indirect impacts	t NO _{3eq} /year	0.000	0.006	-0.561	-0.004
Avoided impacts	t NO _{3eq} /year	0.000	0.000	0.000	0.000
Total impacts	t NO _{3eq} /year	0.022	0.006	-0.500	-0.004
POCP					
Direct impacts	t Ethylene _{eq} /year	0.052	0.095	0.007	0.070
Indirect impacts	t Ethylene _{eq} /year	0.000	0.073	-7.230	-0.045
Avoided impacts	t Ethylene _{eq} /year	0.000	-1.810	-0.034	-1.270
Total impacts	t Ethylene _{eq} /year	0.052	-1.640	-7.260	-1.250
HTP					
Direct impacts	t 1-4-dichlorobenzene _{eq} /year	1.000	0.000	0.000	0.000
Indirect impacts	t 1-4-dichlorobenzene _{eq} /year	0.000	0.007	-0.651	-0.004
Avoided impacts	t 1-4-dichlorobenzene _{eq} /year	0.000	0.000	0.000	0.000
Total impacts	t 1-4-dichlorobenzene _{eq} /year	1.000	0.007	-0.651	-0.004

Table 6. Results of LCA for the two considered scenarios.

The final energy audit is negative (Figure 6), where negative values indicate that avoided impacts are greater than the added impacts to the environment due to the recovery activity. The higher energy saving is due to the recovery of NFe, because for the secondary production of aluminum is one-twentieth of the necessary energy for primary production (aluminum is the metal with the highest energy intensity needed for extraction on a world scale [19]). However, the drawbacks of using low-quality Al scrap in refining processes were not included because of a lack of quantitative data, suggesting actual benefits could be somewhat smaller [45].

Ξ

-30,000,000

-40,000,000

Scen 1 - B.A. Scen. 2 - Fe Scen. 2 - NFe Scen. 2 - Inert landfill recovery recovery recovery

Energy audit

Figure 6. Comparison of energy audit between scenario 1 and activities of scenario 2.

The avoided emissions of greenhouse gases equivalents are related to the recovery of aluminum (Figure 7), due, as above mentioned, to the large difference in energy demand between primary and secondary aluminum production. The remaining part is related to the recovery of inert materials and of Fe scraps, this is connected to the avoided impacts due to transport and extraction consumptions. The emissions produced by BA disposal in landfill can be overlooked with respect to the avoided emissions deriving from BA treatment with materials recovery.



Global Warming Potential

Figure 7. Comparison of GWP between scenario 1 and activities of scenario 2.

AP and POCP show positive results (Figures 8 and 9), this means potential emissions are not introduced in environment. The greater benefits are due to NFe recovery. Regarding AP, potential impacts were caused by gaseous emissions of SO_2 from combustion of fossil fuels for electricity production. Indeed, for POCP, potential impacts were caused by emissions of methane and Volatile Organic Compounds (VOC) produced by transport.



Acidification Potential

Figure 8. Comparison of AP between scenario 1 and activities of scenario 2.

Photochemical Ozone Creation Potential





In the graphics of EP and HTP analysis, the most interesting result concerns the hypothesis of BA disposal in landfill (Figures 10 and 11).



Eutrophication Potential

Figure 10. Comparison of Eutrophication Potential (EP) between scenario 1 and activities of scenario 2.



Human Toxicity Potential

Figure 11. Comparison of Human Toxicity Potential (HTP) between scenario 1 and activities of scenario 2.

The results provide evidence that the BA disposal can avoid the issue of heavy metal with high potential of human toxicity [46]. The substances issued in the water with EP and HTP are reported in Table 7, where the leaching tests of BA samples are shown.

Parameter	Unit	Limit Values	Bottom Ashes 1	Bottom Ashes 2	Bottom Ashes 3	Bottom * Ashes 4	Bottom * Asesh 5	Bottom * Ashes 6
Fluorides	mg/L	≤1.5	0.121	0.138	0.16	< 0.9	<0.9	< 0.9
Chlorides	mg/L	≤100	78.3	113	165	78.9	173	168
Nitrates	mg/L	≤50	2.27	2.24	2.44	4.3	<1	<1
Sulphates	mg/L	≤250	3.53	3.64	4.32	<5	<5	8.8
Cyanides	mg/L	≤0.05	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01	< 0.01
Arsenic	mg/L	≤0.05	< 0.01	< 0.01	< 0.01	0.005	< 0.005	< 0.005
Barium	mg/L	≤1	4.18	2.76	2.5	4.4	3.4	4.2
Beryllium	mg/L	≤0.01	< 0.005	< 0.005	< 0.005	< 0.0005	< 0.0005	< 0.0005
Cadmium	mg/L	≤0.005	< 0.002	< 0.002	< 0.002	0.0003	< 0.0001	< 0.0001
Cobalt	mg/L	≤0.25	< 0.002	< 0.002	< 0.002	< 0.002	< 0.002	< 0.002
Total Chrome	mg/L	≤0.05	0.009	0.011	0.013	0.008	0.016	0.011
Copper	mg/L	≤0.05	0.085	0.095	0.07	0.059	0.08	0.076
Mercury	mg/L	≤ 0.001	< 0.001	< 0.001	< 0.001	< 0.0005	< 0.0005	< 0.0005
Nickel	mg/L	≤0.01	< 0.005	< 0.005	< 0.005	< 0.0004	< 0.0004	< 0.0004
Lead	mg/L	≤0.05	2.3	1.39	1.13	3.8	3.6	7.2
Selenium	mg/L	≤0.01	< 0.005	< 0.005	< 0.005	< 0.005	< 0.005	< 0.005
Vanadium	mg/L	≤0.25	0.007	0.007	< 0.005	< 0.003	< 0.003	< 0.003
Zinc	mg/L	≤3	1.11	0.64	0.53	1.3	0.87	1.2
COD	mg/L	≤30	59	45	80	69	113	97.7

Table 7. Leachate tests results.

* Measures carried out in a second different laboratory.

The end life cycle of ceramic tiles mixture with BA is not taken into consideration, but tests performed on tiles show leachate values below the legal limits (Table 7). Nevertheless, the most important environmental impacts that might arise when residues are disposed in landfills or utilized in ceramics tiles are the impacts related to the presence of heavy metals and salts and the potential long-term leaching [47]. A favorable point to be considered is due to the toxicity results avoided for metallurgical activities that are excluded from the considered system. Regarding Al recycling, savings related to HTP and EP are due to avoided disposal of red mud from bauxite mining, while the avoided burdens on HTP are connected to the production of Zn, to be used as an alloying element [45].

A positive result derives from a gain in the non-renewable resources extraction. From the characterization of the BA during the experimentation phases, the substances affecting the impact indicator are copper, lead, nickel, zinc, bauxite, iron, and manganese. Although there is a higher recovery of Fe than of NFe (Figure 5), the RDP shows a better result for the case of NFe recovery (Figure 12). The obtained result is related to the aluminum indicator of RDP, which is more important than iron in terms of extractable resources. The hypothesis of disposal in landfill of BA has a null impact because materials recovery is not expected; furthermore, the activity of inert recovery is null because impact indicators of RDP do not include clay.



Resource Depletion Potential

Figure 12. Comparison of Resource Depletion Potential (RDP) between scenario 1 and activities of scenario 2.

The results of this study show large benefits related to the reuse of BA in ceramic tiles production. The described industrial experimentation represents an alternative and innovative use of MSWI BA in order to avoid their disposal in landfill. In Italy there are no specific rules about the use of MSWI BA in the production of ceramic tiles. Moreover, MSWI BA are classified as special and hazardous waste, which present high disposal costs. For these reasons, the national policies should promote development of protocols to regulate the reuse of BA in ceramic products.

5. Conclusions

The LCA methodology together with the current industrial experimentation, confirms environmental and energy benefits related to the use of MSWI BA in production of ceramic tiles. Therefore, the MSWI BA recovery system presents undoubted advantages with respect to the MSWI BA disposal. In this regard, "La Sapienza" University of Rome has carried out several leaching tests on the BA and on the produced tiles. Results were utilized to set the data used for the LCA application. By adding all the activities adopted in BA treatment together with the recovery of materials, considering all the avoided activities deriving from the recycling of recovered materials and from the BA landfilling, results show a positive judge (Table 6). The negative values of the total impacts indicate that avoided impacts are larger than the impacts caused from the activity of recovery, reuse, and final disposal. The treatment of MSWI BA that brings the recovery of NFe, Fe, inert material, and the avoided potential impacts due to the absence of landfilling, push towards environmental benefits. Benefits regarding the Energy Audit and the GWP categories are related to recovery of Al due to the large difference in energy demand between primary and derived aluminum production. Indirect impacts produced by the recovered energy during Al recycling avoid the emission of $6877 \text{ t } \text{CO}_{2eg}$ /year. At the same way, recovery of inert materials and recovery of Fe scrubs prevent GWP impacts due to transport and extraction consumptions. Specifically, inert materials recovery avoids the emission of 1681 t CO_{2eq}/year,

whereas Fe recovery prevents the release of 1188 t $CO_{2eq}/year$. A lower AP is due to the recovery of NFe, even if potential impacts were caused by emissions into the air of SO₂ from combustion of fossil fuels for electricity production. In fact, Table 6 shows that the highest avoided AP (amounting to 52.40 t SO_{2 eq}/year) is related to the indirect impacts of NFe recovery. The POCP impacts are caused by the emissions into atmosphere of methane and VOC mainly due to transport emissions. Recovery of inert material and Fe prevents emissions of 1.81 t Ethylene_{eq}/year and 1.27 t Ethylene_{eq}/year, respectively. EP impacts avoided are due to recovery of NFe (0.56 t NO_{3eq}/year), thanks to a lower emission into water of NH₃ coming from the necessary production of electricity. An important element, even if not directly involved in the LCA analysis, is the avoided risks of contamination of water due to disposal of BA in landfill. We can find an indirect evaluation of this aspect into the HTP avoided (amounting to 1 t 1-4-dichlorobenzene_{eq}/year), because it means that several heavy metals with high potential of human toxicity, as shown by the experiments carried out and confirmed by the literature data, cannot be dispersed in the environment. At the end, the RDP impacts are also positive during the treatment phases because the substances affecting impact indicators, like copper, lead, nickel, zinc, bauxite, iron, and manganese, are recovered.

Based on the positive results obtained from the LCA, the main future challenges regard the improvement of the environmental benefits caused by the BA reuse. In this context, further investigations will be performed using a different percentage of BA in the mixture to maximize the benefits and the environmental sustainability of the process. Furthermore, future research will be focused on the metals recovery system to improve their separation from the other inert materials. By avoiding heavy metal leachate during the life cycle until the disposal of the product deriving from BA reuse, it is possible to increase the positive environmental and human health impacts. In this way, the BA could be reused widely in the productive process of civil engineering, supporting the Green Economy. Regarding the industrial sector, BA reuse is an economic virtuous process because it is a way for companies to save on waste disposal and primary materials costs. Additionally, waste reuse can assist companies in obtaining labels of environmental excellence, which increases their profitability and reputation.

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Abbreviations

AP	Acidification Potential
BA	Bottom Ash
ECS	Eddy Current Separator
EF	Equivalence Factor
EFs	Equivalence Factors
EfP	Effect Potential
EDIP	Environmental Design of Industrial Products
EP	Eutrophication Potential
Fe	Ferrous metals
GWP	Global Warming Potential
HTP	Human Toxicity Potential
LCA	Life Cycle Assessment
LCI	Life Cycle Inventory
LCIA	Life Cycle Impact Assessment
MSWI	Municipal Solid Waste Incineration
NFe	Non-ferrous metals
POCP	Photochemical Ozone Creation Potential
VOC	Volatile Organic Compounds

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