UNIVERSITÀ DEGLI STUDI

DI MODENA E REGGIO EMILIA

Dipartimento di Scienze Chimiche e Geologiche

Dottorato di ricerca in

Models and Methods for Materials and Environmental Sciences

Ciclo XXXIV

Waste inertisation and recycling: environmental analysis of prototypes and industrial applications

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“To my Family”

Summary

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Abstract

The PhD was awarded by the University of Modena and Reggio Emilia (UNIMORE) and was carried out in collaboration with the National Research Council - Institute of Atmospheric Pollution Research. During the three years of my PhD financed by the CNR, I carried out research activities on the circular economy and in particular on the recycling and reuse of waste in collaboration with UNIMORE.

Several projects on circular economy have been financed by public funds and I took part in several activities, in particular related to environmental impacts and the application of new strategies for waste recycling and reuse.

I have participated in training activities offered by UNIMORE and other boards and I have attended international and national conferences with oral contribution. The period outside UNIMORE was carried out at AzzeroCo$_2$ for the application of the Life Cycle Assessment (LCA) methodology in order to evaluate the impact of recycling and reuse processes of materials that are destined for landfill.

The aim of this thesis was to analyse the viability of recycling, reuse and environmental analysis of three case studies concerning waste materials that are currently destined to landfill disposal because they cannot be recycled by the current supply chains and are therefore not used as secondary raw materials.

The three case studies were subject of three research public projects aimed at assessing the applicability of different strategies to produce secondary raw materials and the environmental impacts of their recycling.

The first case study focused on the recycling of materials containing asbestos and Man-Made Vitreous Fibres. In particular, the atmospheric emissions related to the thermal treatment process were evaluated, and the subsequent environmental impact was assessed using the LCA method.

The second case study assessed the feasibility of recycling household hard plastics. The possibility of separating the different hard plastics by their density was demonstrated, and it was possible to obtain a blend with good mechanical properties composed of post-consumer and post-industrial plastics. The LCA analysis showed a considerable reduction in environmental impacts compared to the production of new virgin plastics.

The third case study analysed the possibility of reusing cellulose from Absorbent Hygiene Products (AHP) obtained from Fater S.P.A.'s patent recycling process. Tests carried out during the project allowed to evaluate the use of recycled cellulose both for the production of insulating panels and as cellulose flakes. It was demonstrated that recycled cellulose flakes has the same properties as virgin cellulose as far as the thermal insulation of buildings is concerned and that the application of insulation panels is a valid method of use with a low environmental impact.

Il dottorato di ricerca è stato assegnato dall'Università di Modena e Reggio Emilia ed è stato svolto in collaborazione con il Consiglio Nazionale delle Ricerche - Istituto sull'Inquinamento Atmosferico (CNR-IIA). Durante i tre anni di dottorato finanziato dal CNR ho svolto attività di ricerca sull'economia circolare e in particolare sul riciclo e riutilizzo di rifiuti in collaborazione con UNIMORE.

Sono stati finanziati diversi progetti sull'economia circolare da fondi pubblici a cui ho preso parte in diverse attività, in particolare legate agli impatti ambientali e dell'applicazione di nuove strategie per il ciclo e riutilizzo di rifiuti.

Ho partecipato ad attività formative offerte da UNIMORE e da altre istituzioni e ho partecipato a convegni nazionali e internazionali con contributo orale. Il periodo esterno ad UNIMORE è stato svolto presso AzzeroCo$_2$ per l'applicazione della metodologia Life Cycle Assessment (LCA) al fine di valutare l'impatto di processi di riciclo e riutilizzo di materiali che sono destinati in discarica.

Lo scopo della tesi è stato quello di analizzare la fattibilità del riciclo, del riutilizzo e dell'analisi ambientale di tre casi studio relativi a materiali di scarto che attualmente sono destinati allo smaltimento in discarica perché non possono essere riciclati dalle attuali filiere e quindi non sono utilizzati come materie prime secondarie.

I tre casi studio sono stati oggetto di tre progetti pubblici di ricerca volti a valutare l'applicabilità di diverse strategie per produrre materie prime secondarie e gli impatti ambientali del loro riciclo.

Il primo caso di studio si è concentrato sul riciclaggio di materiali contenenti amianto e fibre vitree artificiali. In particolare sono state valutate le emissioni in atmosfera legate al processo di trattamento termico ed è stato valutato il conseguente impatto ambientale mediante il metodo LCA.

Il secondo caso di studio ha valutato la fattibilità del riciclaggio della plastica dura per uso domestico. È stata dimostrata la possibilità di separare le diverse plastiche dure in base alla loro densità, ed è stato possibile ottenere una miscela con buone proprietà meccaniche composta da plastiche post-consumo e post-industriali. L'analisi LCA ha mostrato una notevole riduzione degli impatti ambientali rispetto alla produzione di nuove plastiche vergini.

Il terzo caso di studio ha analizzato la possibilità di riutilizzare la cellulosa da Prodotti Sanitari Assorbenti (PSA) ottenuta dal processo di riciclo del brevetto dell'azienda Fater S.P.A.. I test effettuati durante il progetto hanno permesso di valutare l'utilizzo di cellulosa riciclata sia per la produzione di pannelli isolanti che come scaglie di cellulosa. È stato dimostrato che i fiocchi di cellulosa riciclata hanno le stesse proprietà della cellulosa vergine per quanto riguarda l'isolamento termico degli edifici e che l'applicazione di pannelli isolanti è una valida modalità di utilizzo a basso impatto ambientale.

1 Introduction

Waste production and its disposal are one of the most important environmental concerns of modern society. Consequently, it is important to develop new solutions that allow waste to be recycled and reused as secondary raw material.

The average European citizen generates around 5 t of waste, of which only a limited share (39% for 2014 with a total EU waste production of 2,6 billion t) is recycled. Much of the rest still ends up in landfills or incinerators (European Commission, 2021b).

EU waste policy provides a framework for improving waste management, stimulating innovation in waste separation and recycling, limiting the use of landfill and creating incentives to change consumers' behaviour. It also aims to reduce the actual amount of waste generated and the number of harmful substances it contains.

In order to protect environment and human health, the European Commission has issued the "Waste Framework Directive" that sets the basic concepts and definitions related to waste management, including definitions of waste, recycling and recovery.

To comply with the objectives of this Directive, EU countries shall take the necessary measures to achieve the following targets:

- by 2020, the preparation for reuse and the recycling of waste materials (such as paper, metal, plastic and glass) from households shall be increased to a minimum of overall 50 % by weight;
- by 2020, the preparation for reuse, recycling and other material recovery, including backfilling operations using waste to substitute other materials, of non-hazardous construction and demolition waste shall be increased to a minimum of 70 % by weight;
- by 2025, the preparation for reuse and the recycling of municipal waste shall be increased to a minimum of 55 %, 60% and 65% by weight by 2025, 2030 and 2035 respectively.

In addition to defining this directive, the Commission has adopted new Circular Economy Action Plan (CEAP), in March 2020 a package of measures and legislative proposals to boost sustainable growth and help Europe make the transition towards a more circular economy, in order to reduce the pressure on natural resources and create sustainable growth and jobs. Moreover, the ultimate goal is to achieve the EU's 2050 climate neutrality target and to halt biodiversity loss.

The plan consists of several interrelated initiatives designed to develop a strategic framework in which sustainable products, services and business models will be the driving force for adopting more sustainable consumption patterns.

In addition to the design of sustainable products, a focal aspect of the action plan addresses the main value chains, which are required to make an effort to the sustainability challenge of the main value chains urgently requires comprehensive and coordinated actions that will be an integral part of the strategic framework on sustainable products. Among these value chains, plastics and building construction/demolition must be addressed.

A further relevant aspect highlighted by the European policy is aimed at improving circularity in a toxic-free environment by replacing hazardous substances, supporting the development of solutions for high quality sorting and removal of contaminants from waste, developing methodologies to minimise the presence of substances of health or environmental concern in recycled materials and the products obtained from them.

About plastic waste, almost 26 million t of plastic waste is generated in Europe every year. Thus, the EU is taking into account different strategy to accelerate the transition to circular economy. Specific targets and rules were identified in the EU plastic strategy, part of the EU's circular economy action plan. Different actions were defined, including:

- Increase the demand for recycled plastic content;
- improving the separation and collection methods of plastic waste;
- scaling up support for innovation, with an additional €100 million to develop smarter and more recyclable plastics materials, to make recycling processes more efficient, and to trace and remove hazardous substances and contaminants from recycled plastics.

Construction and demolition waste (CDW) accounts for more than one third of all the waste generated in the EU. It contains a wide variety of materials such as concrete, bricks, wood, glass, metals and plastics. Alongside these materials for disposal, there are also materials containing asbestos, such as "Construction materials containing asbestos", which must be managed separately from other waste because of their hazardousness. The separate collection of this hazardous waste from Construction and Demolition waste, facilitate the recycling process of the material, because after collection it becomes very selected.

The Waste Framework Directive set the construction and demolition waste as priority waste. It sets the following objectives by the 2020 follow these objectives:

- the preparation for reuse, recycling and other material recovery of non-hazardous construction and demolition waste (excluding naturally occurring material defined in category 17 05 04 in the list of waste) shall be increased to a minimum of 70 % by weight;
- promote selective demolition to enable removal and safe handling of hazardous substances and facilitate reuse and high-quality recycling by selective removal of materials and establishing sorting systems;
- reduce waste generation.

Given these European and member state targets, it is relevant to evaluate solutions and processes for the recycling and reuse of waste that is currently not or cannot be included in supply chains. A greener economy means sustainable growth and job opportunities. As well as generating an environmental benefit, EU businesses could make net savings of up to 600 billion euro by applying the principles of eco-design, waste prevention and reuse of raw materials. This could generate 2 million additional jobs and a 1% grow in GDP by increasing resource productivity by 30% by 2030 (European Commission, 2021a)

The residues or waste produced do not necessarily remain categorised in this way: some never become waste (by-products), others lose their waste status as a result of a recovery process (residues become secondary raw material or "end of waste"). It all depends on the legal framework and its evolution.

In order to no longer be considered waste, it is necessary to supplement EU directives, transposed, for example, by the Italian state with a special decree drawn up by the Ministry of Ecological Transition, which clearly defines the criteria for ceasing to be considered waste, the so-called "end of waste".

The concept of "end of waste" refers to a recovery process carried out on a waste, at the end of which it loses this status and acquires that of product. The term "end of waste" therefore does not refer to the final result, but to the process that actually allows a waste to play a useful and a profiting role as a product again.

The European legislation on waste contained in Directive 2006/12/EC, which the EU Parliament and Council followed up by adopting Directive 2008/98/EC of 19 November 2008, is known as the Waste Framework Directive (WDF). The latter directive places waste prevention at the top of the priority scale of waste management methods, followed immediately by preparation for reuse:

Article 4 of Directive 2008/98 - Waste hierarchy:

"1. *The following waste hierarchy shall apply as the order of priority of legislation and policy on waste prevention and management*:

1. prevention;
2. preparation for reuse;
3. recycling;
4. other recovery, for example energy recovery; and
5. disposal [...]'.

In order to art no. 6 of the WFD, a waste ceases to be a waste when it has undergone a recovery operation and fulfils the precise conditions, which are as follows:

(a) *it is commonly used for specific purposes:* it must be widespread, generally applied in known areas and capable of performing known and defined functions;

(b*) there is a market or demand for the substance or object*: the fact that there is a market shows that the recovered object is unlikely to be abandoned;

(c) *the substance or object fulfils the technical requirements for the specific purposes and complies with existing legislation and standards applicable to products*: i.e. the object must be able to provide the required performance under the concrete conditions of use or consumption, in accordance with both the legal and the technical standards for the specific good;

(d) *the use of the substance or object will not lead to overall negative impacts on the environment or human health.*

Condition (b) is also commented by the EU Commission in its Communication to the Council and the European Parliament on waste and by-products 2007 (European Commission, 2007) , which clarifies that " *If it subsequently turns out that the waste can in fact serve a useful purpose, then the material will lose its waste status when it is ready for use as a recovered product* [...] *the existence of long term contracts between the material holder and its subsequent users can be an indication that the material covered by the contract will be used and therefore that certainty of use is present [....] Where a manufacturer can sell the material concerned for a profit, this can indicate that it is more likely that such a material will certainly be used. However, this alone is not definitive*", i.e. a possible profit is not in itself an indication of reuse. In fact, it is not the economic value of the material that is decisive (as required by the previous Article 181-bis of Legislative Decree 152/2006 with respect to secondary raw materials) but rather the fact that the usefulness of the material can be demonstrated in the same way as any other product. To this aim, it is required that long-term contracts must be signed to demonstrate that the material will be used, ruling out the option of "wild" or illegal disposal.

Once all the conditions set out in Article 6 are met, the object resulting from the recovery process is no longer waste since it has objectively become a product.

With reference to the concept of recovery, the directive explicitly opens up the possibility to consider the monitoring waste to ascertain whether it satisfies the criteria for defining when waste ceases to be waste as a recovery operation.

Thus, it is necessary to introduce the waste into a recovery path so that it can be considered as end-of-waste. It is necessary to implement a transformation operation that gives the waste a useful role, replacing other materials that would otherwise be used to fulfil a particular function within the process or in the economy in general.

Based on these concepts, the research work focused on waste that does not currently have a destination and therefore constitutes unsorted waste. Today, there are many wastes that have been reintroduced as secondary raw materials into the same or other supply chains: the European Commission has set targets for specific waste streams, such as electronic equipment, cars, batteries, construction, demolition, municipal and packaging waste, as well as for reducing the landfilling of biodegradable waste. However, there are still many other wastes that are not recycled but have great potential for reuse even with less environmental impact than using virgin raw materials.

The aim of the research was to assess the environmental impacts of three case studies involving three categories of waste that are currently destined to landfill disposal. The advantages/disadvantages of using them as secondary raw materials is described in the following paragraphs:

- Materials containing fibres, both man-made vitreous fibers (MMVF) and asbestos fibers;

- Hard polyolefin based domestic plastic waste;

- Absorbent Health Products.

The first case study has focused on the treatment for reuse of two building materials that were widely used in the past and today: Asbestos Containing Materials (ACM) and Man Made Vitreous Fibres (MMVF). In the countries all over the world where asbestos is banned, it has been progressively removed from the environment and in many cases substituted with synthetic fibres.

The terms “Asbestos” includes a family of fibrous silicate minerals that currently used in several countries or used in the past for their extraordinary proprieties, such as the great tensile strength, poor heat conduction, non-biodegradability, high electrical and alkali/acid attack resistance and sound absorption capabilities (Leonelli et al., 2006). Unfortunately, it was discovered that asbestos fibres are toxic/pathogenic and the six asbestos mineral species have been classified by the International Agency for Research on Cancer as carcinogen for humans (International Agency of Research on Cancer (IARC), 1987).

There are several fibrous minerals in nature, but the main ones, classified by Italian law as 'asbestos' are: chrysotile (serpentine asbestos) and the amphibole asbestos species (actinolite asbestos, amosite, anthophyllite asbestos, crocidolite and tremolite asbestos). The article 247 of Italian Legislative Decree n. 81 of 09 April 2008 designates these fibrous/asbestiform silicates as “asbestos”, as they are the most commonly used in the commercial sector, and therefore considered a risk for exposed workers.

The serpentine asbestos, also named white asbestos, have an ideal chemical formula $Mg_3(OH)_4Si_2O_5$. This mineral, structured with layers of silicates (with a tetrahedral and an octahedral sheet), is the most often commercially used form of asbestos and cover more than 90 % of world production of asbestos (Yanagisawa et al., 2009). The amphiboles asbestos are double-chain silicates with chain structure and assume a fibrous habit being structurally elongated in one preferred crystal direction (Hawthorne & Oberti, 2018). The mineralogical composition, structure and physical properties determined the use of these minerals for industrial applications. Each type of asbestos has different physical characteristics. Chrysotile fibres are extremely thin and are flexible and soft, which can provide weaving, in turn, amphibole asbestos fibres are harsher and more brittle than chrysotile (Kusiorowski et al., 2012). The chemical and physical properties of asbestos are shown in Table 1.

Table 1 Physical and chemical properties of asbestos (Klein et al., 2002)

Name	**Ideal formula**	**Colour**	**Luster**	**Density (gcm^{-3})**	**Hardness (Mohs scale)**	**Decomposition Temperature (°C)**	**Fusion temperature of residual material (°C)**
chrysotile	$Mg_3(OH)_4(Si_2O_5)$	White to grayish green	Silky	2.4 – 2.6	2.5 – 4	450-700	1500
crocidolite	$Na_2(Fe^{2+}{}_3Fe^{3+}{}_2)Si_8O_{22}(OH)_2$	Blue to dark	Silky to dull	3.2 – 3.3	4	400-600	1200
amosite	$Fe^{2+}{}_2Fe^{2+}{}_5Si_8O_{22}(OH)_2$	Yellowish gray to dark brown	Vitreous to pearly	3.1 – 3.2	5.5 – 6	600-800	1400
anthopyllite asbestos	$Mg_2Mg_5Si_8O_{22}(OH)_2$	Gray to green	Vitreous, pearly on cleavage	2.85 – 3.2	5.5 – 6	800-850	1450
tremolite asbestos	$Ca_2(Mg_{5.04.5}Fe^{2+}{}_{0.0-0.5})Si_8O_{22}(OH)_2$	White, gray.	Vitreous and silky	2.99 – 3.03	5 – 6	950-1040	1315
actinolite asbestos	$Ca_2(Mg_{4.5-2.5}Fe^{2+}{}_{0.5-2.5})$ $Si_8O_{22}(OH)_2$	Pale to dark green	Vitreous to dull	3	5 – 6	620-960	1400

Thanks to its properties, asbestos minerals have been used for a long time, until the carcinogenic properties of asbestos were confirmed by experimental evidence. As a matter of fact, as early as 1973 the IARC assessed the carcinogenic effects of inhaling asbestos fibres, particularly on workers and among families of asbestos workers. Thus, in 2012 IARC classified the six above-mentioned minerals as cancerogenic belonging to group 1 (Carcinogenic to humans).

In particular, professional or environmental exposure to asbestos may cause mesothelioma, a fatal malignancy of the cells lining the pleura, peritoneum and pericardium. Given a long latency period (20-40 years), a reduction in the frequency of mesothelioma only occurs in those countries that have implemented policies to control and abandon asbestos since the 1970s. Worldwide there are still more than 90.000 people dying every year due to asbestos-related diseases including mesothelioma and asbestosis resulting from occupational exposure. Moreover, it is estimated that several thousands of deaths can be attributed to other asbestos-related diseases as well to non-occupational exposure to asbestos (Public Health and the Environment Word Health Organization, 2006).

For example, in Italy, Law No 257 of 27 March 1992 - Regulations on the cessation of the use of asbestos - banned the extraction, import, processing, use, marketing, treatment and disposal of asbestos in the national territory, as well as the export of asbestos, which caused tens of thousands of deaths, mainly in the areas where these materials were produced. In spite of the many deaths, there are still many countries which continue to produce and use materials containing asbestos. Therefore, is a global issue. In particular, of the 195 world countries, 65 (33%) countries had asbestos bans and 130 (67%) countries did not (Arachi et al., 2021). Some relevant countries such as China, India, and Russia have not banned the use of chrysotile yet.

According to current Italian legislation, ACW must be removed and properly managed in accordance with safety regulations. Even if European regulation considers the recycling of ACW after inertisation, disposal in landfill after inertisation treatments has been taken as the only possible solution.

The European Parliament resolution 2012/2065 (INI) of 14 March 2013 (“asbestos related occupational health threats and prospects for abolishing all existing asbestos”) (European Parliament, 2006) consider that: "delivering asbestos waste to landfills would not appear to be the safest way of definitively eliminating the release of asbestos fibres into the environment (particularly into air and groundwater) and therefore it would be far preferable to opt for asbestos inertisation plants " and that "creating landfills for asbestos waste is only a temporary solution to the problem, which in this way is left to be dealt with by future generations, as asbestos fibres are virtually indestructible over time".

Several techniques have been developed worldwide for the inertisation of asbestos that modify completely the the crystal-chemical structure of asbestos thereby undermining the hazardous properties. From to the treatment is possible obtain a product can be considered as new raw material and that therefore can be destined for recycling in other supply chains such as cement industry, ceramic industry and others one (Gualtieri et al., 2011b; Gualtieri & Tartaglia, 2000; Ligabue et al., 2020).

All the processes for the inertisation can be classified into the following three macro-categories: thermal, chemical and mechano-chemical treatments. Thermal treatments are use heat to transform asbestos into an inert material, using a temperature range in which fibres are not stable anymore; chemical treatments use chemical reactions to convert asbestos into harmless compounds; in mechano-chemical treatments, fibres are degraded by mechanical milling.

Thermal treatments are very articulated and it is also the only solution where the most important industrial experiences are concentrated. Almost all of the applications identified and the analysed studies foresee the use of temperatures up to or above 1200 °C. However, it should be pointed out that a recent study has demonstrated that a temperature of 700 °C is sufficient to make chrysotile-rich (not amphibole asbestos-rich) asbestos-cement inert (Kusiorowski et al., 2015).

Asbestos minerals thermal decomposition consists of three stages: the first is associated with the loss of adsorbed water, the second is the removal of structural hydroxyl groups from the structure of asbestos minerals and the third is responsible for the crystallization of amorphous materials and the growth the new phases occurs after dehydroxylation. The main advantages connected to thermal treatment are: (i) incorporation of large amounts of heavy metal ions which are chemically bonded inside an inorganic amorphous network; (ii) the final process product is inert towards most chemical or biological agents and can be disposed in landfills; (iii) flexibility to treat wastes of various type; (iv) consolidated technology; (v) a reduced amount of waste is obtained.

The common critical issue for all thermal treatments is the high energy required to heat a thermally inert material such as asbestos. In this regard, it should be recalled that the most common type of ACW (Asbestos Containg Waste) is asbestos-cement, where asbestos fibres are dispersed in a thermally inert material as well.

Other issues are related to the formation of atmospheric pollutants during the heating phases. In this view, a particular problematic material is vinyl-asbestos, since the thermal treatment of halogenated polymers can lead to the formation of persistent organic pollutants such as dioxins and polychlorinated biphenyls.

There are several subcategories of thermal treatment, which can be classified according to the technology used to heat, the presence of additives and the heat-up processes (recrystallisation). Depending on the subcategory chosen, the obtained material may be an inert glass mass to be conferred to the landfill as non-hazardous waste, a pottery or a potentially re-usable material with good mechanical properties, for example in the building industry or civil works.

The research carried out focused on a consolidated methodology developed in the European Patent EP2027943B1 named “KRY·AS”, that consists of thermal conversion by ceramisation of the ACW using a continuous furnace (Gualtieri et al., 2011c; Gualtieri & Boccaletti, 2011; Ligabue et al., 2020), developed in 2007 by University of Modena and Reggio Emilia in collaboration with the company ZETADI S.r.l.. The experimental conditions for this process are well known (Bloise et al., 2016; Gualtieri et al., 2008), as well as the thermo-chemical reactions involved (Gualtieri, et al., 2008; Viani et al., 2013).

Basically, the process involves three main steps, i.e. dehydration, dehydroxylation with consequent destruction of the structure of the asbestos fibres, and recrystallization. The obtained solid residue can be used for several industrial applications, including ceramics, clay bricks, concrete etc, as indicated above.

Treatment does not include grinding prior to firing and big bags containing asbestos-cement plates remain sealed in such a way as to avoid the release of fibres. The cooking cycle uses a "tunnel" continuous industrial gas oven, generally used for brick production, of a length not less than 50 m. The process (combustion of big bags, dehydration, decarbonation and dehydroxylation) involves heating at a maximum temperature of 1200-1300 °C for not less than 8 h. Subsequently, the material is conveyed in the firing zone at the same temperature for about 12 h and finally cooled to room temperature for about 6 h in the final section of the oven.

The second material, Man-Made Vitreous Fibres are currently the most commercially important group of all man-made inorganic fibres and were already widely used in the 1930s for thermal and acoustic insulation, and later as reinforcement for plastics, due to their chemical and physical properties, in the textile industry and other industrial activities.

These materials include glass fibres (used in glass wool and continuous glass filament), rock or stone wool, slag wool and refractory ceramic fibres (Figure 1). All main MMVFs are composed by silica-based and contain various amounts of other inorganic oxides.

Respirability and biopersistence in the lung of fibres have been monitored over the years to reduce the risk, thus the industry has introduced some new, less biopersistent fibre compositions than the older types. This not addresses the problem of disposing of old types of MMVFs.

The International Agency of Research on Cancer (IARC) (2002) classifies special-purpose glass fibres such as E-glass and ‘475’ glass fibres and Refractory ceramic fibres are possibly carcinogenic to humans (Group 2B). Insulation glass wool, continuous glass filament, rock (stone) wool and slag wool are not classifiable as to their carcinogenicity to humans (Group 3). It should be remarked that for new generation fibres, IARC has not given an assessment due to insufficient data in the literature, although these fibres appear to have low carcinogenicity.

While for Classification, Packaging and Labelling (CPL) Regulation (European Parliament and the Council of the European Union, 2008), the fibres are exempt from classification as carcinogenic if they have a length-weighted geometric mean diameter minus two standard errors (DLG-2ES) greater than 6 μm (Note R). In addition, the classification as a carcinogen does not apply to those fibres for which it can be demonstrated that they have low biopersistence (Note Q). So, for CPL regulation this type of fibres may fall into the categories: Carc. 1B - may cause cancer by inhalation and Carc. 2 - suspected for causing cancer.

Possible waste classifications for MMVF are as follows:

- 17.06.03* (special hazardous waste);

- 17.06.04 (non-hazardous special waste).

Once again, it is the chemical and physical characteristics of MMVF (content of alkaline and alkaline/earth oxides and geometric mean diameter weighted on fibre length, DLG-2ES) that determine its classification.

In particular, for the attribution of the EEC code, waste consisting of MMVF must be analysed prior to disposal.

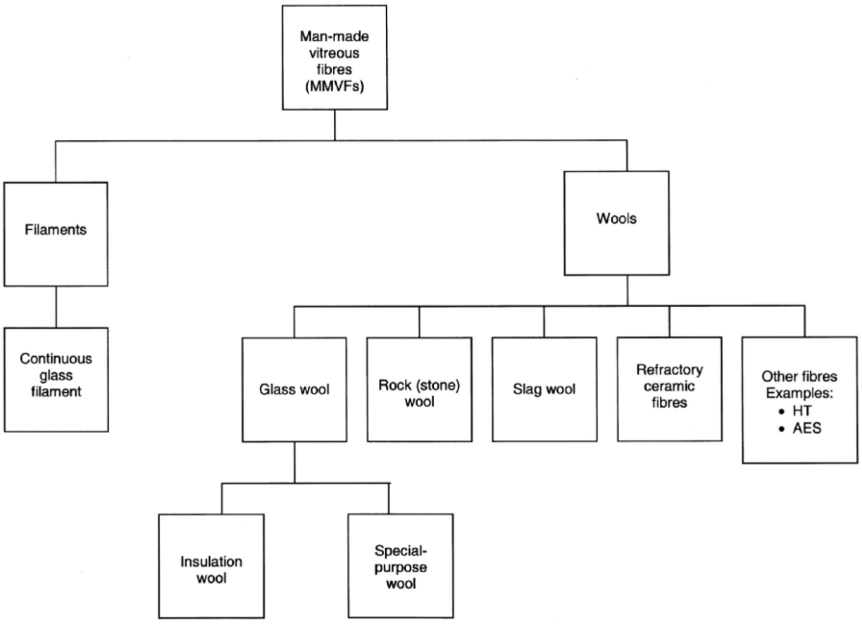


Figure 1 Classification of Man-made vitreous fibres (MMVFs)

If these materials are classified as MMVF, they can undergo the same thermal treatment process as performed on asbestos to produce glass that can be successively recycled as a secondary raw material (Gualtieri et al., 2009), in the 2021 have already carried out tests to verify that the material produced was safe according to the regulations and the possibility of recycling these products into stoneware tile mixtures, for the production of mineral wool from secondary raw material and as filler for the plastic production.

The second case study is focussed on plastics, in particular the household hard plastic that which cannot be recycled today. In the last decades this product has surged to a position of near prominence as a crafting material not only for common household goods, but also for its contribution to overcoming recent and constantly emerging environmental challenges. Global production since the 1960s has increased twenty-fold, up to 368 million t in 2019 (Plastics Europe, 2020). Plastics, for example may be employed as insulation material to improve energy efficiency for heating and cooling, and as lighter materials in mobility applications, such as planes or cars, to save fuel and reduce polluting emissions, or, more recently, for 3D printing (Goh & Yeong, 2018). Nonetheless, the production, use and discarding of plastic is more often than not wasteful and harmful for the environment, lacking a circular approach that could reduce some of these impacts. Plastics, and in particular polyolefins, are a low-cost, hydrophobic, possibly transparent and bio-inert alternative to other, classic materials.

Each year, more than 29 million t of plastic waste are produced in Europe, with the largest share being made of thermoplastic polyolefins polypropylene (PP) and polyethylene (PE) (Plastics Europe, 2020). PP, representing 19% of plastics in Europe, while less dense, is generally harder and more thermally and chemically resistant than PE, which comprises nearly 30% of plastics in Europe, summing both low-density (LD) and high-density (HD) PE. PP is commonly employed for food wraps, automotive parts or pipes, while

LD-PE is used for reusable bags, containers or food wrapping films, and HD-PE can be found in bottles, toys or pipes.

Of all produced and wasted plastic in Europe, nearly 25% ends up in a landfill (Plastics Europe, 2020). Landfilled plastic is, on one hand, a missed opportunity for either energy recovery or recycling, and on the other hand, a direct, harmful pressure on the environment, for example, as marine litter entangling, or ingested by, marine organisms (Law, 2017), as microplastics produced by degradation of larger waste plastic and transported through the atmosphere even to remote locales, with known harmful effects on human health (Prata, 2018), as a source of persistent organic pollutants such as polychlorinated biphenyls (Melnyk et al., 2015); or a cause behind the loss of potentially productive land (Ilyas et al., 2018). Energy recovery through incineration directly leads to the emission of greenhouse gases and to pollutants harmful to human health, such as formaldehyde or dioxins, where the off-gas is uncontrolled and released without thorough purification, making it an environmentally suboptimal choice (Gradus et al., 2017; Nagy & Kuti, 2016). Moreover, the production of plastics is very energy intensive, with resins incorporating 62–108 MJ/kg of energy, much higher than required for paper, wood, glass or most metals, and about 4% of fossil fuels is yearly employed for the production of plastics, with another 3 - 4% being used in their manufacture to supply energy (Ilyas et al., 2018; Lebreton & Andrady, 2019).

To date, plastic waste has not been specifically covered in EU legislation. Only Packaging Directive 94/62/EC has set a specific recycling target for plastic packaging. In contrast, the Waste Framework Directive (2008/98/EC) set a general recycling target for household waste, which also applies to plastic waste, and established a waste hierarchy (prevention, preparation for reuse, recycling, recovery, and disposal). The latest EU Multiannual Financial Framework (MFF) set a so called “plastic tax” of 0.80 EUR/kg for non-recycled plastic packaging as of January 1st, 2021. Nonetheless, while hard plastic recycling is not obligatory and not registered, several European companies already process hard plastic waste (HPW), and recycling is already common in multiple European countries.

Consequently, considering the costs and drawbacks of producing, landfilling or incinerating plastic, there is a growing, urgent need to increase the effort toward plastic recycling. Two important causes that explain why only about a fourth of all waste plastic is actually recycled are that recycled plastic may not be used for the same application for health and environmental protection reasons, and that post-consumer plastic generally consists of blends of polymers that are difficult to separate (Bajracharya et al., 2016).

Plastic recycling begins at collection, either via source separation by private citizens or via mechanical recovery from mixed municipal solid waste. In both cases, techniques have been developed for the separation of the mixed plastic streams by exploiting differences in chemical and physical properties of plastics, such as gravimetric, optic or electrostatic (Turku et al., 2017), while technology is still struggling regarding multi-material waste (Ashton et al., 2016). PE and PP are usually separated together from the plastic waste stream as gravimetric methods are easy and cost-effective; these polyolefins generally comprise the lighter fraction of the waste stream, as opposed to polymers with density higher than water (Turku et al., 2017). The properties of recycled plastic are generally less desirable than those of virgin plastic (Gu et al., 2016) because of physical and chemical changes to the molecular structure of plastic polymers, or because of common contaminants such as alcohols, esters and ketones, which are rarely present in virgin plastic (Luijsterburg & Goossens, 2014). A common method of enhancing the inferior characteristics (such as rheological and mechanical properties) of recycled plastic is blending polymers with fillers, such as talc or glass fibres (Gu et al., 2016). Recycling plastic is a sensible choice when the environmental benefits outstrip the potential economic costs. The largest environmental benefits result from a reduction in fossil hydrocarbons needed in the manufacture of virgin plastic and the consequent avoided negative greenhouse gas emissions, particulate matter emissions and acidification potential (Schwarz et al., 2021). The current recycling rate of 9% for plastics (Geyer et al., 2017) needs to be increased for any benefit to be felt and for plastic recycling to significantly impact climate change mitigation. One way to evaluate the environmental benefits of alternative scenarios and technology is to perform a life cycle assessment (LCA) analysis. Several literature reviews and case studies exist (Arena et al., 2003; Lazarevic et al., 2010; Rigamonti et al., 2014; Schwarz et al., 2021), offering a strong framework and basis for further and more specific applications of the LCA methodology, although most previous studies focus on plastic wraps and containers instead of hard plastics. In general, no single recycling technology is universally superior to other choices, requiring a thorough evaluation for each specific real-life scenario to identify the most effective solution, whether it may be reuse or energy recovery. Being an energy and cost intensive process, plastic waste recycling is generally supported by national systems based on different taxation frameworks and/or incentives. However, these systems are often limited to specific kinds of plastic waste, namely, containers such as bottles, boxes, blisters etc. Hard plastic products that are not intended to be used as containers are generally excluded from these recycling chains: for instance, this is the case for chairs, basins, crates, toys etc. As a consequence, today these materials are generally not recycled and are instead sent to incinerators or landfills, despite generally being made of PE and PP similarly to container plastic waste (Faraca et al., 2019). As described above, new policies strongly support a further increase in the share of recycled plastic, and consequently it is expected that hard plastic waste management will significantly and rapidly change in this upcoming decade.

Moreover, the quality of recycled plastic should be considered when comparing recycling scenarios with virgin plastic production. During production, plastic polymers are blended together with dyes, plasticizers, fillers and inclusions. As such, recovered materials are not pure and usually are characterized by lower quality and, consequently, need to be down-cycled to be used again (Arena et al., 2003; Schwarz et al., 2021), which can be seen as a secondary recycling technique (Singh et al., 2017).

This translates directly to open-loop strategies, where recycled plastic derived from specific items or applications cannot substitute virgin plastic needed for the same purpose because of its decreased quality and properties. Optimal strategies to overcome this obstacle would be upstream policy changes, ensuring the production of less contaminated plastics, or downstream technological developments, pushing toward the recovery of higher quality recycled polymers. Thus, there is an urgent need to understand the properties of plastic blends prepared from hard plastic waste. Unfortunately, most of the previous literature has been focused on plastic blends made on container plastic waste, generally wasted bottles. Recycled plastic packages have to contend with physical degradation, polymeric or molecular contamination (Vilaplana & Karlsson, 2008) and hygroscopicity due to polar contaminants originating from prints, glues, etc. (Garofalo et al., 2021). Contaminants such as VOCs are generally the result of insufficient cleaning during recycling, of degradation processes or of additives in the polymeric matrix (Cabanes et al., 2020; Chen et al., 2020; Strangl et al., 2021). HPW, on the other hand, must contend with a different set of obstacles: for example, due to the long life of most hard plastic consumer products compared to that of plastic packaging, degradation processes are arguably more significant for the development of secondary contaminants, together with additives and residues from additives. At the same time, hard plastic items contain fewer components that are made from different polymers, so polymeric contamination is more likely to originate from sorting mistakes, if present, than from components made from different polymers. Hard plastic items are also devoid of inks, glues or other hygroscopic contaminants, and are less likely to be employed as containers, reducing the impact of product residues as molecular contaminants. Finally, hard plastic is prominently produced via injection molding, while plastic packages are produced with a mixture of injection and blow molding technologies, resulting in different MFI profiles.

The blending of virgin and recycled plastic has also been studied for thermoplastics employed in 3D printing (Pakkanen et al., 2017) and for plastic packages. For instance, a recent study (Curtzwiler et al., 2019) demonstrated the usefulness of blending virgin polypropylene with recycled mixed polyolefins from beverage bottle enclosures, highlighting how mixing defined quantities of post-consumer polyolefins with virgin plastic could lead to better results for desired properties. For example, 80% of post-consumer polyolefins mixed with 20% of virgin polypropylene directly produced a 14% increase in the tensile modulus compared to pure virgin polypropylene. Curtzwiler et al. (2019) report on a series of analyses, such as thermogravimetric analysis, melt flow index testing and electromechanical testing, to define the contribution of post-consumer polyolefin to the physical and chemical properties of blends, concluding that in most cases the law of mixing may be used to predict the blends' properties as relations are generally linear.

However, to the best of our knowledge, the effects of post-consumer hard plastic on the properties of plastic blends have not been investigated yet. This is particularly critical because hard plastic materials have a completely different composition in terms of additives, plasticizers, etc.

The last case study taken into account is focussed om the Absorbent Hygiene Products (AHP), products that are widely used daily by millions of people including baby nappies, sanitary protection pads, tampons, adult incontinence products and personal care wipes.

AHP are usually collected together with the undifferentiated residual fraction of municipal solid waste (MSW) and then disposed of in landfills or by incineration, only in some parts of Europe limited recycling processes are implemented. This waste category is 2-3% of the total municipal solid waste and represent up to 15-25% of the residual waste stream in some treatment facilities, in sites where a separate collection rate of more than 70% is achieved (Dri M. et al., 2018)Thus, this does not allow an easy quantification of the production of this category of waste.

They constitute a considerable fraction in quantity and weight: used AHP can have up to three times the weight of unused AHP (Arena et al., 2016).

A recent estimate from the JRC preliminary report (Revision of EU Ecolabel criteria for Absorbent Hygiene Products)(Pérez Arribas et al., 2021) indicates that the total AHP sales volume in Europe was valued at just over EUR 6 bn in 2020 representing a volume of around 59 bn units of these products In the EU 27 and the UK. The main market, around of 35% corresponds to baby diapers, to follow panty liners (30%), Feminine care pads (less than 25%) tampons (10%).

The production of this waste is of environmental relevance, so many companies are studying different approaches to reduce the waste generated in the production processes and find innovative solutions to recycle and reuse AHP waste both as material and for energy purposes.

Modern absorbent sanitary products are built up in layers to allow urine and faeces to be transferred and distributed to the absorbent material. The elementary layers consist of an outer shell made of polyethylene film or non-woven composite that prevents the escape of moisture and inner absorbent layers based on cellulose pulp and absorbent polymers for moisture retention and a final layer of non-woven material in contact with the skin.

According to data compiled by Ambiente Italia (Ambiente Italia s.r.l., 2013), the weighted average composition of AHPs is shown in Table 2.

The cellulose fibre is natural insulation materials with a similar characteristic to synthetic materials and a low environment impact, low embodied energy, and good thermal and acoustic properties. Thanks to these properties and the development of a more sustainable economy this type of insulation material is becoming more and more popular (Lopez Hurtado et al., 2016a).

Virgin cellulose is made from recycled paper fibres such as unsold newspapers or recovered from waste collection. They are then milled and treated with inorganic additives such as flame retardants and mould growth inhibitors. Further advantageous technical characteristics of cellulose are related to its high coefficient of thermal expansion and high surface area, and the associated environmental aspect plays an important role. Cellulose has a high durability and is recyclable and environmentally friendly (Pal et al., 2021).

Two products can be made from cellulose: a prefabricated panel, obtained by compression and moulding with a binder, or, more commonly, loose fibres in flakes to be blown into cavities, ceilings etc.

Table 2 Composition of AHP used and unused

	AHP unused	**AHP used**
Cellulose	48.0%	23.8%
Super Absorbent polymer (polyacrylate)	23.0%	11.6%
Polypropylene	13.0%	6.6%
Polyethylene	8.0%	3.9%
Other (PE/PP/PET/Lycra)	8.0%	4.0%
urine and faeces	0.0%	50.0%

Currently, some private companies are developing emerging recycling technologies that focus on separating the main components of absorbent hygiene products (AHP), such as nappies, to recover materials for various uses. Generally, the techniques available today for the recovery and separation of AHP are based on a first step of sterilisation of the input stream using steam at high temperature and pressure, to sanitise the material and eliminate all pathogens. Afterwards, the material is dismembered by blades and the plastic fractions separated from the cellulosic and other components. Other solutions include using nappies as raw materials to produce fuels. Other research attempts have followed a different approach: degradation of nappies by the Pleurotus ostreatus fungus with the use of urban garden waste as co-substrates, Composting and anaerobic digestion of the biodegradable fraction with activated sludge waste (Tsigkou et al., 2020).

An Italian patent developed by Fater S.pA. enterprise (Patent number JP2013111480 (A)) enables the recovery of plastic granules and high quality organic-cellulosic material. In particular, 75 kg of plastic and 225 kg of organic-cellulosic material are produced from 1 t of used products. Fater S.pA. has built an industrial plant in Lovadina di Spresiano (Treviso), capable of recycling 100% and processing 8,000 t/a of AHP waste (Arena et al., 2016).

The new disposal scenario for this waste fraction opens up new possibilities for the management of this waste stream and is particularly important if we consider that its separate collection has already been introduced in more than 800 Italian municipalities (about 12 million inhabitants).

Italian legislation has also moved in this direction: the Official Gazette of 2 November 2019 published Law 128 of 2 November 2019, the law converting Decree Law No. 101 of 3 September 2019, "containing urgent provisions for the protection of employment and for the resolution of company crises", which, in Article 14 bis, contains the reform of the "end of waste status", commonly referred to as "End of Waste" (EoW).

Italy is the first country in the world to introduce a regulation "for the recovery of secondary raw materials" from used AHP. The decree is an important milestone for the environment, which finally sees the above-mentioned product fraction no longer as waste, but as a resource to be recovered.

This opens up an important technological and regulatory pathway, in which AHP can be collected separately and recycled.

In chapters 2, 3 and 4 respectively, the methodologies and results of each case study are discussed.

2 Case study 1: Treatment of Asbestos Containing Materials (ACW) and of Man Made Vitreous Fibres (MMVFs) wastes

2.1 Case study description

This case study aims at improving the knowledge on the environmental sustainability of asbestos inertisation and treatment of rock wool, considering that emissions are a crucial aspect for the environmental assessment of the use of thermal processes.

During the research period, several experiments have been conducted on two different prototypes to monitoring and evaluate the emission deriving for thermal processes that could be realistically applied on an industrial scale in the near future. The first one is a laboratory prototype (a), and second one a pre-industrial prototype (b) (Figure 2). Three different kinds of waste were tested in collaboration with industrial partner – Zetadì S.r.l., in order to monitor the impact of waste composition on the emissions. The two research bodies, the CNR-IIA and the University of Modena and Reggio Emilia, have worked together to carry out these two monitoring campaigns.

In the first case, a sampling strategy was developed on a laboratory prototype. In the second case, measurements necessary for the environmental assessment procedures of the industrial plant to be built by Zetadì S.r.l. were carried out, so it was necessary to comply with national and international regulations in order to carry out the measurements and analyses.

The measurement on the laboratory prototype were carried out thanks to the project financed by Ministry of Ecological Transition under the Agreement RINDEC-20016-4 of November 25th 2015, entitled “Development of a novel methodology for asbestos inertisation, emission abatement and analysis of the supply chain (www.filieraamianto.it).

Thanks to the prototype of a pre-industrial oven for the thermal processing of mineral wool of Zetadi s.r.l. in Ferno (VA), it was possible to investigate the emission products caused by the vitrification process.

In the laboratory prototype, two different kinds of ACW were used: asbestos cement sheets, and an asbestos waste including a plastic polymer (polyurethane). These two kinds of waste were chosen because they are the most common types of ACW, so that results obtained can be generalised to most of future asbestos inertisation processes.

A wide range of atmospheric pollutants was considered, including gaseous inorganic molecules (SO_2, NO_x, CO etc), VOCs, and particulate matter (PM). Since no standard methods were available, a dedicated methodology has been developed and applied for the measurement of total PM concentration, asbestos fibres, and several particulate-phase trace carcinogenic pollutants: heavy metals, PAHs, dl-PCBs, PCDD/Fs. The measurement of such a large list of pollutants was aimed at obtaining a very detailed picture of the environmental impact of asbestos inertisation.

In the pre-industrial prototype, only one kind of waste has been used consisting of mineral wool waste. This kind of waste is very common in the past and today to thermal insulation. Standard methods were used to monitoring several atmospheric pollutants such as: gaseous inorganic molecules (SO_2, NOx, CO), particulate matter (PM), heavy metals, PAHs, dl-PCBs, PCDD/Fs.

Paragraph 2.2 and Paragraph 2.3 presents materials and methods and results for the emissions assessment form the two prototypes, respectively.

Figure 2 Prototypes used for the tests to evaluate the emissions of the a) laboratory prototype b) pre-industrial prototype

2.2 Monitoring emissions of laboratory prototype

2.2.1 Materials and methods

2.2.1.1 The prototype characteristics

The prototype basically consists in a muffle furnace (model CVK/S Nannetti; inner dimensions: 380 mm length; 470 mm depth; 280 mm height, of which 210 mm available for sample housing) equipped with a customised cylinder smokestack (5 m height, 30 cm diameter), made in in stainless steel. At 3 m of height, the smokestack has a removable circular opening of 5 cm diameter, in order to collect and analyse the emissions derived from the thermal treatment.

The muffle furnace was used with two different temperature programs on the basis of the different sample type: a slower heating rate and a longer isothermal time were necessary for samples of type C. Figure 3 reports the temperature programs: total time was 450 min (7.5 h) for samples C and 210 min (3.5 h) for samples CP. The definition of different temperature programs was based on the amount of asbestos in the sample, according to previous experiments (Bloise et al., 2016).

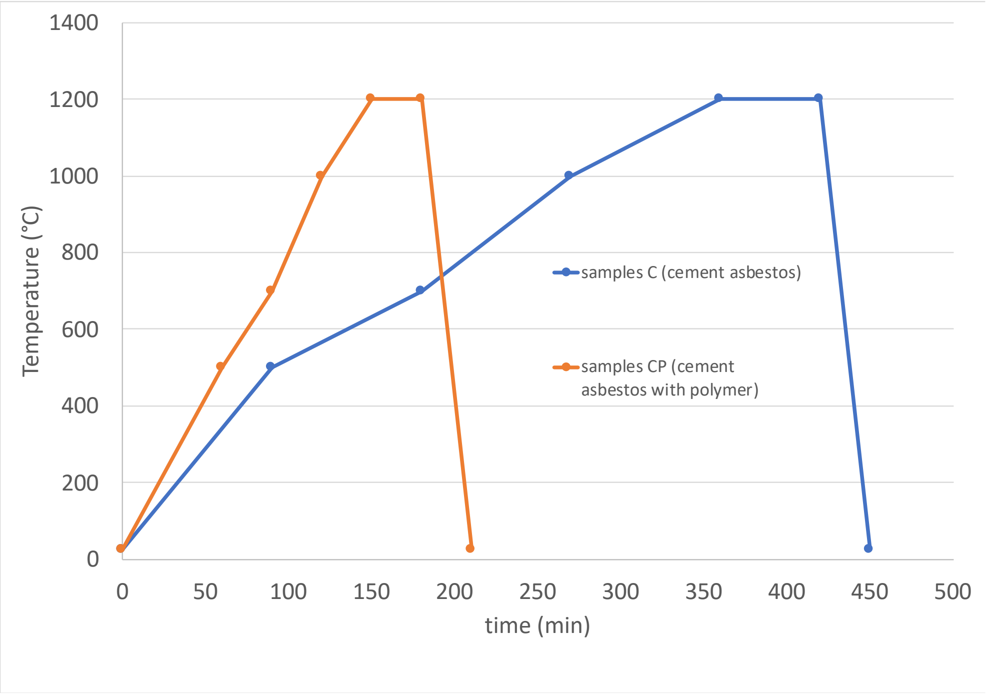


Figure 3 Temperature programs used in this study

2.2.1.2 *Asbestos Containing Waste samples*

Samples of ACW were kindly provided by ZETADI s.r.l. Two different kinds of ACW were used, in order to observe the impact of waste composition on the emissions.

The first kind of waste (sample code C) is commercial asbestos cement, and was chosen because it is the most prevalent kind of ACW. Two samples C weighing 5,294 and 4,627 kg each were prepared. The samples consisted in a stack of 8 asbestos cement sheets. The chemical composition (oxides in wt%) of the asbestos cement in this sample was: SiO_2=27.38; TiO_2=0.05; Al_2O_3=2.80; Fe_2O_3=1.50; MnO=0.03; MgO=6.94; CaO=34.9; Na_2O=0.04; K_2O=0.01; SO_3=1.25; Loss on ignition (L.o.i.)=25.1%. The mineralogical composition (wt%) of the asbestos cement in this sample as determined by the Rietveld method was: chrysotile asbestos=8,0(4); amphibole asbestos (crocidolite)=2.0(1); calcite 50.3(2); quartz=2.5(2); C_2S=3.5(2); portlandite=1.5(3); amorphous phase=32.2(5).

The second kind of waste (sample code CP) was asbestos cement with polymers. It was chosen because it is also a quite common kind of ACW and a significant emission of pollutants was expected, due to the presence of organic polymers. In this case, for this kind of waste, samples were prepared with two asbestos cement sheets between a polyurethane block weighing 1,035 and 1,135 kg each. The chemical composition (oxides in wt%) of the asbestos cement in this sample was: SiO_2=22.94; TiO_2=0.02; Al_2O_3=2.20; Fe_2O_3=1.10; MnO=0.01; MgO=8.34; CaO=32.7; Na_2O=0.02; K_2O=0.02; SO_3=1.55; Loss on ignition (L.o.i.)=31.1. The mineralogical composition (wt%) of the asbestos cement in this sample as determined by the Rietveld method was: chrysotile asbestos=11.5(4); calcite 63.5(2); quartz=1.1(2); bassanite=0.4(2); C_2S=0.5(2); amorphous phase=23.1(6).

For both types of waste (C and CP), two samples were prepared. The samples were packaged to simulate the security packaging used for asbestos removal. Such packaging consists in a polyethylene bag. In fact, asbestos thermal treatment would be performed on ACW including the packaging, due to safety reasons. It is also important to highlight that asbestos cement sheets are already coated with a vinyl chloride polymer, which is used during asbestos removal from roofs to avoid fibres dispersion. A schematic representation and pictures of samples are given in Figure 4 and Figure 5, respectively.

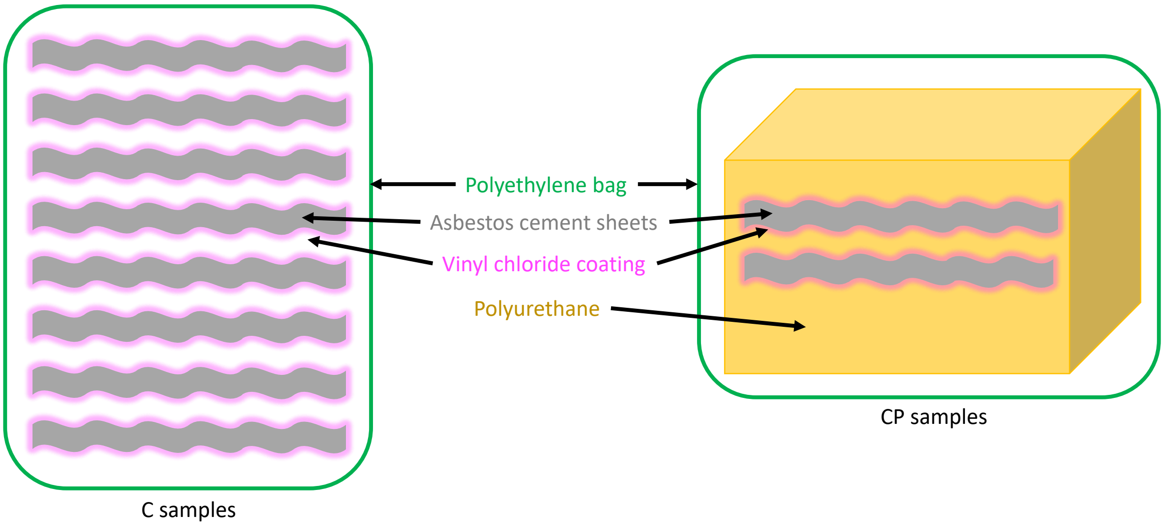


Figure 4 Schematic representation of samples composition

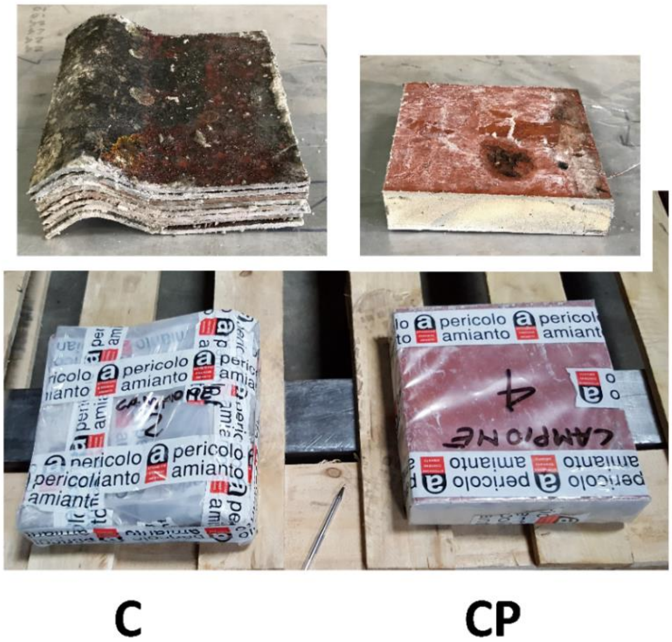


Figure 5 Pictures of samples C and CP with (up) and without (down) the polyethylene package

2.2.1.3 Analysis of gaseous emissions

During the thermal treatment of ACW, the atmospheric pollutants emitted were collected from the smokestack and analysed. The whole monitoring strategy is graphically represented in Figure 6.

Gaseous inorganic molecules were analysed online with a multi-gas analyser PG-250 (Horiba; Kyoto Cl, Northampton NN3 6FL, UK). The analyser allows the concentration measurement of the pollutants NO_x, SO_2 and CO using chemiluminescence method, UV fluorescence method and non-dispersive infrared (NDIR) method, respectively. Furthermore, the concentration of CO_2 (with an NDIR sensor) and O_2 (with a paramagnetic sensor) are also available. These two latter compounds are not atmospheric pollutants, but they were monitored to have an insight on the chemical reactions occurring during the thermal treatment of ACW. The high temperature of the tests (up to 1,200 °C) led to a high concentration of moisture (in terms of absolute volumetric concentration of water), and the damp fumes were capable of condensing when the gaseous effluent was cooled to be analysed. Hence, in order to avoid water condensation inside the measurement chambers, the PG-250 was equipped with a double stage moisture removal system, consisting in a gravity discharge separator followed by a thermal-electric cooling device.

Volatile organic compounds (VOCs) were collected with a portable dynamic dilution sampler, model DDS (TCR Tecora; 10 Rue de la Prairie, 91140 Villebon-sur-Yvettes, France). This device allows to dilute the sampled gas with clean air (using activated carbon filters), and its use was necessary in order to collect a significant volume of emissions without saturating the adsorption cartridge. VOCs were sampled with a stainless-steel probe, diluted with clean air (dilution ratio 1:4) and adsorbed on a cartridge filled with activated and graphitized carbon (200 mL min^{-1}). The combination of activated and graphitized carbon was chosen to avoid saturation with moisture (Brancaleoni et al., 1999; Paolini et al., 2019). Cartridges were analysed in laboratory using thermal desorption, cryo-focusing and gas-chromatography coupled to quadrupole mass spectrometry (TD-GC/MS) in full scan mode. The instrumental equipment consisted in a 100-xr TD desorption unit (Markes International) and an Agilent 7000 GC/MS system. The detailed instrumental conditions are reported in a previous study (Paolini, Petracchini, Carnevale, et al., 2018). Compounds were identified by comparison with mass spectra libraries, using the NIST MS Search software (National Institute of Standards and Technology; Gaithersburg, Maryland USA).

2.2.1.4 Analysis of particulate emissions

The total suspended particles (TSP) were isokinetically sampled during the thermal treatment of ACW. An isokinetic sampling equipment basically consists in a sampling probe with a known inner diameter, a Darcy tube and a thermocouple: the linear velocity of the emission is continuously monitored with the Darcy tube, and the sampling flow rate of the probe is regulated in order to have the same liner velocity of the emission. In this study, the isokinetic sampler Isostack (TCR Tecora) was used. During the final isotherm, the maximum temperature reached by the sampling probe was 556 °C. Isokinetic sampling was used for the measurement of TSP, asbestos fibres, heavy metals, and organic micropollutants (PAHs, dl-PCBs, PCDD/Fs), as described below. Figure 6 gives a graphical overview of the monitoring strategy.

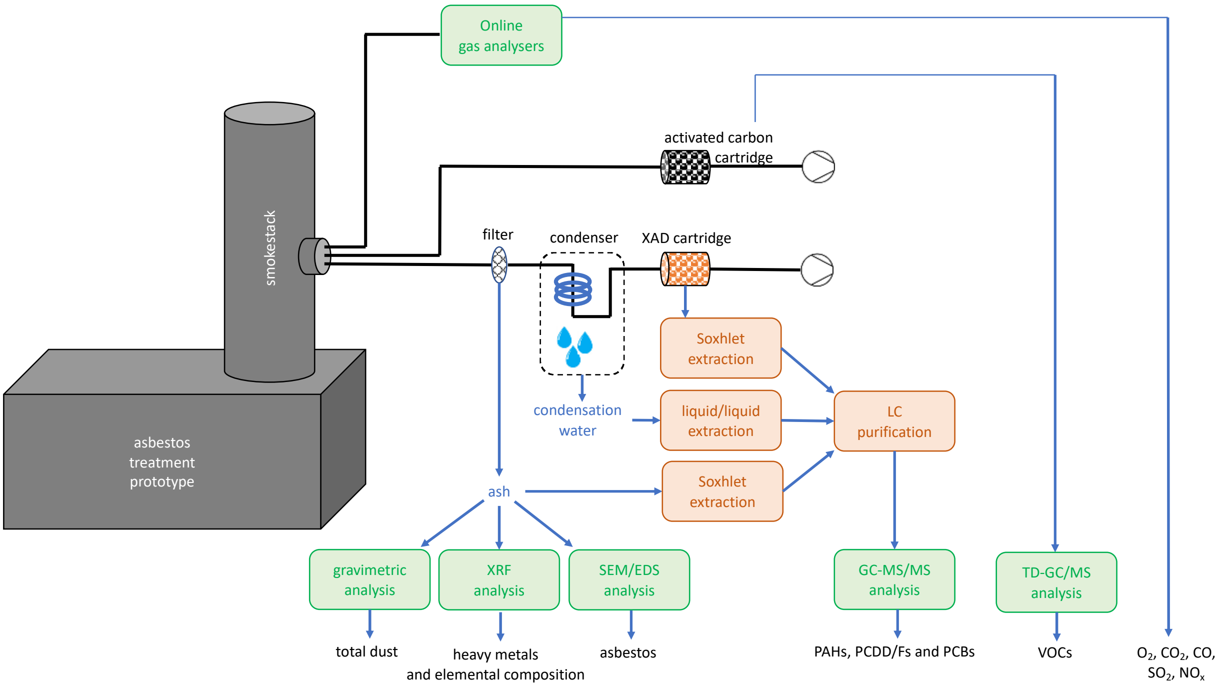


Figure 6 Monitoring strategy of emissions

The sampling equipment after the probe consisted of a filter, a condenser and an adsorption cartridge. A titanium sampling probe was used; sampling probe and filter holder were kept at 120 ± 5 °C: these operating conditions are necessary in order to avoid chemical degradation of organic micropollutants. Despite quartz fibre filters are the optimal device for sampling organic micropollutants, Teflon filters (47 mm diameter, 0.4 μm diameter) with high thermal stability (Sartorius; Otto-Brenner-Straße 20 37079 Goettingen, Germany) were used in this study, in order to allow the subsequent SEM and XRF analyses. After the filter, water was collected with a condenser at (6 ± 2) °C, and the dried gas was adsorbed in a cartridge filled XAD (poly-styrene/divinylbenzene) microspheres.

The filters were weighed in order to measure TSP concentration. Subsequently, an aliquot (1 cm^2) of each filter was cut and analysed by scanning electron microscopy (SEM) in order to verify the absence of asbestos fibres, using an ESEM Quanta-200 (Fei Company; Oxford Instruments) with 20 kV accelerating voltage, 4.0 μA beam current and high vacuum conditions. The filters were trimmed and placed on an aluminium stub with double-stick carbon tape and sputter-coated with gold (10 nm of thickness), using a Gold Sputter Coater - Emitech K550. Images were acquired using the signal of secondary electrons. The Surface of the filters was investigated working at different magnification levels, from 500× up to 20000× and 11 mm working distance. The chemical composition of particle specimens was investigated using an energy-dispersive X-ray spectrometer, X-EDS Oxford INCA-350. For each sample, 200 fields were analysed all over the surface. An EDS elemental analysis was carried out on every fibre. The combination of chemical and morphological analysis allows to assess the absence of asbestos fibres.

The remaining part of the filter was used for the analysis of organic micropollutants (PAHs, dl-PCBs, PCDD/Fs), together with condensation water and XAD cartridge. Quantification was carried out with isotopic dilution method, using perdeuterated PAHs and ^{13}C isotopes of dl-PCBs and PCDD/Fs. Filters and XAD were Soxhlet-extracted, while condensation water was extracted in a separation funnel. All extracts were concentrated in a rotary evaporator, before the subsequent steps of purification and gas-chromatographic quantification.

PAHs were purified in an alumina column (5 g deactivated with 2.5% of water; operating conditions as described in a previous study (Balducci et al., 2018) and analysed by gas-chromatography coupled to single quadrupole mass spectrometry in single ion monitoring mode, using the instrumental conditions described in a previous study (Paolini et al., 2016)

PCDD/Fs and dl-PCBs were purified with a double step liquid chromatography: a clean-up column with Na_2SO_4 and silica with H_2SO_4 and $AgNO_3$ and a second column with basic alumina for the separation of hydrocarbons, dl-PCBs, and PCDD/Fs. The purified samples were analysed by gas-chromatography coupled to triple quadrupole tandem mass spectrometry, in single reaction monitoring mode. The mass transitions associated to the loss of Cl_2 and CoCl were used for the determination of dl-PCBs and PCDD/Fs, respectively. The whole procedure is described in full details in a previous study (Benedetti et al., 2017).

All analyses of PAHs, PCDD/Fs and dl-PCBs were carried out in an Ultra Trace GC coupled to a TSQ mass spectrometer, with a TriPlus autosampler; the software used is Xcalibur (hardware and software purchased from Thermo Fisher Scientific).

2.2.1.5 Calculation of emission factors

The emission factors were calculated in order to define the emissions as a function of the mass entering the furnace chamber.

The emission factors (*EF*s) are defined as the ratio between the mass of pollutant emitted m_p and the mass of waste treated m_W, and are calculated as follows:

$$EF = \frac{m_p}{m_w} = \frac{C_p \times Q}{m_w}$$

where C_p is the concentration of the pollutant and Q is the normalised dry flow rate. This latter was measured with the Darcy tube of the isokinetic sampler, knowing the section of the smokestack.

2.2.1.6 Analysis of solid residue

The solid residue inside of the muffle furnace at the end of the heat treatment was collected and analysed. This solid by-product is actually can have several industrial applications but the recycling of this secondary raw material is allowed only if it is totally free of asbestos fibres. For this purpose, a phase analysis was carried out using X-ray powder diffraction (XRPD) and scanning electron microscopy (SEM). For each treatment cycle, different portions of solid residue were collected in the top, in the middle and in the bottom.

The samples were gently ground in an agate mortar for 2 min and the obtained powder was analysed by the XRPD technique to accomplish a qualitative phase analysis. The instrument used was θ/θ diffractometer (Malvern Panalytical; The Bruel & Kjaer, Jarman Way, Royston SG8 5BQ, UK) with Cu Kα radiation, 40 kV and 40 mA, equipped with a real time multiple strip (RTMS) detector. A virtual step scan (0.0167 °2θ) was performed in the 3-70 °2θ range.

The microstructure of the raw samples was investigated by scanning electron microscopy (SEM) using a FEI Quanta-200 instrument. The specimens were attached on aluminium stubs using carbon glue and subsequently coated with a 10 nm thick gold film.

2.2.2 Results and discussion

2.2.2.1 *Gaseous inorganic pollutants*

Figure 7 shows the emissions of CO_2 and O_2, for sample C1. Similar results were obtained for all other samples. Figure 8 and Figure 9 show the concentration of CO, NO_x and SO_2 for samples C1 and CP1, respectively. Similar results were obtained for duplicate tests C2 and CP2. Therefore, results can be considered statistically representative of typical emissions of a hypothetical plant used for the thermal treatment of asbestos. The average and maximum concentration is reported in Table 3.

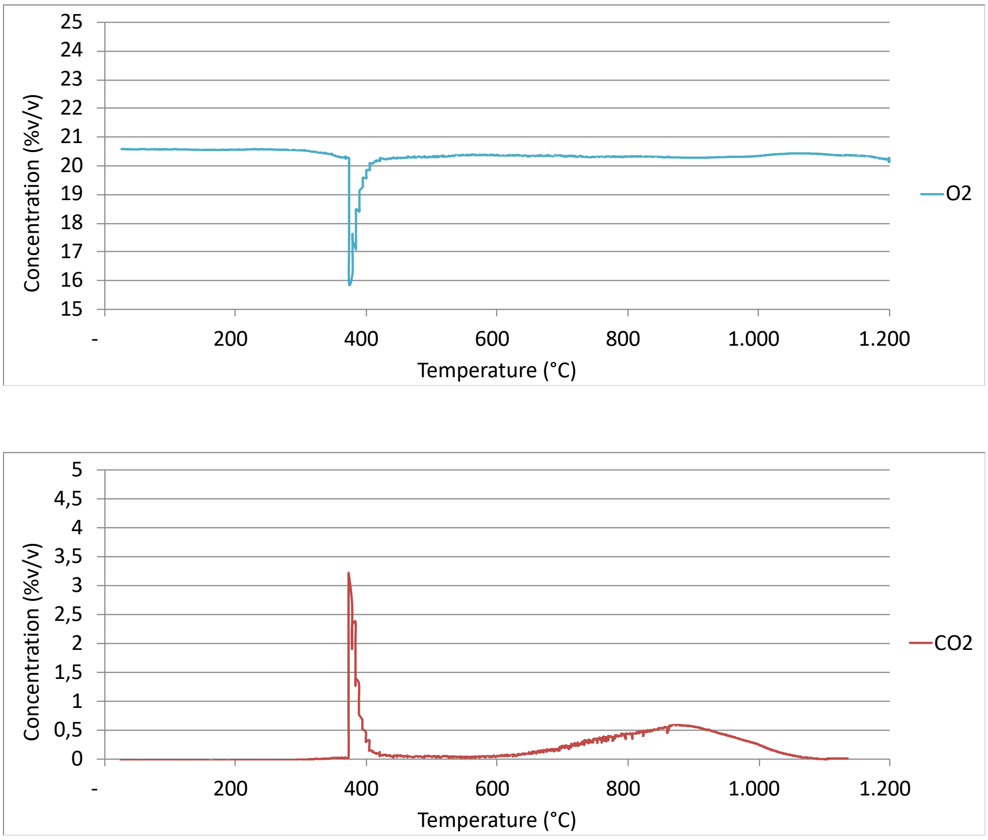


Figure 7 Concentration of O_2 and CO_2 during the treatment of sample C1

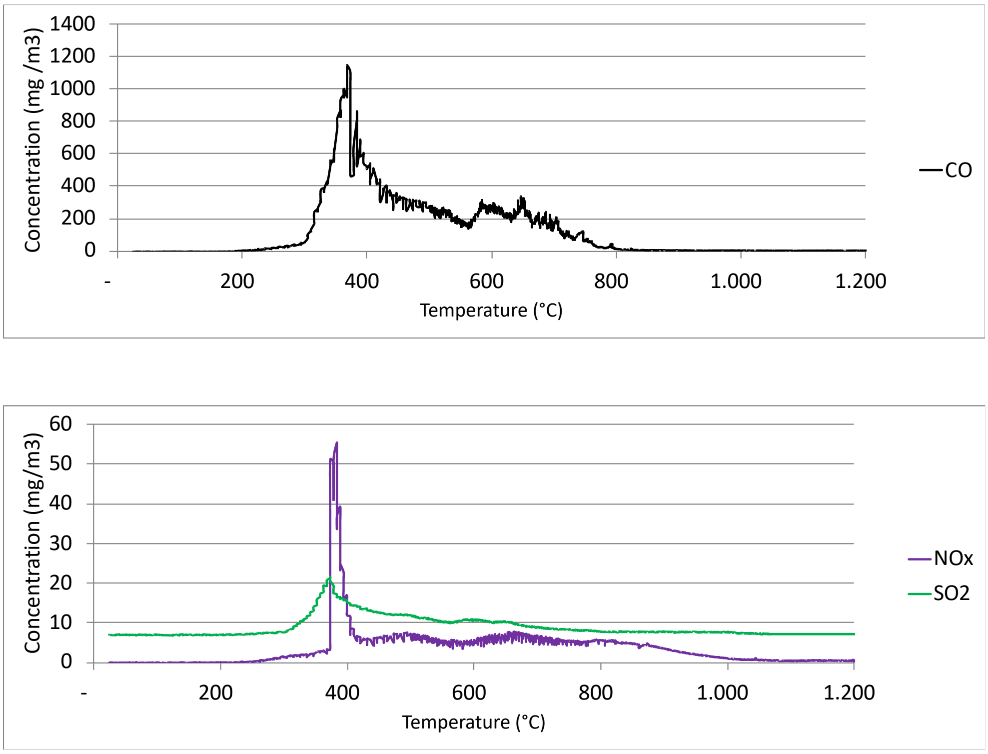


Figure 8 Concentration of CO, NO_x and SO_2 during the treatment of sample C1

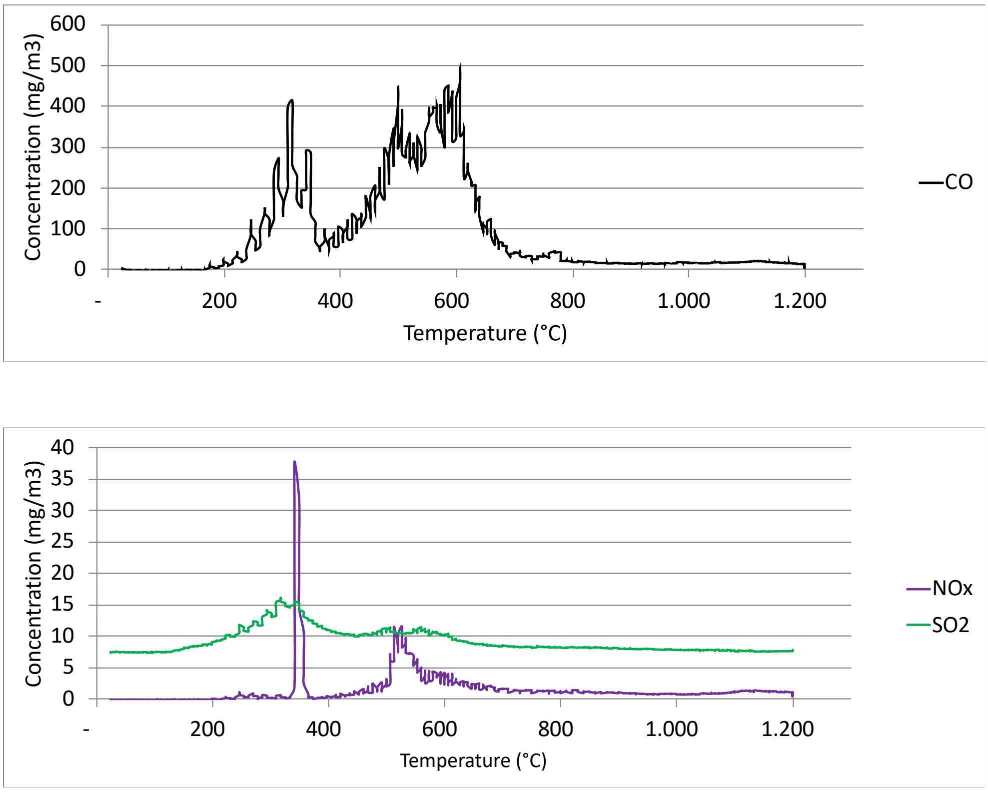


Figure 9. Concentration of CO, NO_x and SO_2 during the treatment of sample CP1.

Table 3. Average and maximum concentration of CO, NO_x and SO_2.

Parameter		**Samples C**	**Samples CP**
Average concentration (mg m^{-3})	CO	95	79
	NO_x	3.13	1.60
	SO_2	8.65	9.02
Maximum concentration (mg m^{-3})	CO	1140	493
	NO_x	55.56	37.80
	SO_2	21.57	16.20

The major CO_2 emission peak is related to the thermochemical conversion of carbonate from asbestos cement, in the temperature range 600-1050 °C. Other CO_2 peaks are related to the combustion of the organic fraction of ACW: this is the case of the temperature range 350-425 °C. In this case, the O_2 reduction is directly due to oxidation reactions. In this temperature range, CO emission peaks are also observed, due to the incomplete combustion: indeed, both samples C and CP are packaged in a polyethylene bag. Small amounts of organic materials are also present in asbestos cement sheets, including e.g. moss, lichens, soil etc. For CP samples, CO emission is also due to the oxidation of polyurethane.

The peaks of NO_x formation are observed at its typical stability temperature (Glarborg et al., 2018). Higher NO_x emissions were expected for samples CP, due to the combustion of polyurethane (Guo et al., 2014): instead, maximum and average NO_x concentration is higher in samples C. This suggests that NO_x emissions are mainly due to other components such as the polyethylene bag, rather than to the polyurethane. Finally, it is interesting to observe the SO_2 emission due to the combustion of sulfur. Traces of this element are always present in every kind of waste, resulting in the formation of SO_2 (Courtemanche & Levendis, 1998; McPhail et al., 2014).

2.2.2.2 Volatile organic compounds

The volatile organic compounds (VOCs) identified during the experiments are reported in Table 4. VOC sampling was performed during the maximum combustion peaks. These combustion peaks were selected in correspondence of CO and CO_2 maximum concentration. Indeed, the maximum VOC concentration was expected during these processes.

Table 4 VOC concentration in mg m^{-3}

	Samples C	**Samples CP**
Acetone	0.10	0.18
1-chloro-1-propene	-	0.13
1-hexene	-	0.25
Chloro-butane	0.71	0.27
Benzene	12.7	16.5
1,2-dichloropropane	0.39	0.25
Toluene	1.53	2.80
Chlorobenzene	0.33	0.18
Ethylbenzene	0.18	0.45
1,3-xylene and 1,4-xylene	0.33	0.91
1,2-xylene	0.05	0.13
Styrene	6.89	2.98
1-ethyl-2-methyl-benzene	-	0.11
1-methylstyrene	0.32	0.20
Naphthalene	-	0.49

Most of identified VOCs are aromatic compounds, such as benzene, toluene, styrene, xylenes, ethylbenzene and naphthalene. They are commonly associated to the incomplete combustion of the carbon-containing substances, so they can be attributed to the oxidation of the plastic bag were the ACW was confined. For samples CP, aromatic VOCs can also be due to the combustion of polyurethane: indeed, the highest VOC concentrations are observed for CP samples.

The concentration ratios are consistent with the type of emission: in particular, the toluene/benzene and toluene/xylenes ratio are typical of high temperature emissions, similar to what is observed in steelworks, solid fuel burning, cement plants and waste incinerators (Chattopadhyay et al., 1996; Hedberg et al., 2002). The high concentration of styrene is also consistent with expected values of this kind of process. Indeed, styrene is a quite reactive and instable compound: consequently, its concentration is generally low in atmospheric air, but relatively high styrene levels can be observed in some industrial emissions, shortly after his formation (Batterman et al., 2002; Jia et al., 2008; Wei et al., 2008)

A surprisingly high concentration of chlorinated VOCs is observed for both sample matrices. This is likely due to the combustion of the coating used for ACW removal and transportation: indeed, this coating is a vinyl resin, consisting of vinyl chloride and vinyl acetate copolymers. The combustion of polyvinylchloride is known to be associated with emissions of organochlorine compounds (Aracil et al., 2005; Ruokojärvi et al., 2000): hence, the emissions of chlorinated VOCs is likely due to the vinyl coating rather than to the combustion of other components of ACW.

In general, VOC emissions are higher for samples containing a mixed matrix (asbestos and plastics, CP samples), rather than for samples containing only asbestos (C samples). For these latter samples, the emission might be mainly attributed to the combustion of the bag.

2.2.2.3 Total suspended particles

Total suspended particles (TSP) were sampled on filters and weighted, before performing the analyses described in the next paragraphs. The absolute concentration is higher for samples containing only asbestos cement (7.328 and 8.935 mg m^{-3} for C1 and C2 samples, respectively), while samples containing asbestos cement with polymers (3.837 and 2.408 mg m^{-3} for CP1 and CP2, respectively) led to an emission with a lower concentration of total suspended particles.

The higher concentration of TSP for C samples is partially due to the larger amount of ACW used for these tests, compared to CP. In order to normalise the measurements, an emission factor was calculated, dividing the total mass flow rate for the mass of waste used in each cycle. As reported in subsequent section 2.2.2.8, similar emissions are observed for samples C and CP. Consequently, the emission of particles is mainly related to the thermal degradation of concrete, rather than to the combustion of polyurethane.

2.2.2.4 Emission of asbestos fibres

Filters with collected particles were analysed by SEM-EDS to verify whether asbestos fibres are released in the atmosphere during the thermal degradation of asbestos waste. The presence of asbestos fibres was identified by using a combination of morphological and chemical parameters, as described in paragraph 2.2.1.4.

The SEM-EDS analysis confirmed the presence of a typical structure resulting from chrysotile degradation, which has been observed and described in a previous study (Gualtieri, et al., 2008). The microstructure is completely different from that of pristine asbestos: indeed, on the fibre, it can be observed the growth of crystals of sub-spherical silicates, which are typical of the minerals constituting the inert material. The EDS spectra confirm the presence of Ca- and Mg-silicates produced during the thermal inertisation. As an example, Figure 10 reports the SEM image and EDS spectrum for one fibre observed for sample C1.

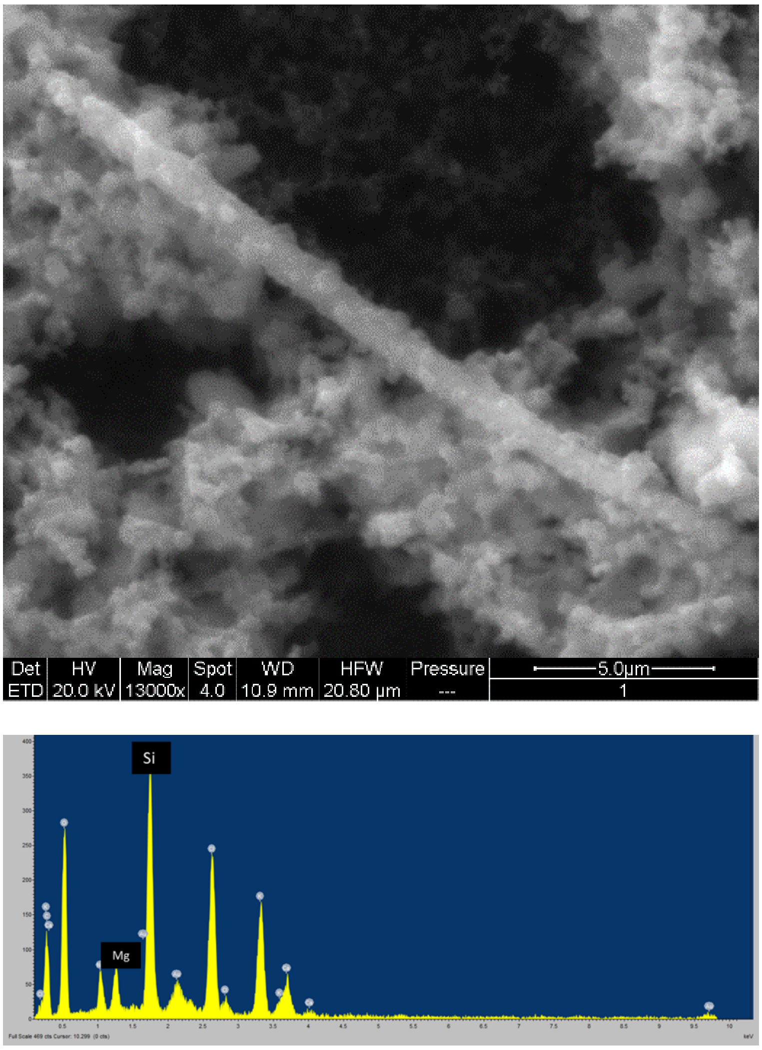


Figure 10 SEM images (top) and EDS spectrum (bottom) of a recrystallized asbestos fibre detected in the particulate released from sample C1

Such a structure has already been observed in the solid residue of ACW after thermal treatments, while this is the first experimental evidence of its presence in the released particulate.

In general, a very limited number of fibres was detected. None of them can be classified as asbestos. Most of the detected fibres are formed through the process described above of thermal degradation of chrysotile. The other fibres are exotic, being ceramic refractory fibres released from the wool medium used in the experiment to provide a better insulation of the furnace and pipes. They should be neglected in the study because all of them display diameters greater than 3 µm and hence, are not classified as respirable fibres. Besides that, their presence can be eliminated simply changing the type of refractory insulating medium for the furnace and pipe systems. Table 5 summarises all the identified fibres.

Table 5 Fibres identified in the emitted particles

Sample	Fibres from chrysotile degradation	Other fibres	Total number of identified fibres
C1	2	0	2
C2	2	2	4
CP1	2	6	8
CP2	1	2	3

The absence of asbestos fibres in the emission from ACW thermal treatment is a very promising result, which confirms the safety and the replicability of the proposed process, as a valid and sustainable alternative to landfilling.

2.2.2.5 Elemental composition of particles

The elemental composition of total suspended particles is reported in Table 6. Typical elements of thermal degradation of cement are easily identifiable, such as Ca. Some of the elements related to the degradation of asbestos such as Mg are below the detection limit, probably because they remained mainly associated with the bottom ashes inside the furnace: nevertheless, as described in previous paragraph 2.2.1.4, the SEM-EDS analysis allowed to detect the specific presence of Mg on fibres resulting from the thermal degradation of chrysotile.

Results confirm the presence of particles released from the degradation of cement: indeed, the inlet ACW and the sampled emissions have a similar elemental profile and concentration distribution of heavy metals. For instance, the concentration of Fe and Si is relatively high, while low concentrations are observed for other elements such as Zn.

An unexpectedly high concentration of Cl was observed, ranging from 0.9 to 1.2 mg m^{-3}, indicating that Cl can constitute up to 14% of total suspended particles. Such an abundance of this element in the emissions the thermal treatments of solid waste is quite unusual, regardless of the type of feedstock (Colangelo et al., 2012; Margallo et al., 2015). As previously discussed for chlorinated VOCs in paragraph 2.2.2.2, the chlorine source probably lies in the ACW coating, containing vinyl chloride.

It is worth mentioning that the high concentration of Cl can increase the volatilisation of heavy metals (Yu et al., 2017), hence the substitution of the vinyl chloride coating with a chlorine free resin might reduce the concentration of heavy metals in the emission. For instance, $TiCl_4$ has a boiling point of only 409,5 K, and the volatility of this compound has probably contributed to the relatively high elemental concentration of Ti in the emission.

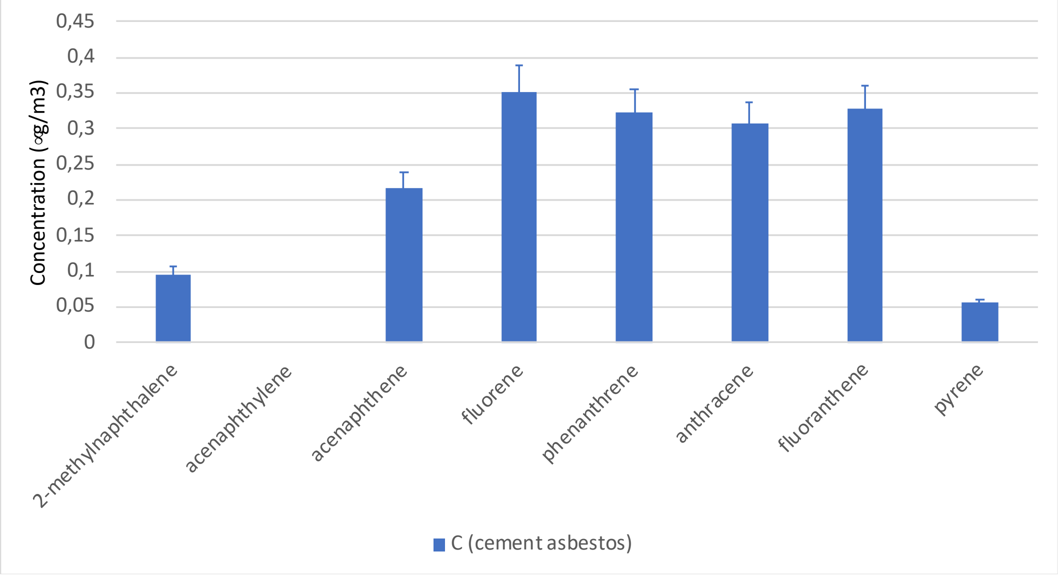
Table 6 Elemental composition of total suspended particles (ng m-3)

	Samples C	**Samples CP**	**uncertainty**
Na	288700	<1200	400
Mg	<300	<300	100
Al	8250	2050	100
Si	4990	4095	70
P	1130	1395	10
S	321000	10400	200
Cl	1043850	101595	70
K	1851000	6550	160
Ca	2590	2285	30
Ti	19196	145	20
V	77	<15	5
Mn	118	208	5
Fe	905	10650	20
Co	<30	<30	10
Ni	115	180	10
Cu	29625	930	50
Zn	1885	300	10
Ga	54.5	80.5	5
Ge	<15	<15	5
As	<15	<15	5
Se	69.5	<15	5
Br	2935	97	5
Rb	8450	<300	100

2.2.2.6 *Polycyclic aromatic hydrocarbons*

The concentration polycyclic aromatic hydrocarbons (PAHs) is reported in Figure 11. For both C and CP samples, the most abundant PAHs are those with 3 aromatic rings (acenaphthene, acenaphthylene, phenanthrene and anthracene), which are also those with the least toxicity to humans (International Agency of Research on Cancer (IARC), 1987, 2010, 2012). A typical profile for an industrial emission can be observed: compared to measurements in ambient air, there are high ratios of anthracene/phenanthrene and benzo(a)pyrene/benzo(e)pyrene. Indeed, these less stable compounds (i.e. anthracene and benzo(a)pyrene) are relatively more concentrated in emissions: once they are released into the atmosphere, they degrade faster than their more stable isomers (i.e. phenanthrene and benzo(e)pyrene, respectively) (Cecinato et al., 2014; Katsoyiannis & Breivik, 2014; Petracchini et al., 2017). This degradation is mainly due to oxidative processes (Balducci et al., 2017). Similarly, it was possible to detect some particularly reactive PAHs which are typical of emissions and rarely quantifiable in ambient air, such as cyclopenta(c,d)pyrene (Yagishita et al., 2015).

As expected, the experiments carried out in the presence of polyurethane (CP samples) led to higher emissions of PAHs: therefore, it is confirmed that the emission of these pollutants is mainly related to the presence of other carbon-containing components in the ACW feedstock, rather than to asbestos cement.



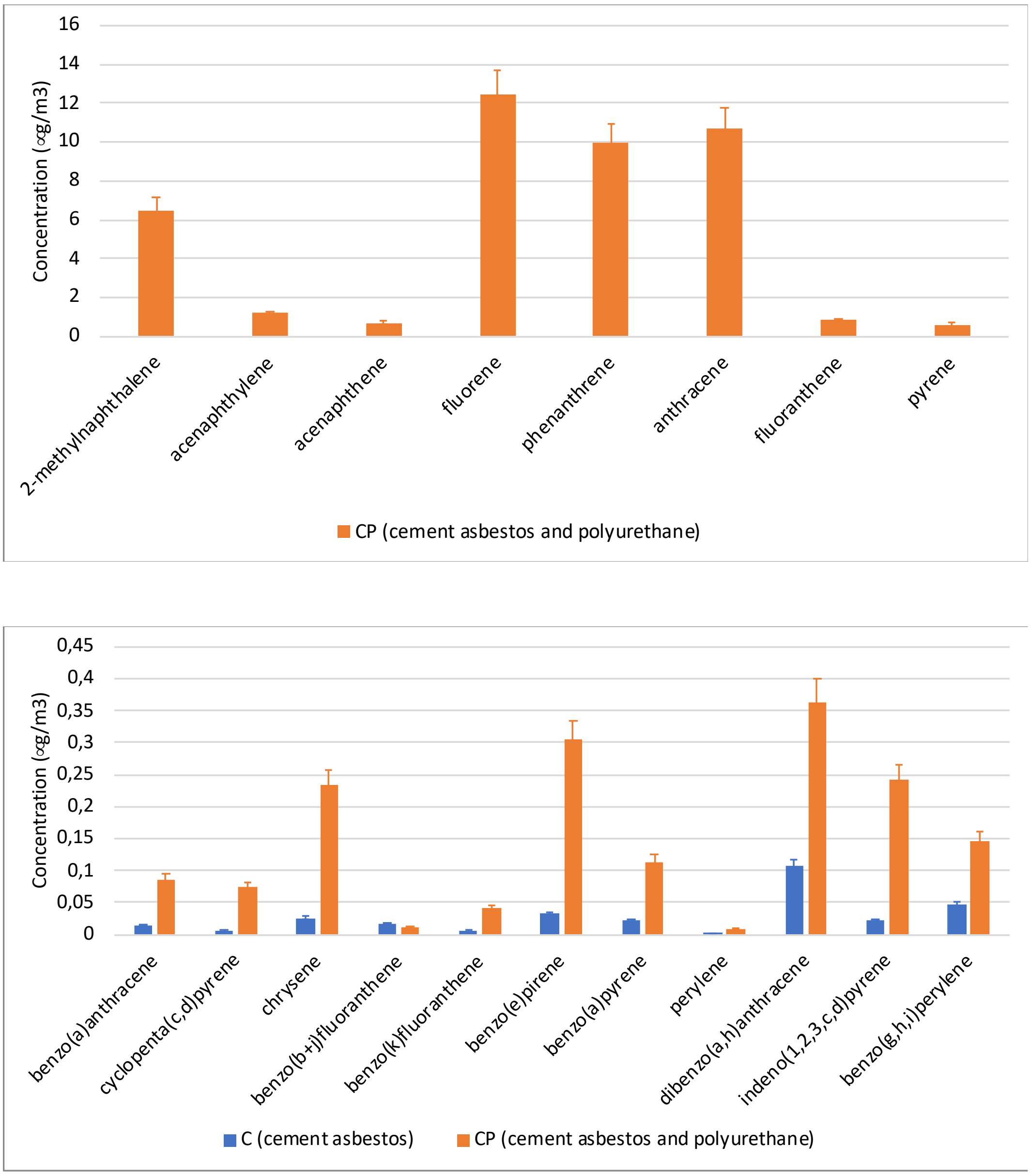


Figure 11 Concentration of PAHs

2.2.2.7 PCDD/Fs and dl-PCBs

Figure 12 reports the concentration of polychlorodibenzo-p-dioxins (PCDD) and polychlorodibenzofurans (PCDF). Values are expressed both in absolute concentration and in toxic equivalence (TEQ). In fact, all the compounds belonging to this class have the same carcinogenic effect on humans, mediated by the aryl hydrocarbon receptor (AhR) (Denison & Nagy, 2003; Fernandez-Salguero et al., 1996; Hankinson, 1995). However, for some compounds this effect is stronger than others. The compound with the most marked carcinogenic effect is the 2,3,7,8-tetrachlorodibenzo-p-dioxin (2,3,7,8-TCDD). The toxicity of the other compounds is expressed in relation to 2,3,7,8-TCDD through toxic equivalency factors (TEF) defined by the World Health Organization (van den Berg et al., 2006).

Despite its highest TEF, the 2,3,7,8-TCDD contributes less to the TEQ, due to its relatively low concentration. This finding is quite common in environmental monitoring (Bogdal et al., 2013; Breivik et al., 2004). Instead, the main contribution is given by TeCDF and PeCDD/Fs that are present at higher concentrations. Compared to atmospheric air measurements with a stronger predominance of more persistent congeners (HpCDD/Fs and OCDD/Fs) (Bogdal et al., 2013; M. S. Wang, Chen, Huang, etal., 2010), in this study there is a relative abundance of PeCDDs, which is typical of industrial emissions, such as municipal solid waste incinerators (L. C. Wang et al., 2010; G. Zhang et al., 2012) and production of cement (Conesa et al., 2011; C. Li et al., 2014) and steel (Gao et al., 2019; Y. F. Li et al., 2010). Even though the formation of PCDD/Fs in the thermal treatment of other hazardous waste was previously reported (van Caneghem et al., 2010; C. Wang et al., 2019), this is the first experimental evidence of the presence of PCDD/Fs in the emission of asbestos thermal treatments.

Differently from PAHs (previous paragraph 2.2.2.6), the highest concentration is observed for samples with only asbestos cement (C samples), while the presence of polymers in the ACW (CP samples) did not increase the emission of PCDD/Fs. Consequently, the emission of these pollutants is little related to the combustion of the polyurethane polymer. Since it is not likely that PCDD/Fs are produced from cement (which is a substantially inert and chlorine-free material (Conesa et al., 2011; C. Li et al., 2014), the formation of these pollutants is attributed to the coating. Indeed, as already discussed in previous sections for chlorinated VOCs (paragraph 2.2.2.2) and for chlorine content in TSP (paragraph 2.2.2.3), the coating is a vinyl acetate and vinyl chloride resin. The combustion of the latter polymer is a well-known source of PCDD/Fs, and a relationship has been clearly established between the presence of PVC in incinerators feedstock and the concentration of PCDD/Fs in the flue gas (Stec et al., 2013; Yasuhara et al., 2006). The relatively high formation rate of dioxins observed in this study can be mainly attributed to the high temperature used for the asbestos inertisation, since temperature has been identified as the leading factor for PCDD/Fs “de novo” synthesis from PVC (Kim et al., 2004; Takasuga et al., 2007). Moreover, dioxin formation can also be promoted by the presence of metals (Yasuhara et al., 2005) and nanoparticles (Font et al., 2010), which are released during the degradation of cement asbestos.

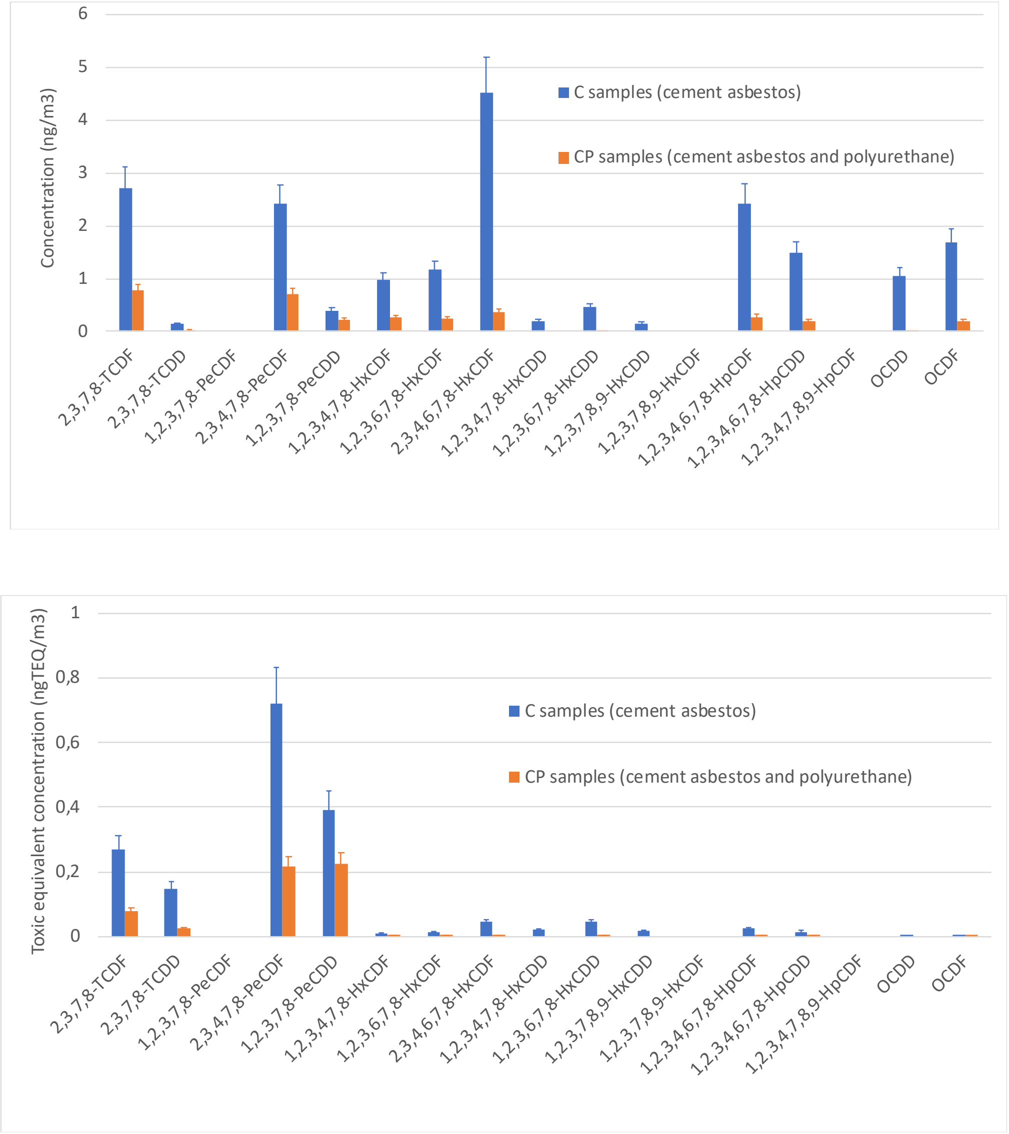


Figure 12 Concentration of PCDD/Fs

This result is very important in order to plan a sustainable development of the proposed methodology: indeed, before carrying out a thermal treatment of ACW on an industrial scale, the coating should be totally substituted with a different kind of polymer, without vinyl chloride. Since similar problems have also been reported for other chlorinated polymers (Aracil et al., 2010), a chlorine-free polymeric resin should be recommended.

In addition to PCDD/Fs, dioxin-like polychlorinated biphenyls (dl-PCBs) were also monitored. As these compounds have a similar toxicity, they also have a TEF referred to 2,3,7,8-TCDD (van den Berg et al., 2006). Compared to PCDD/Fs, dl-PCBs have lower TEFs: however, they are monitored because of their higher concentration. Results obtained are reported in Figure 13.

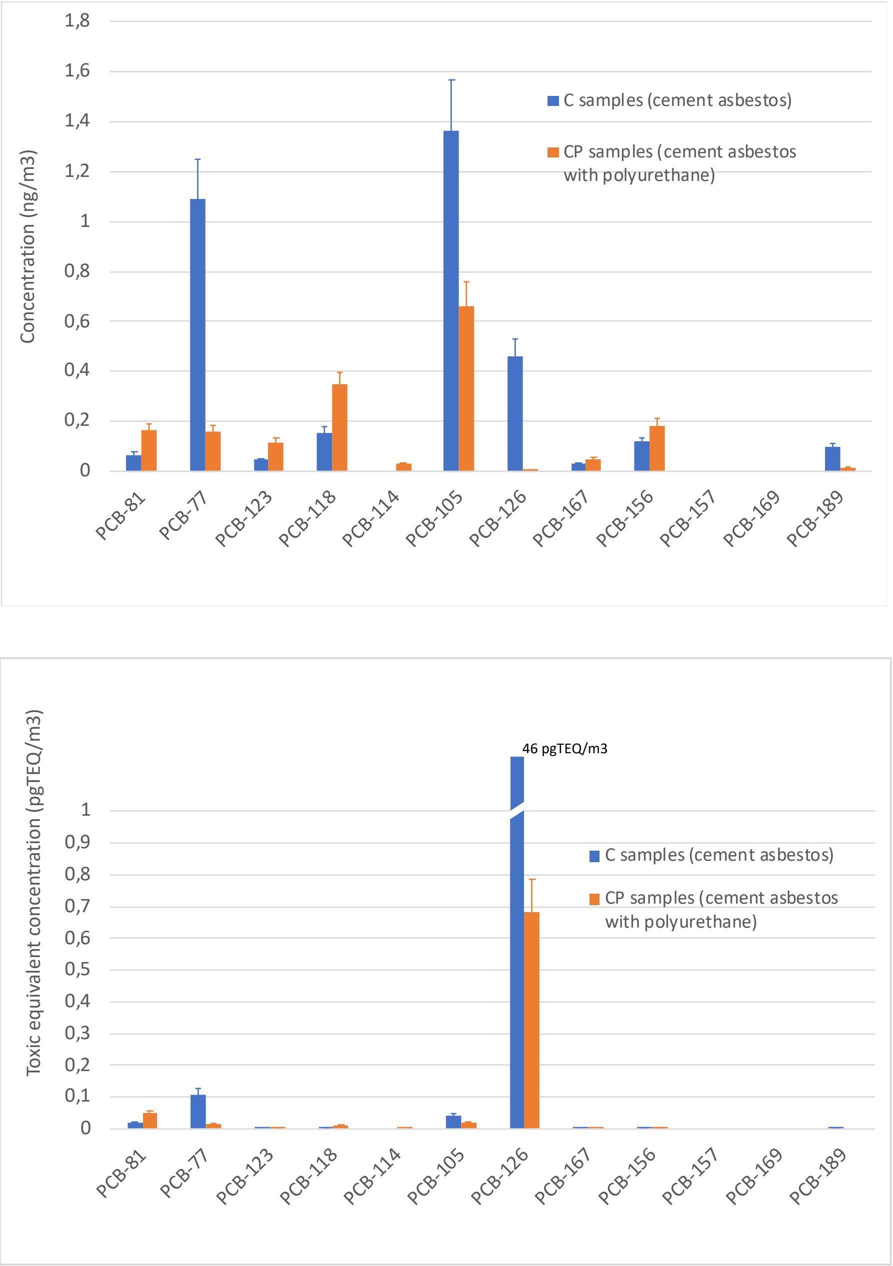


Figure 13 Concentration of dl-PCBs

Like PCDD/Fs, also for dl-PCBs the highest emissions are observed for CP samples. Consequently, the emissions of dl-PCBs can be attributed to the PVC coating rather than to the combustion of the polyurethane. Indeed, the formation of dl-PCBs from PVC combustion has been known since the late 1970s (Ahling et al., 1978).

Compared to measurements carried out in ambient air (Y. F. Li et al., 2010; Rudel et al., 2010), a predominance of PCBs with a lower degree of chlorination is observed, which is typical of emissions (M. S. Wang, Chen, Lai, et al., 2010). However, the contribution to the TEQ is substantially given by the PCB-126 due to its relatively higher TEF.

It is worth noting that dl-PCBs are less concentrated than PCDD/Fs: similar results have been reported in few previous works (Takasuga et al., 2004; M. S. Wang, Chen, Lai, et al., 2010; Yasuhara et al., 2006). A consequence of the lower concentration of PCBs is their small contribution to the TEQ, which is essentially given by the PCDD/Fs.For C samples, the toxic equivalent concentration is 1,720 and 46 pgTEQ/m^3 for PCDD/Fs and dl-PCBs, respectively (total 1,766 pgTEQ/m^3). For CP samples, the toxic equivalent concentration is 558 and 079 pgTEQ/m^3 for PCDD/Fs and dl-PCBs, respectively (total 559 pgTEQ/m^3).

2.2.2.8 Emission factors

Table 7 reports the emission factors (EFs) of selected pollutants, calculated as described in paragraph 2.2.1.5. In order to evaluate the emission factors for an industrial scale plant, the eventual addition of some emission abatement system is also taken into account. In total, four scenarios are considered: asbestos containing waste with and without plastic waste (polyurethane), and with and without emission abatement systems.

Indeed, it is well-known that an 80-99% abatement of heavy metals can be achieved by means of commonly used filters for the removal of particles, such as e.g. cyclones followed by bag filters (Quina et al., 2008; Su et al., 2015), or electrostatic precipitators (Świetlik et al., 2012; G. Zhang et al., 2013).

Similarly, the use of commercial filters in the plant scaling up would significantly reduce these emissions of organic pollutants like PAHs and dioxins (Chung et al., 2010; Jedlička et al., 2012).

More in general, pollutant abatement can rely either on primary or secondary control technologies (Srivastava et al., 2005). The primary control consists in avoiding the formation of pollutants, while the secondary control consists in the removal of pollutants after its formation. In a plant scale up, the accurate control of temperature, air flow and residence time would likely result in a primary reduction of e.g. NO_x and VOC emission factors. However, since this abatement cannot be calculated, the emission factors will be conservatively overestimated, ignoring possible primary control measures.

For secondary control, the following abatement systems are considered: a selective catalytic reduction (SCR) for NO_x, a dry absorption of SO_2 and fabric filter for PM. The removal of VOC and CO by means of a post-combustion system is not taken into account since it would not be realistic due to its cost.

An abatement efficiency of 70%, 90% is calculated for NO_x and SO_2: these are the most conservative performances for the considered technologies (EPA, 2002).

As for PM, its abatement efficiency in fabric filters depends on the particle size. The European Environmental Agency reports an abatement efficiency of 99.9% for total PM (EEA, 2019). The Biomass Energy Research Center (BERC) reported a 99.0% efficiency for both PM_{10} and $PM_{2.5}$ (BERC, 2011). The Nordic Council reports a 95% and 99% efficiency for $PM_{0.1}$ and PM_1, respectively (Lindberg, 2012). The Energy Efficiency and Conservation Authority of New Zealand reports 99.6%, 99.6 %, 99.9% and 99.95% efficiency for PM_1,

PM_2, PM_5 and total PM (EECA, 2012). The US EPA reports an efficiency range between 99.0% and 99.9%, for both PM_{10} and $PM_{2.5}$)(EPA, 2003) Finally, the Dutch agency TNO reports an efficiency range of 90-99 %, 95-98 % and 98-99.5 % for $PM_{0,1}$, $PM_{2,5}$, PM_{10}, respectively (TNO, 2002). In this study, a very conservative abatement efficiency of 99% for total PM will be considered.

Heavy metals, PAHs and dioxins are bound to the fine fraction of particles ($PM_{2.5}$ or below): considering that desorption and vaporisation can also occur during the fabric filtration, an abatement efficiency of 90% will be conservatively calculated for these pollutants.

Table 7 Emission factors (1 ton = 10^6 g)

Pollutant	**unit**	**Cement asbestos**	**Cement asbestos with abatement systems**	**Cement asbestos with polyurethane**	**Cement asbestos with polyurethane with abatement systems**
CO	kg/t	4.877		11.340	
NO_x	kg/t	0.161	0.0483	0.230	0.069
SO_2	kg/t	0.444	0.0444	1.295	129.5
Benzene	kg/t	<0.652		<2.368	
Toluene	kg/t	<0.078		<0.401	
Chlorobenzene	kg/t	<0.016		<0.025	
Styrene	kg/t	<0.353		<0.427	
Total particulate matter	kg/t	0.417	0.0417	0.448	0.0448
Asbestos fibres	fibres/t	0	0	0	0
Ti	g/t	0.986	0.0986	0.021	0.0021
V	g/t	0.003953	0.0003953	<0.002	<0.002
Mn	g/t	0.006084	0.0006084	0.029858	0.0029858
Co	g/t	<0.004	<0.004	<0.004	<0.004
Ni	g/t	0.005904	0.0005904	0.025839	0.0025839
Cu	g/t	1.521	0.1521	0.134	0.0134
Zn	g/t	0.096774	0.0096774	0.043065	0.0043065
Ga	g/t	0.002798	0.0002798	0.011556	0.0011556
As	g/t	<0.002	<0.002	<0.002	<0.002
fluoranthene	g/t	0.016849	0.0016849	0.123035	0.0123035

Pollutant	unit	Cement asbestos	Cement asbestos with abatement systems	Cement asbestos with polyurethane	Cement asbestos with polyurethane with abatement systems
pyrene	g/t	0.002865	0.0002865	0.088340	0.008834
benzo(a)anthracene	g/t	0.000744	0.0000744	0.012273	0.0012273
cyclopenta(c,d)pyrene	g/t	0.000298	0.0000298	0.010637	0.0010637
chrysene	g/t	0.001335	0.0001335	0.033447	0.0033447
benzo(b+j)fluoranthene	g/t	0.000816	0.0000816	0.001794	0.0001794
benzo(k)fluoranthene	g/t	0.000303	0.0000303	0.005972	0.0005972
benzo(e)pirene	g/t	0.001653	0.0001653	0.043754	0.0043754
benzo(a)pyrene	g/t	0.001155	0.0001155	0.016250	0.001625
perylene	g/t	0.000082	0.0000082	0.001392	0.0001392
dibenzo(a,h)anthracene	g/t	0.005452	0.0005452	0.052022	0.0052022
indeno(1,2,3,c,d)pyrene	g/t	0.001129	0.0001129	0.034796	0.0034796
benzo(g,h,i)perylene	g/t	0.002392	0.0002392	0.021087	0.0021087
PCDD/F	mgTEQ/t	0.088303	0.0088303	0.080100	0.00801
dl-PCB	mgTEQ/t	0.002362	0.0002362	0.000113	0.0000113

As already discussed in previous sections, the EFs are very lower compared to the thermal treatment of other waste (EMEP/EEA, 2016), mainly because cement asbestos is an inert material with few carbon sources and a low content of heavy metals. For the same reason, the EFs of VOCs and PAHs are higher in the presence of polyurethane. The EFs of VOCs are overestimated, since their concentration was only measured during the maximum combustion peaks, while VOCs are likely lower or absent during the final isotherm.

2.2.2.9 Comparison with similar emission processes

As previously stated, this is the first experimental measurement of emissions from a thermal treatment of ACW. However, some useful comparison can be carried out with other similar processes such as municipal solid waste incineration (MSWI) and cement production.

For instance, the emission factors of NO_x and SO_2 are comparable with an MSWI (McPhail et al., 2014), especially if the presence of plastic from polyethylene bag and polyurethane is taken into account (S. Singh et al., 2011). It is important to remark that all pollutants are below the maximum concentration allowed by the relevant regulation. For instance, in the Italian case study (Legislative Decree n.152 of 2006), considering the kind of industrial facility with a similar process (cement plant), the emission limit value is 1,800 and 600 mg m^{-3} for NO_x and SO_2, respectively. As for VOCs, the generally low concentration of aromatic compounds and the relatively high percentage of styrene has been often reported for similar high temperature industrial processes (Wei et al., 2008).

In general, most of hazardous elements (e.g. Co, As etc.) are present in relatively low concentration, when compared with typical emissions from thermal treatments of other kinds of waste (A. Font et al., 2015; Guéguen et al., 2011). Other hazardous elements (e.g. Mn, Ni) are more abundant, but their concentration cannot be regarded as an issue for the authorisation of a hypothetical industrial-scale plant, in compliance with the relevant environmental regulation. For instance, in the Italian case study (Legislative Decree n.152 of 2006), considering the kind of industrial facility with the most restrictive regulation (thermal plants with nominal thermal power higher than 100 MW), the emission limit value for Ni is 0.5 mg m^{-3}, i.e. four orders of magnitude higher that the concentration measured in this study.

The concentration of PAHs is comparable to the emissions from the incineration of other kinds of waste (Sarti et al., 2017; T. Wang et al., 2017)

Similarly, the emissions of PCDD/Fs and dl-PCBs are comparable with standard municipal waste incinerators (EMEP/EEA, 2016). For an industrial scale-up, the local regulation on the emission of persistent organic pollutants should be taken into account. In the case study of Italy, the concentration limit is 0.01 and 0.5 for PCDD/Fs and dl-PCBs, respectively.

2.2.2.10 Characterisation of solid residue

The XRPD analyses performed on the solid residue confirmed the crystal-chemical transformation of the asbestos fibres. All the obtained samples are composed by Al-Ca-Mg silicates, namely akermanite, bredigite and merwinite. Chemical formula and characteristic peaks are resumed in Table 8.

Other minor components are larnite (Ca_2SiO_4), monticellite ($CaMgSiO_4$) and ternesite ($Ca_5(SiO_4)_2SO_4$). The latter appears as the shoulder of the main peak of the akermanite, and its presence is confirmed by the sulfur content in the elemental composition of particles. Quartz is also present: it comes from the initial cement matrix and remained unchanged after the thermal treatment. Quartz is mainly present in the bottom of the solid residue, which is the area where the heat from the oven is less intense. A selection of XRPD results are reported in Figure 14. Similar results were obtained for all other samples.

Table 8 Chemical formula and main XRPD peaks of the phases of solid residue

Phase	**Ideal chemical formula**	**Major peaks (°2θ, d-spacing, hkl)**	**JCPDS card**
akermanite	$Ca_2Mg(Si_2O_7)$	29.204, 3.055, 201 31.437, 2.843, 211	01-087-0051
bredigite	$Ca_{14}Mg_2(SiO_4)_8$	33.550, 2.669, 260 32.865, 2.723, 400 32.692, 2.737, 222	00-036-0399
merwinite	$Ca_3Mg(SiO_4)_2$	33.324, 2.686, 013 33.521, 2.671, 411 33.758, 2.653, 020	00-035-0591

Chrysotile or other asbestiform phases like amosite and crocidolite were not found in any sample of solid residue (see the absence of the reflections typical of the asbestos phases in the selected low angle region 9-13 °θ in Figure 13b). It can be concluded that the thermal treatment led to a crystal-chemical transformation of the ACW, since the phases found in all obtained samples correspond to inert, non-hazardous silicates.

This material can be recycled for several applications, including ceramics, clay bricks, concrete etc (Ligabue et al., 2020, Gualtieri, 2013; Viani et al., 2013; Gualtieri et al., 2011a). This is another important advantage compared to landfilling, since recycling avoids the consumption of renewable resources and promotes circular economy.

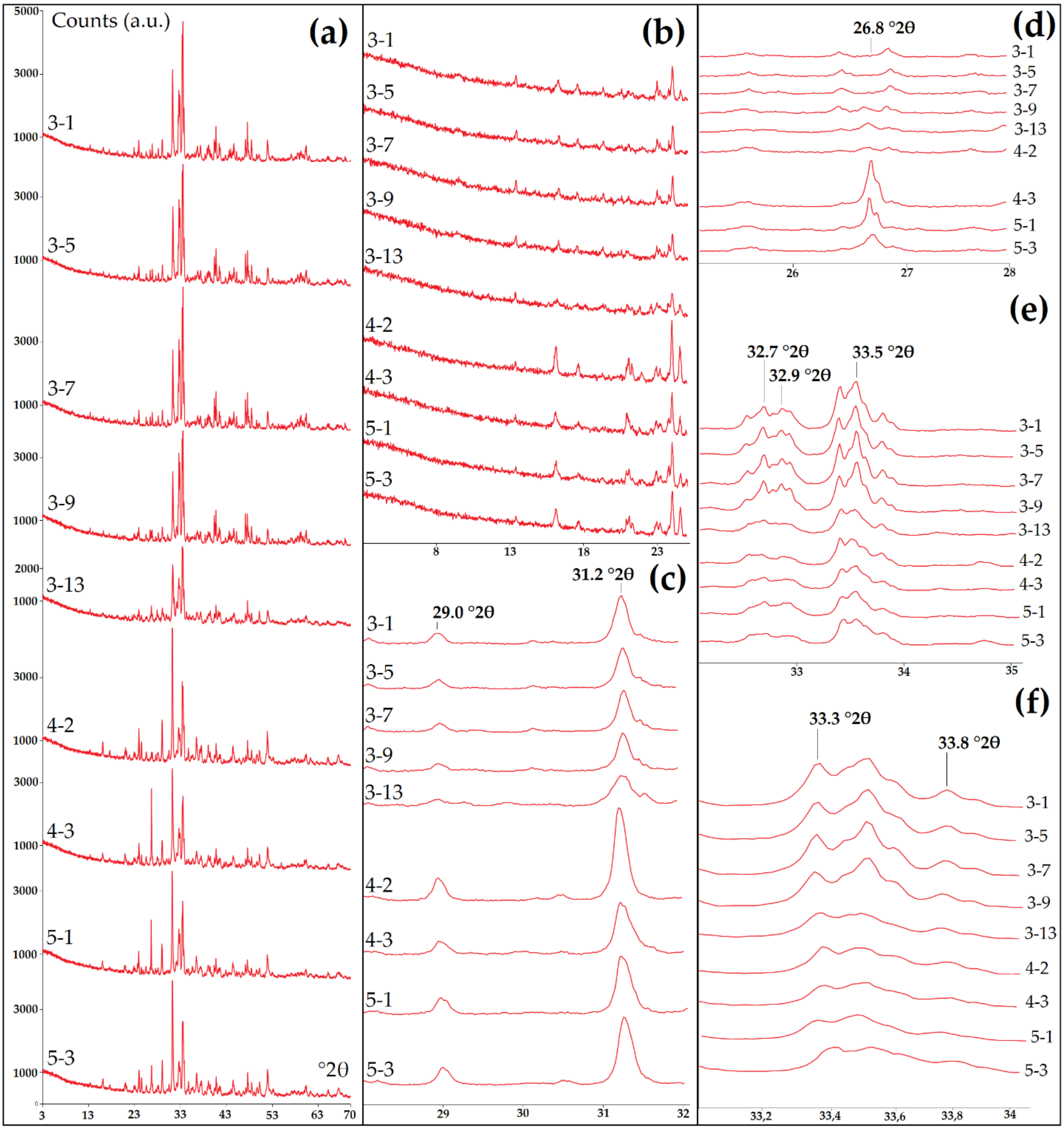


Figure 14. XRPD of solid residue.

Legend: (a) overview of the selected regions; (b) low angle region without asbestos peaks; (c) region showing the major peaks of akermanite; (d) region showing the major peaks of quartz; (e) region showing the major peaks of bredigite; (f) region showing the major peaks of merwinite. Nomenclature of sub-samples: sample CP1 top (3-1), middle-top (3-5), middle (3-7), middle-bottom (3-9) and bottom (3-13); sample CP2 central top (4-2) and side top (4-3); sample C2 side top (5-1) and central top (5-3).

2.3 Monitoring emissions of pre-industrial prototype

2.3.1 Materials and methods

2.3.1.1 The prototype characteristics

The pre-industrial prototype plant of Zetadì s.r.l. is a melting furnace for the inertisation of exhausted mineral wool, consisting of the following elements:

- melting furnace;
- primary burner fuelled by LPG and O_2. The use of O_2 considerably reduces nitrogen oxide (NO_x) emissions and makes it possible to reach the temperatures required for sintering spent mineral wool (approximately 1260 °C);
- an undersized LPG and air-fuelled afterburner to remove any unburnt substances. The use of the afterburner reduces the concentrations of VOCs (Volatile Organic Compounds), which are largely produced by the thermal degradation of the polymers present in the waste that feeds the plant;
- a chimney of the plant.

The furnace is maintained at a temperature of approximately 1000÷1050 °C and approximately 8 h of conditioning are required for its correct operation in nominal conditions.

The nominal feed rate is approximately 60÷70 kg/h of waste with a 20% drop in weight at the end of the process (40÷45 kg/h of secondary raw materials). The vitrified slag is thrown into the water for rapid cooling, which produces the crushing into fine grains useful for subsequent melting and production of glass or rock wool (Figure 15).

After the melting furnace and before the afterburner there is an 8 mm flue gas inlet where the gases have a high temperature (greater than 600 °C). In this inlet it is possible to assess the unburnt before the afterburner by using a sampling probe made of titanium, alumina or a quartz tube.

Due to the high flue gas temperatures, however, all sampling was carried out downstream of the afterburner using the two flue gas intakes installed on the chimney (Figure 16).

The chimney has an internal diameter of 210 mm with two standard sized sampling flanges installed, i.e. approximately 650 mm in diameter (Figure 16).

Figure 15 Melting furnace and slag outlet segment from the melting furnace and water-cooling tank

Figure 16 Chimney and sampling flange

2.3.1.2 Waste samples

Samples of mineral wool were kindly provided by ZETADI s.r.l.

The big bags with mineral wool waste used as samples for the tests were not specially created as in the previous test, but are real waste to be disposed of. This does not allow their specific content to be identified, but only their CER code, that could be CER 17.06.03* and CER 17.06.04. Therefore, there could be waste material from the removal of mineral wool inside the big bags, which could cause a variation in the composition of the emissions, so is possible to observe the real impact of waste composition on the emissions.

The samples composed by the waste were packaged into a big bags used to transport and/or store waste to simulate the security packaging used for asbestos removal. The polyethylene big bags containing waste were loaded directly into the melting furnace for the testing.

2.3.1.3 Methods applied for analysis and determination

The monitoring of pre-industrial prototype plant was carried out using UNI and ISO standards both for sampling and determination. The sampling was necessary for the comparison with the surveys carried out by an accredited laboratory for the purpose of sizing the emission abatement plant necessary for the commissioning and emission authorisations.

The plant showed fluctuations in flow rate and temperature over short periods, so only the average conditions obtained during sampling of organic micropollutants (long-term sampling) have been reported in the results.

As reported in more detail in the subsequent paragraphs, in Table 9 main reference standards used were shown.

Table 9 List of main standards used

Parameter	Standard	Title
Sampling section	UNI EN ISO 15259:2008	Air quality - Measurement of stationary source emissions - Requirements for measurement sections and sites and for the measurement objective, plan and report
Water vapour/humidity	UNI EN 14790:2017	Stationary source emissions - Determination of water vapour in ducts - Standardised reference method
Speed, Flow Rate, Pressure and Temperature: Manual Reference Method	UNI EN ISO 16911-1:2013	Stationary source emissions - Manual and automatic determination of velocity and flow rates of ducted flows - Part 1: Manual reference method
Speed, Flow Rate, Pressure and Temperature: Manual Reference Method	UNI CEN/TR 17078:2018	Stationary source emissions - Guidelines for the application of EN ISO 16911-1

Parameter	Standard	Title
Quality assurance	UNI EN 14181:2015	Stationary source emissions - Quality assurance of automated measuring systems
Carbon monoxide - CO	UNI EN 15058:2017	Stationary source emissions - Determination of carbon monoxide mass concentration - Standard reference method: non-dispersive infrared spectrometry
Nitrogen oxides - NO_x	UNI EN 14792:2017	Stationary source emissions - Determination of the mass concentration of oxides of nitrogen - Standard reference method: chemiluminescence
Oxygen - O_2	UNI EN 14789-2017	Fixed source emissions - Determination of the volumetric concentration of oxygen (O2) - Standard reference method: Paramagnetism (SRM)
Sulphur dioxide- SO_2	UNI 10339:1995	Determination of sulphur dioxide (SO2) in vented gas streams. Instrumental method with direct extractive sampling
Mass concentration of organic substances in gaseous and vaporous form (expressed as TVOC) in emissions from stationary source	UNI EN 12619:2013	Stationary source emissions - Determination of the mass concentration of total organic carbon in gaseous form - Continuous method with flame ionisation detector
PCDD/PCDF	UNI EN 1948-1:2006	Stationary source emissions - Determination of mass concentration of PCDD/PCDFs and dioxin-like PCBs - Part 1: Sampling of PCDD/PCDFs
PCDD/PCDF - purification	UNI EN 1948-2:2006	Stationary source emissions - Determination of mass concentration of PCDD/PCDFs and dioxin-like PCBs - Part 2: Extraction and purification of PCDD/PCDFs
PCDD/PCDF - analysis	UNI EN 1948-3:2006	Stationary source emissions - Determination of the mass concentration of PCDD/PCDFs and dioxin-like PCBs - Part 3: Identification and quantification of PCDD/PCDFs
PCB-DL	UNI EN 1948-4:2006	Stationary source emissions - Determination of mass concentration of PCDD/PCDFs and dioxin-like PCBs - Part 4: Sampling and analysis of dioxin-like PCBs
Metals	UNI EN 14385:2004	Stationary source emissions - Determination of total emission of As, Cd, Cr, Co, Cu, Mn, Ni, Pb, Sb, Tl and V
Particulate sampling: reference method for the measurement (SRM) of low concentrations of	UNI EN 13284-1:2017	Stationary source emissions - Determination of the mass concentration of dust at low

Parameter	Standard	Title
dust in gaseous streams conveyed at concentrations below 50 mg/m3 under normal conditions		concentrations - Part 1: Manual gravimetric method
PAHs	ISO 11338 -1:2003	Stationary source emissions - Determination of gas and particle - phase polyciclic aromatic hydrocarbons - Part1: Sampling
PAHs	ISO 11338 -2:2003	Stationary source emissions – Determination of gas and particle - phase polyciclic aromatic hydrocarbons - Part 2: Sample preparation, clean-up and determination
Monitoring of gaseous macro-pollutants: batch method, measurement of HCl and HF	UNI EN 1911:2010	Stationary source emissions - Determination of the mass concentration of gaseous chlorides expressed as HCI - Standard reference method

2.3.1.4 Analysis of gaseous emissions

The continuous measurements concerned the gaseous inorganic molecules: NO_x, SO_2, CO, CO_2 (Figure 18). Also, O_2 was also monitored continuously to assess the dilution of the fumes and the concentration of total organic carbon (TOC) in accordance with standard EN 12619:2013, to assess the presence of any unburnt substances and therefore to evaluate the efficiency of the combustion process.

The monitoring plan has been reported in Figure 17.

Day	Combustion Characteristic	Samping
13th April	Temperature of furnace: 1250 °C	start 16:40 end 19:30
	Temperature of post combustor: 1100 °C	
14th April	Temperature of furnace: 1250 °C	start 11:10 end 12:00
	Temperature of post combustor: 1100 °C	start 12:10 end 18:00
	Furnace powered until 16:30	
15th April	Temperature of furnace: 1050 °C	start 10:00 end 17:00
	Temperature post combustor: 900 °C	start 17:10 end 18:10
	Furnace powered until 14:30, subsequently, the temperature was raised to 1200 ° C with very slow pushing of the material left in the furnace, without carrying out the complete load	start 18:20 end 20:20

Figure 17 Monitoring plan of gaseous emission

These measurements were carried out using the follow analysers operating continuously:

1. multi-gas analyser PG-250 (Horiba; Kyoto Cl, Northampton NN3 6FL, UK) in compliance with UNI EN 14181:2015 - QAL 1 SRM certified according to UNI EN 14792:2017, UNI EN 15058:2017, UNI EN 14789:2017;

2. Ratfisch RS - Portable FID analyser for measurements of volatile organic substances (TOC) in compliance with EN 12619:2013.

The main features of the analysers used during the sampling campaign are shown in Table 10 and Table 11.

Table 10 Key features of HORIBA PG-250

Model	**PG-250**
Analytes measured	NOx/SO2/CO/CO2/O2
Method	NO_x: Chemiluminescence Detection method (CLD) SO_2/CO/CO_2: Non-dispersive Infrared Absorption (NDIR) O_2: Paramagnetic method
Certification	TÜV Rheinland 936/809014
Measurement range	NO_x: 0 to 25/50/100/250/500/1000/2500 ppm SO_2: 0 to 200/500/1000/3000 ppm CO: 0 to 200/500/1000/2000/5000 ppm, CO_2: 0 to 5/10/20 vol% O_2: 0 to 5/10/25 vol%
Repeatability	±0.5% of b.s. (NO_x: >= 100 ppm range, CO: >= 1000 ppm range) ±1.0% of b.s.
Linearity	±2.0% of b.s.
Drift	±1.0% of b.s./day (SO_2: ±2.0% of b.s./day)

Table 11 Key features of Ratisch RS-53T

Model	**RS-53T**
Measurement range	0-10/100/1.000/10.000/100.000 ppm (mg C/m^3)
Response time (T90)	1 second (without transport line)
Linearity	≤2% of b.s.
Sensitivity	≤2% of b.s. (gamma 1: external zero gas required)
Drift	± 2% of b.s./week
Span drift	± 4% of b.s./week
O_2 interference	≤ 2% of b.s. (with H_2/He)
Other interference	≤ 4% of b.s.

Figure 18 Analyser of gaseous emissions

2.3.1.5 Analysis of total suspended particles

The total suspended particles (TSP) were isokinetically sampled during the thermal treatment of samples according to the UNI CEN/TS 13649:2015 standard. In this study, the isokinetic sampler Isostack (TCR Tecora) was used. Isokinetic sampling was used for the measurement of TSP and organic micropollutants (PAHs, dl-PCBs, PCDD/Fs), as illustrate in paragraph 2.3.1.8.

Quartz filters with a diameter of 47 mm were conditioned at 180 °C and then brought to a constant weight by keeping them at a constant temperature and humidity for 48 h inside the conditioning chamber. After sampling, the filtering membranes were conditioned in an oven at 160 °C and brought to constant weight under a controlled humidity and temperature hood (50% relative humidity and 20 °C) integrated with a ionisation system (Climatic Cabinet). The filters were weighed using a robotic system equipped with a microbalance (Sartorius ME 5) with an accuracy of 0,001 mg and compared with a field blank in order to measure TSP concentration.

2.3.1.6 Analysis of metals

Heavy metals were sampled and determined according to the UNI EN 14385:2004 method using the isokinetic conditions provided by the UNI CEN/TS 13649:2015 method. The sampling train consisted of a heated titanium sampling probe equipped with a quartz filter. A system of 4 bubblers, three of which filled with HNO_3/H_2O_2, was inserted downstream of the filter. The analysis of the elements was carried out using inductively coupled plasma spectroscopy with a mass detector (ICP-MS).

2.3.1.7 Analysis of HCl and HF

The measurement was carried out during kinetic particulate sampling and was performed using PTFE filters in order to reduce the reactivity of the gaseous fluoride on the filter present in the probe heated to a temperature of 130°. The determination of hydrochloric acid was performed by applying the UNI EN 1911:2010 bubbler method. For the determination of hydrofluoric acid, the same UNI EN was used.

Figure 19 – HCl and HF sampling

2.3.1.8 Analysis of organic micropollutants

The sampling of emissions of organic micropollutants was carried out according to a single method, which involves adopting the technical standards UNI EN 1948-1:2006 for dibenzo-p-dioxins and dibenzo-p-furans (PCDDs and PCDFs), UNI EN 1948-4:2014 for dioxin-like or dioxin-like polychlorinated biphenyls (dl-PCBs) and ISO 11338-1:2003 (for polycyclic aromatic hydrocarbons - PAHs) according to the filter/condenser method.

The sampling line consists of:

- a heated titanium probe;

- a heated filtration apparatus (thimble filled with quartz wool);

- a vapour condensation system

- a non-condensable vapour collection system (XAD-2 resin).

Before starting the sampling phase, the density of the fumes was calculated and the speed profile of the fumes was studied with the applied method UNI EN ISO 16911-1: 2013, in order to evaluate the parameters required for sampling system to obtain an isokinetic collection of the particulates. Before each sampling, each thimble was labelled with the reference solutions for the verification of the sampling efficiency with analyte standards ^{13}C.

The sampling equipment after the probe consisted of a filter, a condenser and an adsorption cartridge. A titanium sampling probe was used; sampling probe and filter holder were kept at 120 ± 5 °C: these operating conditions are necessary in order to avoid chemical degradation of organic micropollutants. After the filter, water was collected with a condenser at (6 ± 2) °C, and the dried gas was adsorbed in a cartridge filled XAD (poly-styrene/divinylbenzene) microspheres.

At the end of each sampling, the probe was washed using polar and apolar solvents (acetone, methanol and dichloromethane). The leak test was then carried out by inserting the probe into the chimney and waiting for the operating temperatures to be reached. Once the leak test was complete, the filter, XAD-2 and probe wash solutions were collected, referred to as a "field blank" and analysed using the same procedure as for sample analysis.

Sampling times varied depending on the conditions of the combustion process. The sampled matrices (filter, condensate water and XAD-2 adsorbent) were extracted and concentrated for subsequent purification and determination. Specifically, the solid matrices (filters and XAD-2) were extracted in Soxhlet with toluene in the presence of sodium sulphate for a period of 36 h, while the condensates were extracted with 1/10 in volume of the total volume of dichloromethane three times. The extract was added to the washing solvents of the sampling line and then combined in the solid matrix extraction Soxhlet. At the end of this step, the extract was reduced to a small volume (approximately 10 ml). The efficiency of the extraction, purification and enrichment steps was assessed by the addition of PCDD/F and dl-PCB labelled standards. An aliquot (1/10) of the extract was used for PAH analysis, the remainder for PCDD/PCDF and dl-PCB.

The portion for PAH analysis, after addition of perdeuterated internal standards, was purified on a microcolumn packed with silica and alumina. The alkane fraction was separated by elution with hexane; the fraction containing PAHs was then collected by elution with a dichloromethane:hexane mixture (1:1) and analysed by high-resolution gas chromatography coupled to mass spectrometry (HRGC/MS) in SIM (Single Ion Monitoring) mode.

The portion for PCDD/F and PCB analysis was quantitatively transferred to a multilayer chromatographic column containing anhydrous Na2SO4, silica, acid silica and silica containing $AgNO_3$. The sample was eluted with hexane and then concentrated. The eluate was transferred to glass microcolumn containing alumina.

The fraction containing PCBs was collected by eluting with a mixture of hexane and 4% methylene chloride; the fraction containing PCDDs and PCDFs was then collected by eluting with a hexane: methylene chloride mixture (1:1).

Solutions containing PCDDs and PCDFs were concentrated and imaged with mixtures of internal standards, useful for recovery calculations; they were then subjected to analysis by gas chromatography and accurate mass spectrometry (HRGC/AMS and GC/MS Orbitrap) in SIM mode.

2.3.2 Results

2.3.2.1 Gaseous emissions

From Figure 20 to Figure 31 are shown for each day of monitoring and for each sampled pollutant the values of the averages carried out every 10 min relating to the concentrations of the following gaseous macro-pollutants: CO, CO_2, O_2, NO_x, SO_2, TOC.

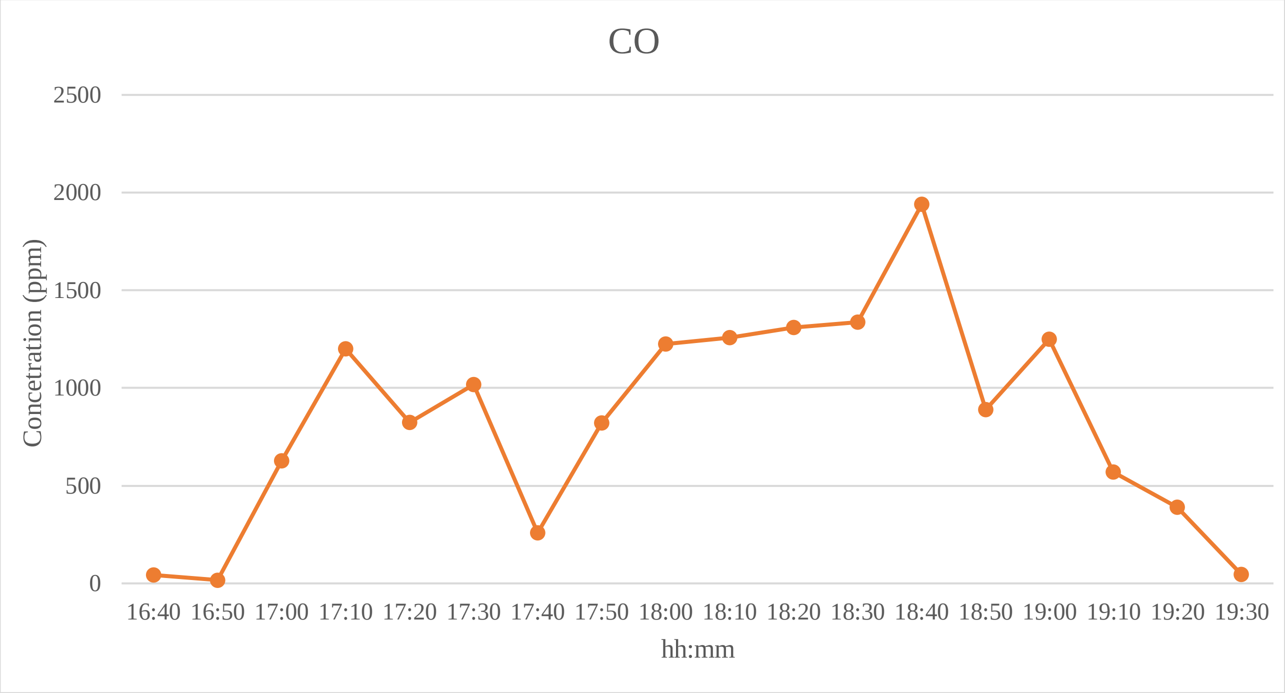


Figure 20 Sampling of 13th April - CO concentrations

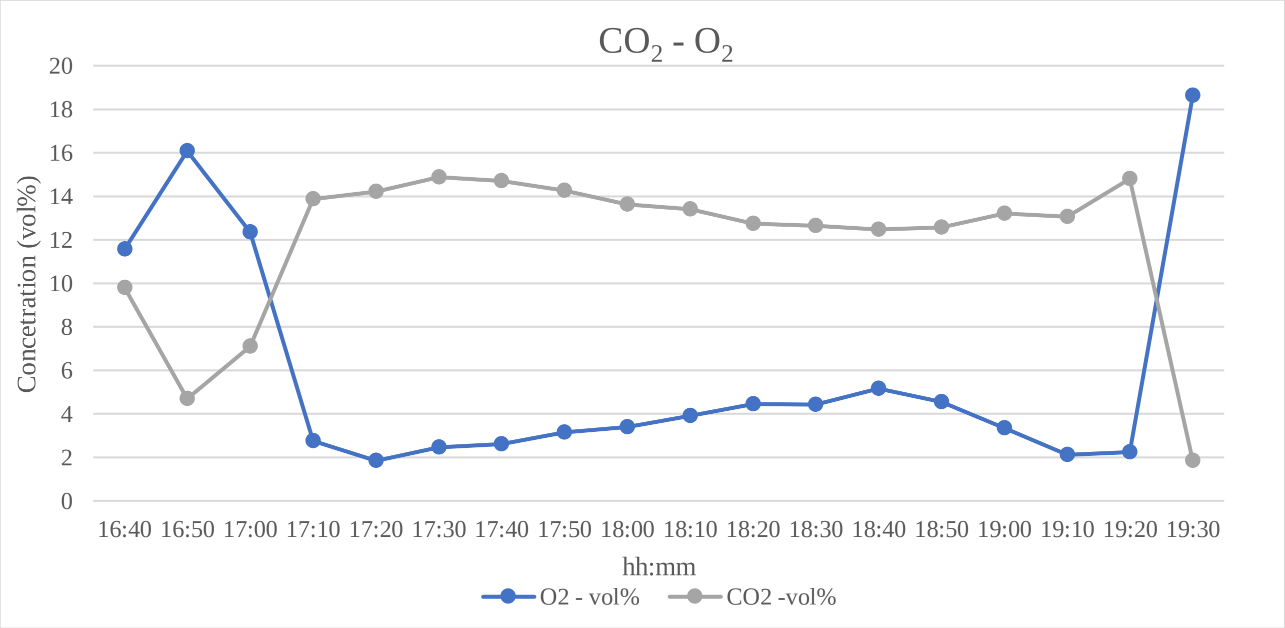


Figure 21 Sampling of 13th April – CO_2 vs O_2 concentrations

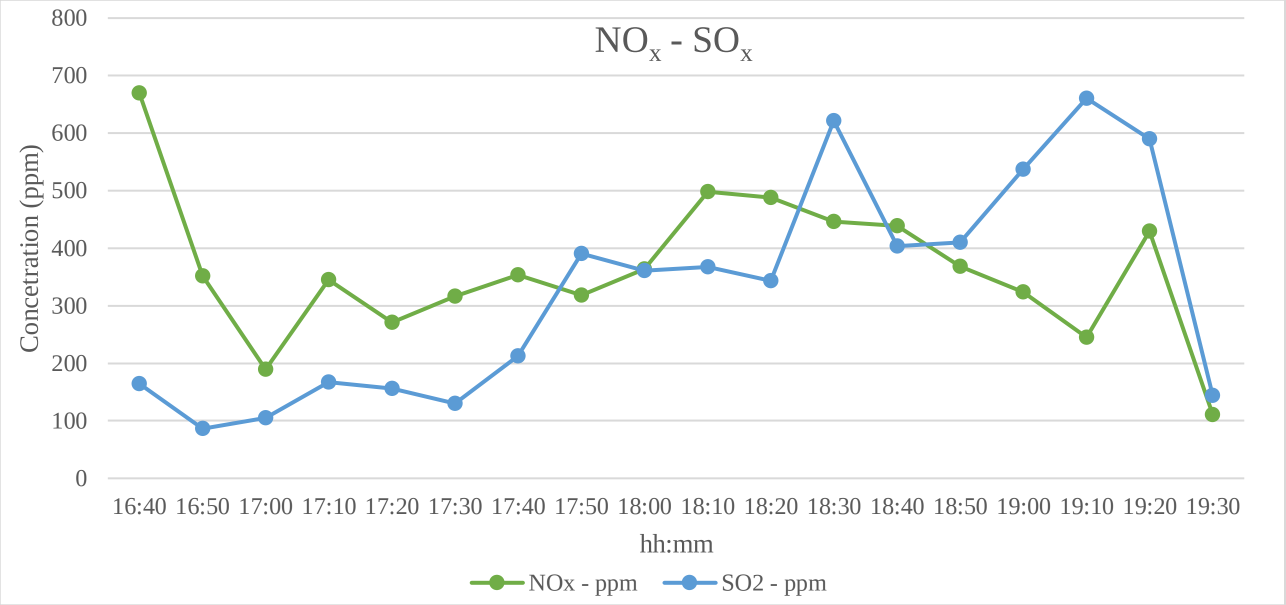


Figure 22 Sampling of 13th April – NO_x and SO_2 concentrations

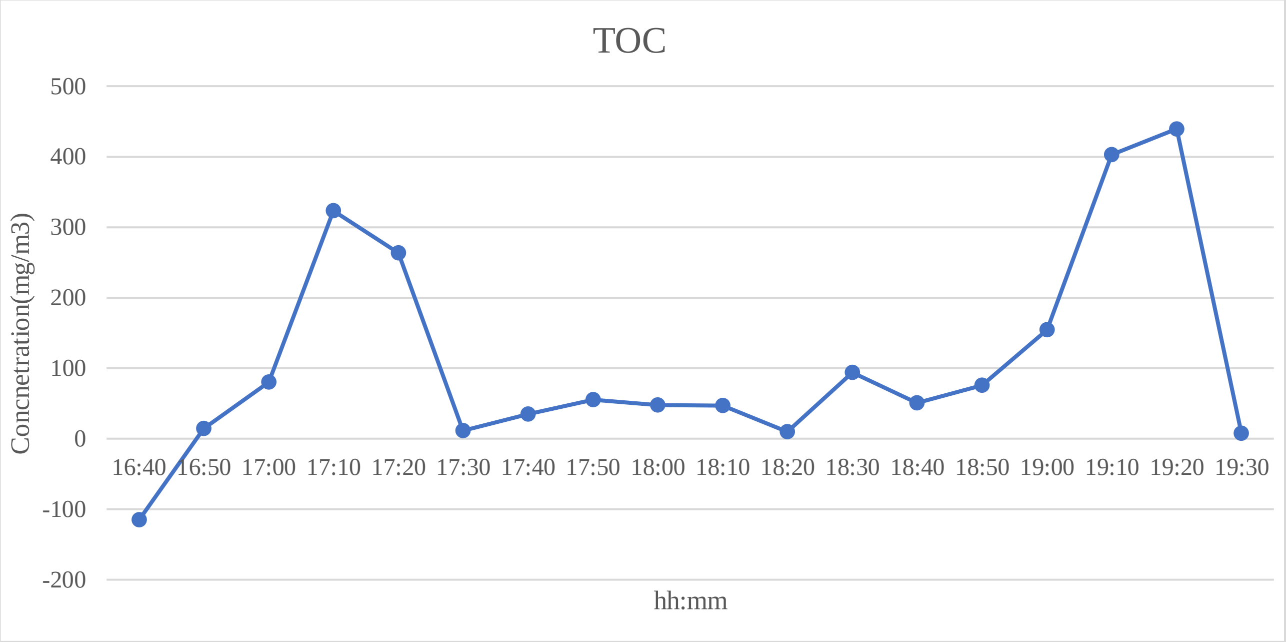


Figure 23 Sampling of 13th April – TOC concentration

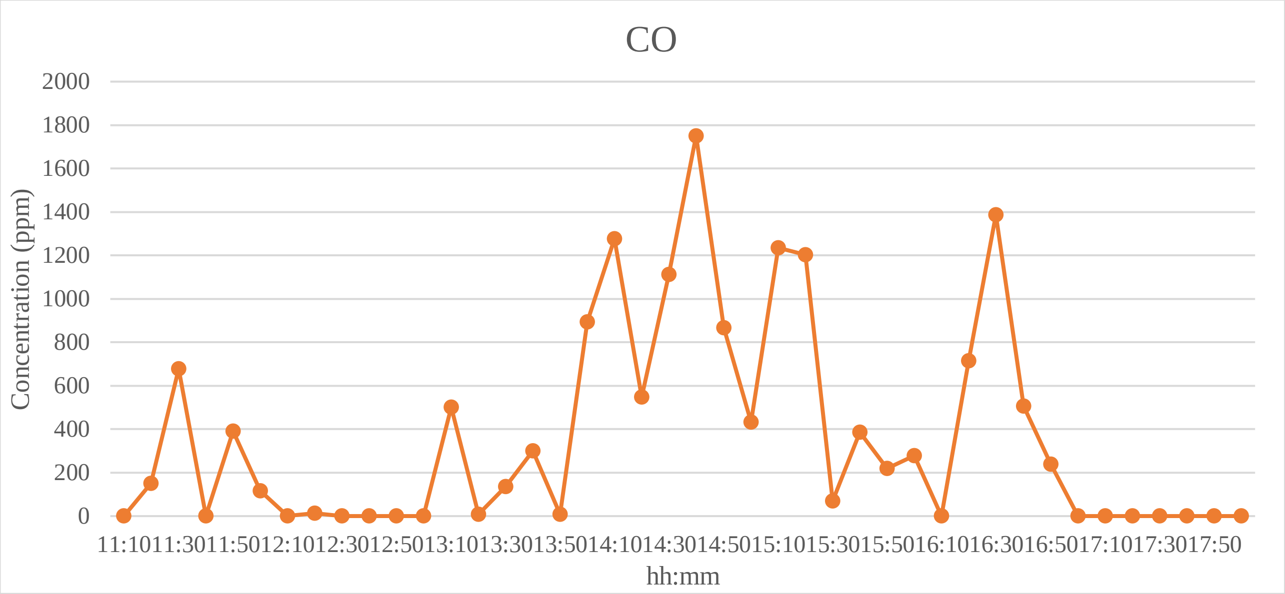


Figure 24 Sampling of 14th April - CO concentrations

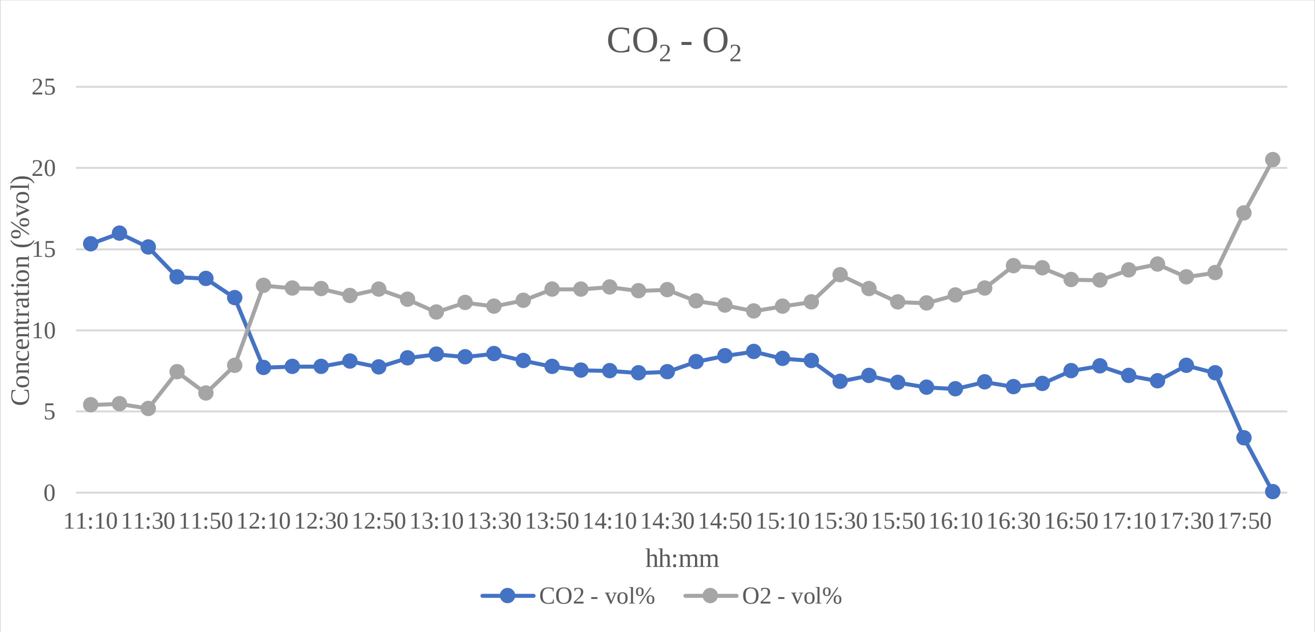


Figure 25 Sampling of 14th April - CO_2 vs O_2 concentrations

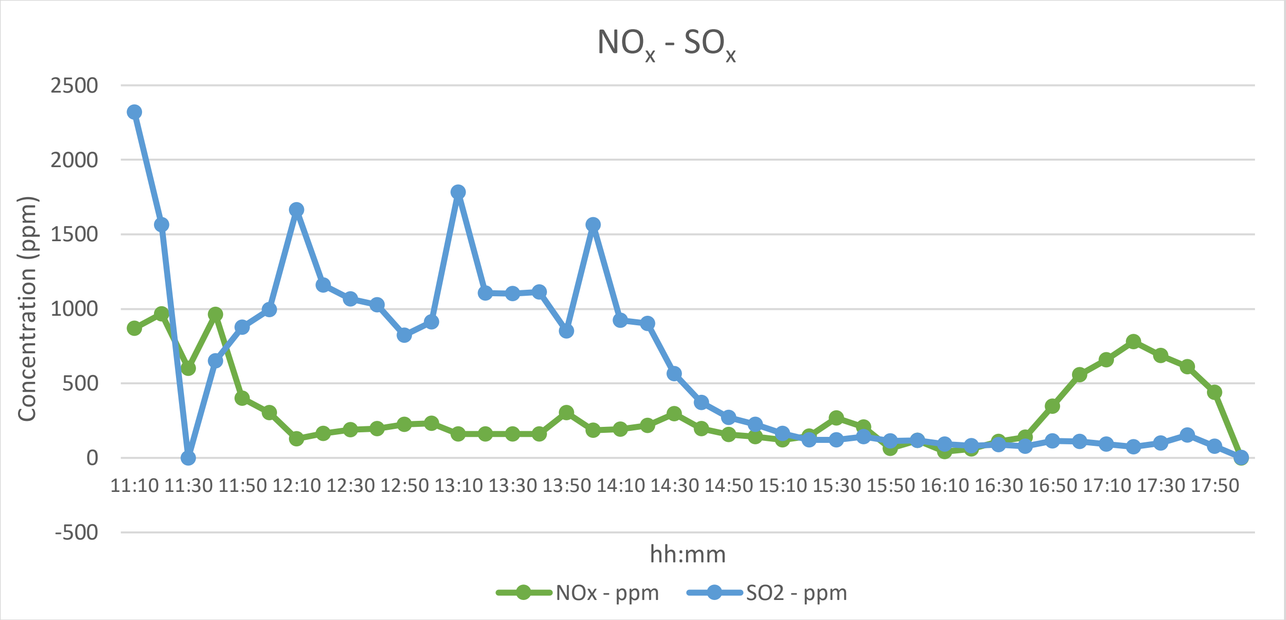


Figure 26 Sampling of 14th April – NO$_x$ and SO$_2$ concentrations

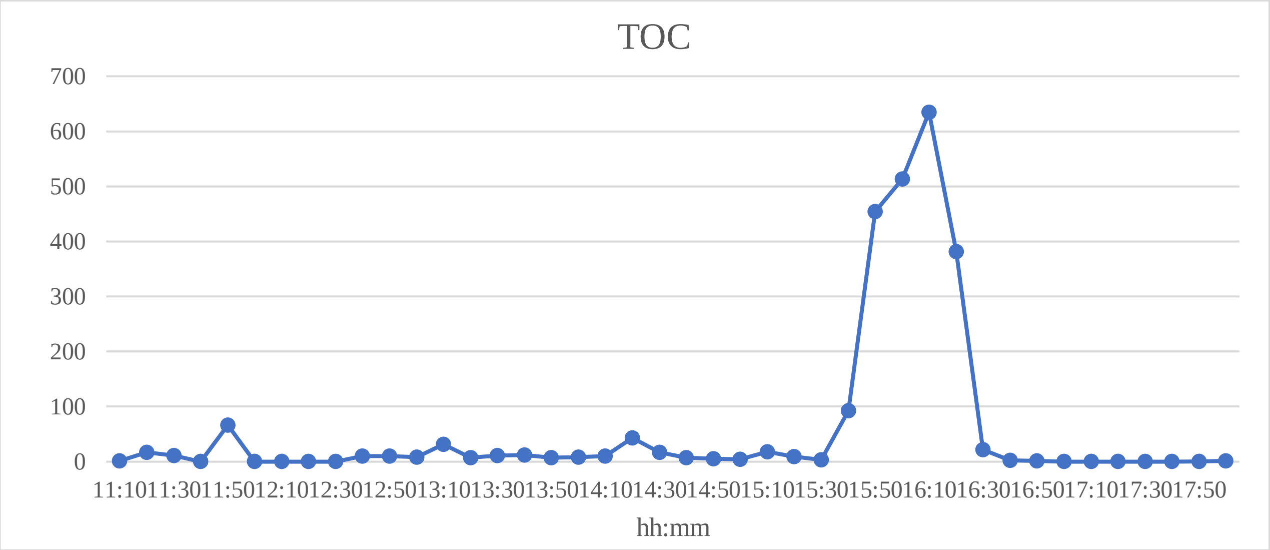


Figure 27 Sampling of 14th April – TOC concentration

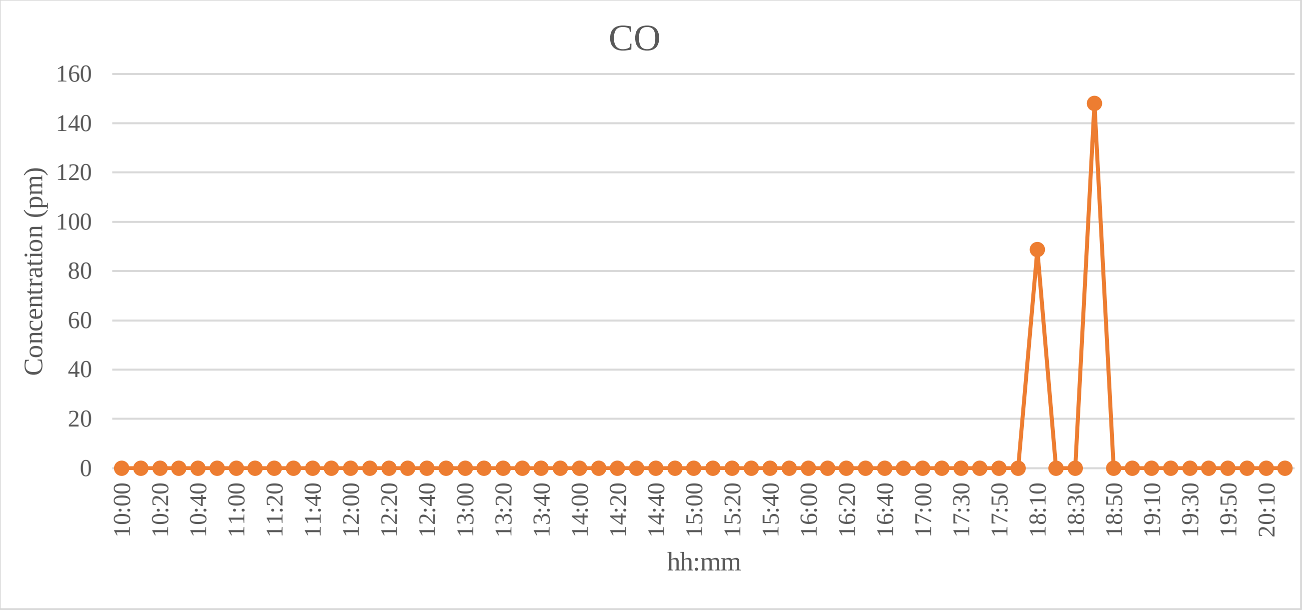


Figure 28 Sampling of 15th April - CO concentrations

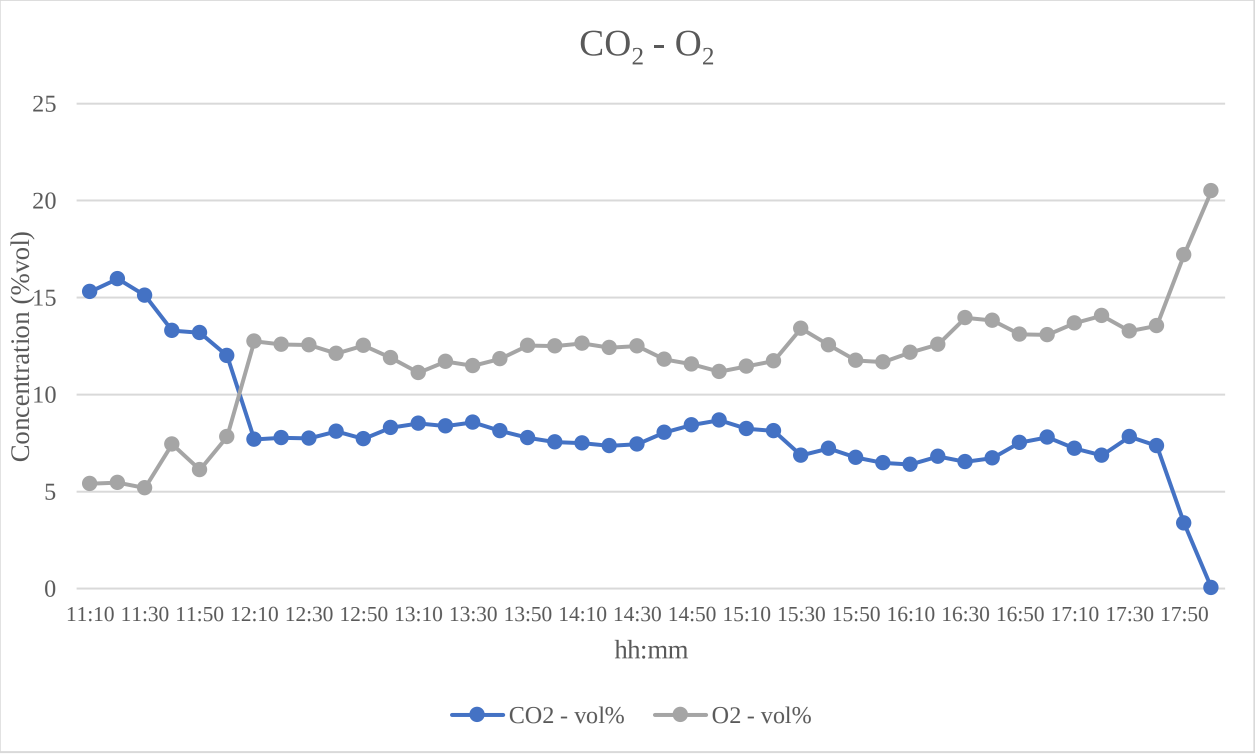


Figure 29 Sampling of 15th April - CO_2 vs O_2 concentrations

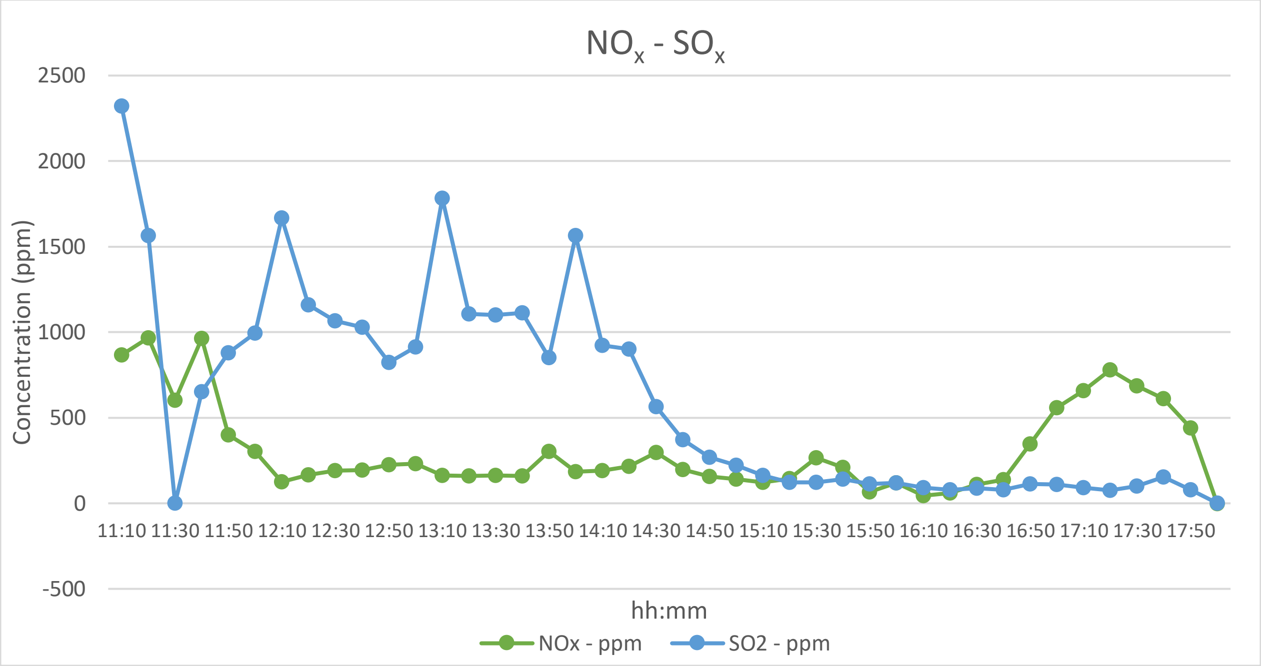


Figure 30 Sampling of 15th April – NOx and SO_2 concentrations

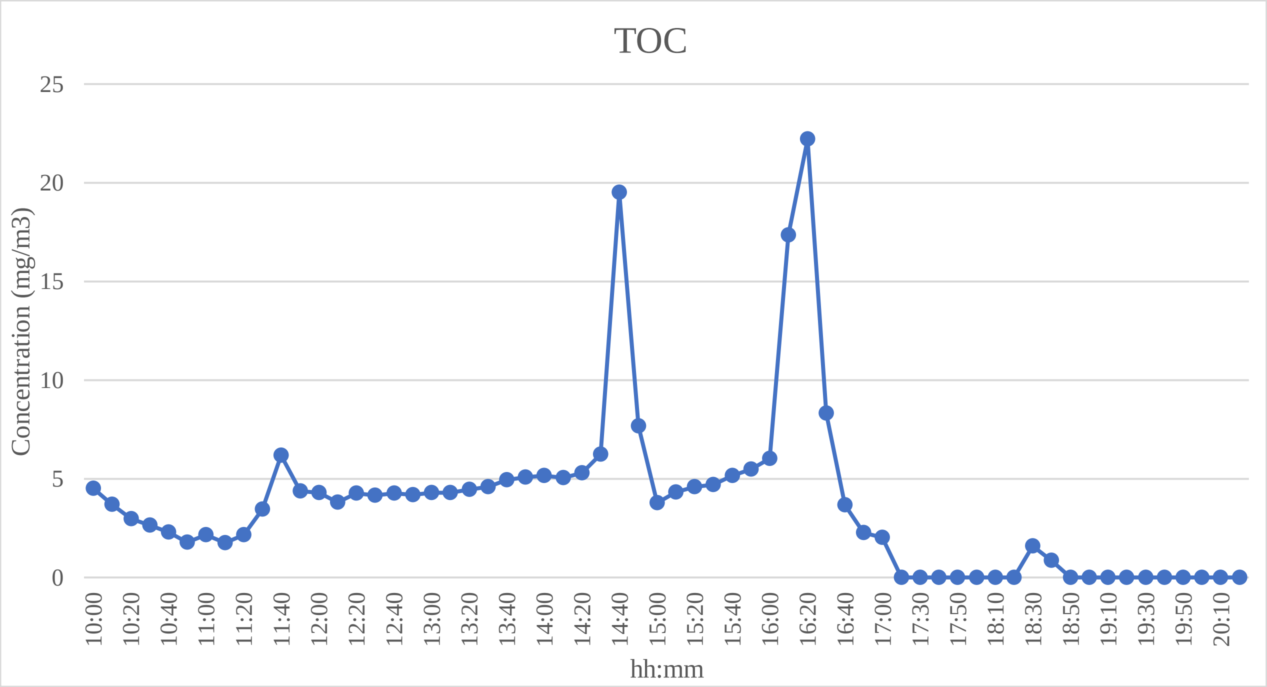


Figure 31 Sampling of 15th April – TOC concentration

The Figure 21, Figure 25 and Figure 29 show carbon monoxide (CO) concentrations are highly variable. The variations are associated both with the combustion of polymers (big bags in all samples, polyurethane in the samples) and partly with thermochemical degradation processes of stone wool/ mineral wool. Also, the TOC values in both the first and second sampling show high standard deviation, indicating poor combustion quality. These emissions could be reduced through the application of primary methods (preventing the generation of flue gases) abatement and if necessary, a secondary method (involving the removal of pollutants resulting from flue gases).

It can be seen that there are emission peaks associated with certain temperatures. For example, CO_2 emissions associated with the thermochemical reactions of the carbonate in the material disposed of in the bag can be seen. At these peaks, there is of course also a decrease in oxygen.

Due to the prototypical nature of the melting furnace, and considering the small size of the chamber which does not allow for uniform and constant loading of the big bags, there are numerous transients. This means that the NO_x and SO_2 concentrations are highly variable depending on the state of loading of the material into the combustion chamber.

This assessment is justified by the values of the daily averages, i.e. the average over the sampling period (days 13, 14 and 15 April) and especially by the high values of the standard deviation (as shown in Table 12).

Table 12 Daily mean values (avg) and standard deviation (std)

13th April	CO (ppm)	CO_2 (vol%)	NO_x (ppm)	O2 (vol%)	SO_2 (ppm)	TOC (mg/Nm^3)
Avg	834.37	11.88	362.57	5.83	325.00	110.84
Std	538.74	3.68	125.33	5.13	188.69	149.05
14th April	**CO (ppm)**	**CO_2 (vol%)**	**NO_x (ppm)**	**O2 (vol%)**	**SO_2 (ppm)**	**TOC (mg/Nm^3)**
Avg	364.36	8.25	307.76	11.87	611.63	56.83
Std	478.99	2.88	255.70	2.85	597.30	148.26
15th April	**CO (ppm)**	**CO_2 (vol%)**	**NO_x (ppm)**	**O2 (vol%)**	**SO_2 (ppm)**	**TOC (mg/Nm^3)**
Avg	3.82	5.80	565.17	15.46	147.05	3.21
Std	23.37	3.96	406.94	3.61	238.08	4.83

As shown in the table above, the values of the daily averages are highly variable on the different sampling days. This observation is corroborated by the test on April 15, 2021. The furnace was not loaded with material for all the previous night and was maintained with a temperature in the combustion chamber of about 1200 °C and about 900 °C in the afterburner. In the first part of the sampling, constant NO_x and SO_2 values were recorded until the material remaining in the loading channel was pushed back into the combustion chamber. In the following hours, a variable concentrations trend was recorded due to the transient phenomena within the combustion chamber that occurred as a result of the introduction of the previously only partially loaded material.

This result is supported by the conclusions of the study by Hart (2001), conclude that the transients are formed due to the semi-batch feed nature of the combustion process and estimate that batch-fed combustion contributes up to 7±18 times more emissions than steady-state combustion will generate.

2.3.2.2 Total suspended particles

In the Table 13 are show the concentration values of total suspended particles expressed in dry mg/Nm^3 and as a function of O_2 reference for the sampling carried out on April 14 and 15, 2021 [1].

The higher concentrations of TSP maybe due to a lot of transient, in particular registered on the day of April 14. For the exception of sampling 3, the all concentrations are above the limit of 10 mg/Nm^3 imposed by "Technical standards and emission limit values for plants waste incineration" of Legislative decree 3 April 2006, n. 152 and subsequent amendments and additions.

Table 13 Total suspended particles values

			Particulate	**O_2 measured**	**particulate O_2 ref.**
date	start hh:mm:ss	stop hh:mm:ss	[mg/Nm^3 dry]	[%]	[mg/Nm^3 dry] O_2 ref.
14/04/2021	12:49:32	13:19:32	348±100	11.76	377±100
15/04/2021	19:27:05	19:57:05	44±13	11.51	46±13
15/04/2021	18:31:10	19:01:10	13±4	7.47	10±4

2.3.2.3 Metals

The Table 14 shows the values of the total concentrations of the main metals expressed as dry mg/Nm^3 and reported as a function of the reference O_2 (O_2 ref. concentration of 11% in v/v). The concentration values for

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[1] Any sampling carried out in the transient periods of the plant was not considered to avoid overestimating the concentrations of airborne particulate emissions from the plant

all three samplings of the following metals are reported: Vanadium, Chromium, Manganese, Cobalt, Nickel, Copper, Arsenic, Cadmium, Antimony, Thallium, Lead.

Table 14 Metals concentrations - O2 ref.

[μg/Nm^3 dry] O_2 ref.

	O_2 % mis.	Vanadium ^{51}V	Chrome ^{52}Cr	Manganese ^{55}Mn	Cobalt ^{59}Co	Nickel ^{60}Ni	Copper ^{65}Cu	Arsenic ^{75}As	Cadmium ^{111}Cd	Antimony ^{121}Sb	Thallium ^{205}Tl	Lead ^{208}Pb
Metals 1	12.04%	46.1	486.6	95.4	6.0	290.9	2,777.6	1,410.0	1,637.0	2,553.3	30.6	39,227.9
Metals 2	14.18%	9.2	186.3	211.8	9.4	318.9	584.7	233.0	2970.7	127.9	10.2	10,604.2
Metals 3	11.62%	4.6	93.4	115.2	12.9	313.2	1,100.0	245.6	715.8	356.4	6.6	5,307.3

High unexpectedly concentrations of Lead and Cadmium have been recorded. In glass and stone wool wastes there are some toxic elements such as Cr, Ba, Ni, Cu, Zn, and Pb (Yliniemi et al., 2021). This could explain the high levels of these metals in the emissions sampled.

According to the Italian legislation - legislative decree of April 2006 no. 152 and subsequent amendments - emission limit values for waste incineration plants are defined. So, on the basis if these limits, the concentrations of metals are above the limits without a complete abatement system.

Table 15 Comparison between legislative decree of April 2006 no. 152 and subsequent amendments and values registered

[mg/Nm^3 dry] O_2 ref.

	O_2 % mis.	Vanadium+Chrome+Manganese+Cobalt+Nickel+Copper+Arsenic+Lead+Antimony	Cadmium+Tallium
Metals 1	12.04 %	46.89	1.67
Metals 2	14.18 %	12.29	2.98
Metals 3	11.62 %	7.55	0.72
Limit value mg/Nm^3	-	0.5	0.05

2.3.2.4 HCI and HF

The emission of HCl is one of the main concerns related to municipal solid waste (MSW) incineration. In particular, the presence of plastics or plastic residues (e.g. PVC) can lead to high contents of organic and inorganic chlorides. HCl and some organic compounds are precursors of polychlorinated dibenzo-p -dioxins and furans (PCDD/F) in municipal solid waste incinerators(H. Zhang et al., 2019).

Some of the industries that use it in production are aluminium industry, oil drilling and refining, the chemical and plastics industries, agricultural and pesticide chemical manufacturers, dye manufacturers, manufacturers of metal parts (NPI, 2021). So, waste containing fluorine can general large quantities of harmful gaseous fluoride, such as HF, that are discharged into the atmosphere during the incineration of this waste (W. han Li et al., 2019).

The following table (Table 16) shows the values of HCl and HF concentrations expressed in dry mg/Nm^3 and as a function of the reference O_2 (O_2 ref. concentration equal to 11% in v/v) for all the samplings carried out.

According to the legislative decree of April 2006 no. 152 and subsequent amendments, the concentrations of hydrochloric acid (HCl) and hydrofluoric acid (HF) are within the limits except for the first monitoring. It should be noted that the plant is not equipped with a complete abatement system, except for the afterburner.

Table 16 Chlorides and Fluorides sampling results

	HCl	**HF**	**HCl**	**HF**	**O_2% measured**
	mg/Nm^3	mg/Nm^3	mg/Nm^3 O_2 ref	mg/Nm^3 O_2 ref	%
Chlorides and Fluorides 1	9.8±1.5	0.87±0.13	10.6±1.5	0.95±0.13	11.8
Chlorides and Fluorides 2	6.1±0.9	0.9±0.14	6.2±0.9	0.92±0.14	11.2
Chlorides and Fluorides 3	3±0.5	0.38±0.06	2.2±0.5	0.28±0.06	7.5

2.3.2.5 Organic micropollutants

The Table 17 shows the concentration values of the PCDD/F compounds expressed as TEQ (pg / Nm^3) and reported as a function of the O_2 reference (O_2 ref. concentration equal to 11% in v/v), and calculated as the sum of the concentrations of the main congeners indicated by current Italian legislation.

Table 17 Concentrations of PCDD/F

Sample	**TEQ (pg/Nm^3)**	**TEQ (pg/Nm^3) O_2 ref.**
DIOX_0458	**45.733**	**29.446**
DIOX_0459	**14.259**	**15.152**
DIOX_0460	**4.261**	**11.293**

Table 18 shows the concentration values of dioxin-like PCB compounds, expressed as TEQ (pg / Nm^3) and as a function of the reference O_2 (O_2 ref. concentration equal to 11% in v/v), calculated as the sum of the concentrations of the main congeners indicated by current Italian legislation.

Table 18 Concentrations of PCB dioxin-like

Sample	**TEQ (pg/Nm^3)**	**TEQ (pg/Nm^3) O_2 ref.**
PCB_0458	**0.2731**	**0.1759**
PCB_0459	**1.5524**	**1.6497**
PCB_0460	**0.4159**	**1.1023**

Finally, Table 19 shows the values of the total concentrations expressed in µg/Nm^3 and as a function of the reference O_2 (O_2 ref. concentration equal to 11% in v/v) calculated as the sum of the chemical species identified in the analysis of the analysed samples. The atmospheric emission limit values for waste incineration plants as defined by Italian legislation were considered as reference for the evaluation of concentrations (Table 21). The average emission limit values obtained set a minimum sampling period of 6 hours and a maximum of 8 hours for organic micropollutants.

The data in the Table 21 show that the sum of the concentrations of dioxins/furans (PCDD + PCDF), dioxin-like PCBs and PAHs are always well below the regulatory limit.

Although the concentrations of PAHs are not of interest for the ducted emissions and therefore are not calculated in the assessment of the final concentration for the purposes of compliance with emission regulations, they displayed high values. In particular, the sum of the PAHs identified in sample IPA_0459 is high due to the high concentrations of Pyrene and Benzo(g,h,i)perylene, both compounds directly correlated to incomplete combustion probably due to the frequent plant transients during the specific sampling, as described in the previous paragraphs.

Table 20 shows only the sum for the chemical species of interest indicated in the Italian regulations in force.

Table 19 Concentrations of PAHs

Sample	**µg/Nm^3**	**µg/Nm^3 O_2 ref.**
IPA_0458	**5.7229**	**3.6847**
IPA_0459	**76.9197**	**81.7354**
IPA_0460	**2.3940**	**6.3444**

The atmospheric emission limit values for waste incineration plants as defined by Italian legislation were considered as reference for the evaluation of concentrations (Table 21). The average emission limit values obtained set a minimum sampling period of 6 hours and a maximum of 8 hours for organic micropollutants.

The data in the Table 21 show that the sum of the concentrations of dioxins/furans (PCDD + PCDF), dioxin-like PCBs and PAHs are always well below the regulatory limit.

Although the concentrations of PAHs are not of interest for the ducted emissions and therefore are not calculated in the assessment of the final concentration for the purposes of compliance with emission regulations, they displayed high values. In particular, the sum of the PAHs identified in sample IPA_0459 is high due to the high concentrations of Pyrene and Benzo(g,h,i)perylene, both compounds directly correlated to incomplete combustion probably due to the frequent plant transients during the specific sampling, as described in the previous paragraphs.

Table 20 Concentrations of single PAHs

Compounds	**Sample**		
	IPA_0458	*IPA_0459*	*IPA_0460*
	µg/Nm^3 O$_2$ ref.	µg/Nm^3 O$_2$ ref.	µg/Nm^3 O$_2$ ref.
Benz[a]antracene	0.0038	0.1063	0.1063
Dibenz[a, h]antracene	0.0199	0.6122	0.3850
Benzo[h]fluorantene	< LOQ	< LOQ	< LOQ
Benzo[j]fluorantene	< LOQ	< LOQ	< LOQ
Benzo[k]fluorantene	0.0024	0.0109	0.0060
Benzo[a]pirene	0.0045	0.3785	0.0564
Dibenzo[a, e]pirene	< LOQ	< LOQ	< LOQ
Dibenzo[a, h]pirene	< LOQ	< LOQ	< LOQ
Dibenzo[a, i]pirene	< LOQ	< LOQ	< LOQ
Dibenzo[a, l]pirene	< LOQ	< LOQ	< LOQ
Indeno [1,2,3 - cd] pirene	0.0064	0.3307	0.0930

Total

	0.0712	**1.4386**	**0.6467**

Table 21 Limit values of $PCDD + PCDF$*, PAHs and PCB dioxin-like emissions*

Compounds	**Limit values**	**units of measurement**
Dioxins e furans (PCDD + PCDF)	**0.1**	**ng/Nm^3**
Polycyclic Aromatic Hydrocarbons (PAHs)	**0.01**	**mg/Nm^3**
PCB dioxin-like	**0.1**	**ng/Nm^3**

2.4 Life Cycle Assessment Analysis

2.4.1 Scope of application

The aim of the Life Cycle Assessment was to evaluate the environmental impacts of the inertisation process of asbestos-cement sheets with compared to landfill disposal.

The study carried out is of the "from Gate to Gate" type, meaning an analysis of the product limited to the intermediate stages of the life cycle, from the entrance to the exit of the plant. Therefore, emissions and environmental production impacts related to the asbestos cement, fuel, use, dismantling and transport phase, as well as the treatment process and products from the secondary raw material were not accounted for.

The function taken as a reference for this study is asbestos-cement waste and the functional unit is 1.000 kg. The reference flow is equal to 749.36 kg of inert material.

2.4.2 Evaluation methods

Life cycle impact assessment (LCIA) calculations were performed in OpenLCA 1.10.3.

For the assessment of environmental impacts, the ecoinvent 3.7.1 consequential database was used.

The following methods were selected for impact assessment, taking into account the different inputs and outputs of the processes.

In detail the methods are:

- ReCiPe EndPoint (E) - A method for assessing impacts by providing results at the midpoint and endpoint of different impact categories;
- TRACI – Method that allows the quantification of stressors that have potential effects, including ozone depletion, global warming, human health effects, ecotoxicity;
- USEtox - Method for measuring impacts on human health.

2.4.3 Life Cycle Inventory Analysis

Inputs and outputs of the two processes have been identified, which consist of raw materials, products, waste and energy consumption both through the use of fuel and electricity. For each process all inputs and outputs from the system have been analysed with their respective quantities.

The flows of the landfilling process have been identified among the processes available in the Ecoinvent inventory, while for the inertisation, the process has been constructed also thanks to the data collected in the monitoring campaign of the laboratory prototype.

The collected data were processed by correlating them with the reference flow of the functional unit.

Table 22 shows the main inputs and outputs for each process.

Table 22 Main input and output of two processes: inertisation and landifill disposal

Inertisation			**Landfill disposal**		
input			**input**		
Flow	**Unit**	**Amount**	**Flow**	**Unit**	**Amount**
Asbestos waste	kg	100	alkyd paint, white, without solvent, in 60% solution state | market for alkyd paint, white, without solvent, in 60% solution state	kg	0.00112
Cement waste	kg	900	anhydrite | market for anhydrite	kg	1.07
electricity, high voltage | electricity production, natural gas, conventional power plant	kWh	26	Asbestos waste	kg	100
Occupation, industrial area	m2*a	1800	cement mortar | market for cement mortar	kg	0.515
waste polyethylene/polypropylene product | treatment of waste polyethylene/polypropylene product, collection for final disposal	kg	2	Cement waste	kg	900
Flow	**Unit**	**Amount**	clay brick | market for clay brick	kg	6.44
output			clay | market for clay	kg	0,22
Arsenic	g	0.002	electricity, medium voltage | market group for electricity, medium voltage	kWh	0.444
asbestos_waste	kg	-100	gravel, crushed | market for gravel, crushed	kg	5.43
Benzo(a)anthracene	g	7.44E-05	packaging film, low density polyethylene | market for packaging film, low density polyethylene	kg	4.27
Benzo(a)pyrene	g	0.00012	plaster mixing | market for plaster mixing	kg	1.59
Benzo(e)pyrene	g	0.00017	sawnwood, softwood, raw, dried (u=10%) | market for sawnwood, softwood, raw, dried (u=10%)	m3	0.0463
Benzo(g,h,i)perylene	g	0.00024	steel, unalloyed | market for steel, unalloyed	kg	98
Benzo(k)fluoranthene	g	3.03E-05	Volume occupied, underground deposit	m3	0.625

Inertisation			**Landfill disposal**		
Cement waste	kg	-900	**Output**		
Chrysene	g	0.000134	**Flow**	**Unit**	**Amount**
Cobalt	g	0.004	Asbestos waste	kg	100
Copper	g	0.1521	Cement waste	kg	900
Dioxins and furans, unspecified	mg	0.00883			
Fluoranthene	g	0.001685			
Indeno(1,2,3-cd)pyrene	g	0.000113			
Inert rock	kg	749.36			
Manganese	g	0.000608			
Nickel	g	0.00059			
Nitrogen dioxide, IT	kg	0.0483			
Particulates	kg	0.0417			
Perylene	g	0.000287			
Polychlorinated biphenyls	mg	0.000236			
Pyrene	g	0.000287			
Sulfur dioxide, IT	kg	0.0444			
Titanium	g	0.0986			

2.4.4 Results

The results of the LCA analyses of the asbestos cement inertisation process are shown below in comparison with disposal in landfills, applying different methods for evaluating the impacts.

A premise must be made about the results produced by the LCA study. It should be noted that the life cycle study evaluates the risk of exposure to asbestos fibres with the USEtox method thanks to the application of the impact factor implemented by Pini et al. (2021). The other methods have been applied to evaluate other categories of impact not considered by the application of USEtox method.

For each evaluation method, the percentage variation for each category is shown graphically in Figure 32, Figure 33, Figure 34. It is evident that the inertisation scenario shows lower environmental impact. In detail, the inertisation reduces the human toxicity - non carcinogenic and total human toxicity applying the USEtox method. The result of ReCiPe method shows that there are also fewer impacts on ecosystems, in particular on water and marine ecosystems. Thanks to the application of TRACi method, a lower impact on human respiratory effects is also shown.

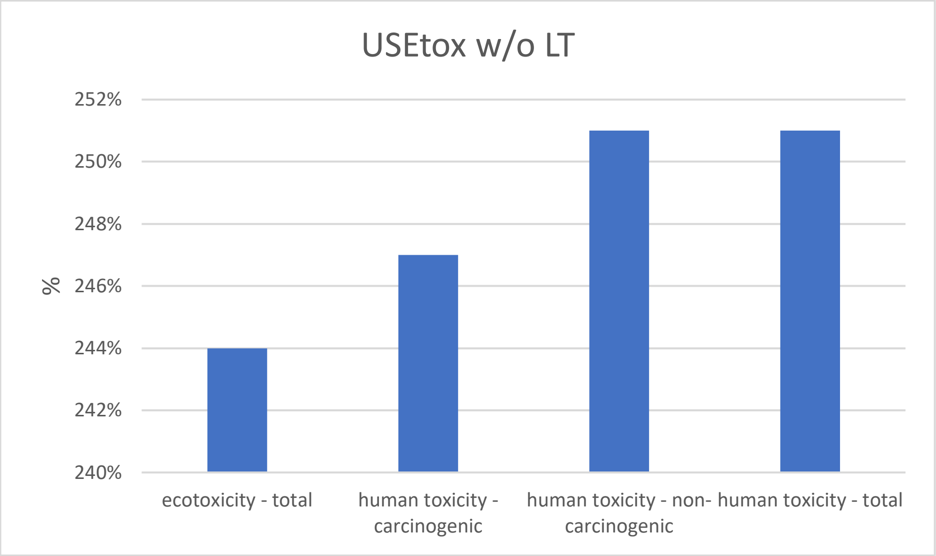


Figure 32 USEtox method: Percentage variation in in each impact category for inertisation compared to the landfill disposal scenario

The previous study (Pini et al., 2021) demonstrates the reduction of human impact and ecotoxicity impact, since it allows the production of an inert secondary raw material. Thus, this study evaluated the emissions of the inertisation process thanks to the monitoring campaign conducted on the laboratory prototype. The campaign demonstrated that no fibres are emitted into the atmosphere and the impact of the emissions of organic and inorganic compounds are contained and guarantee a lower environmental impact compared to landfill disposal.

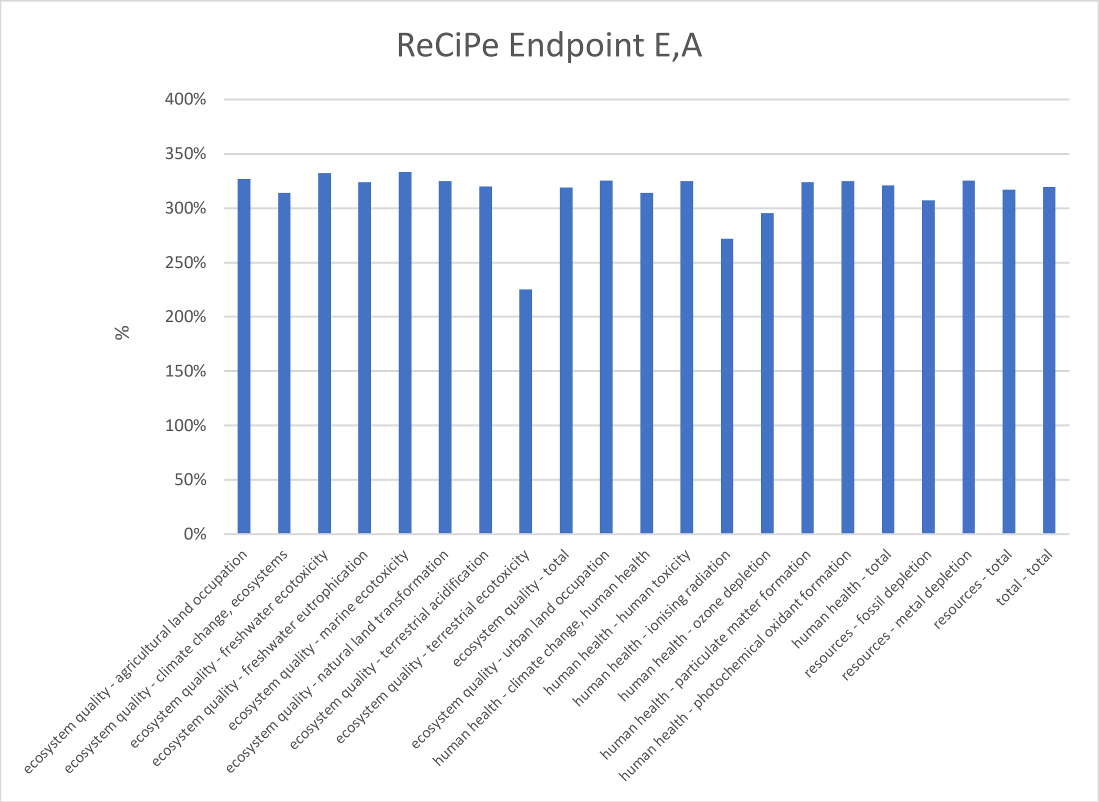


Figure 33 ReCiPe Endpoint (E,A) method: Percentage variation in in each impact category for inertisation compared to the landfill disposal scenario

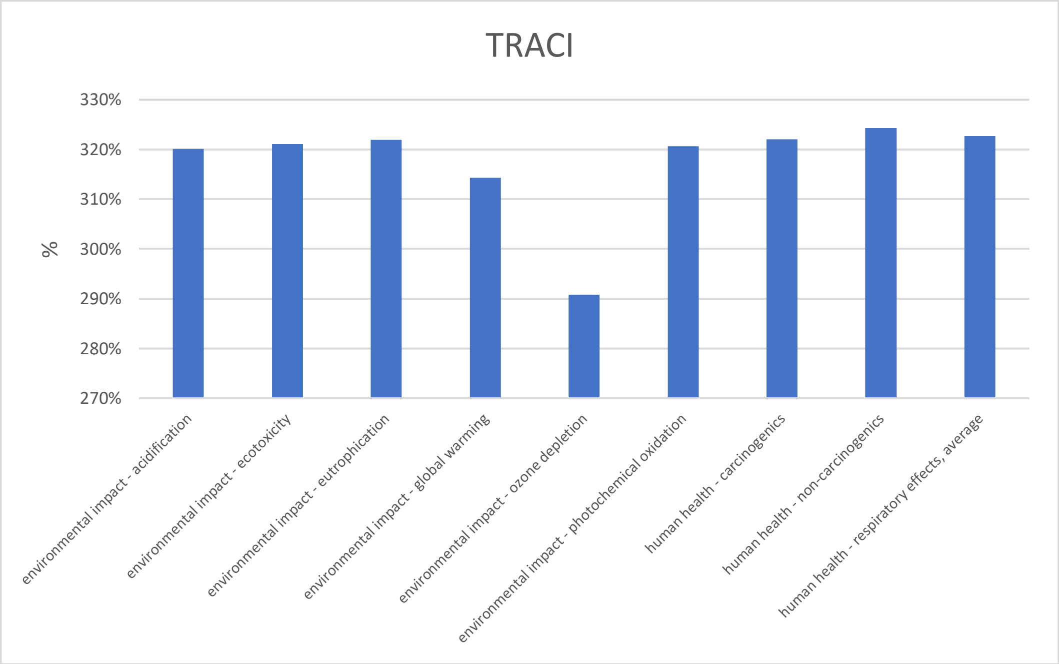


Figure 34 TRACI method: Percentage variation in in each impact category for inertisation compared to the landfill disposal scenario

3 Case study 2: Recycling of post-consumer hard polyolefin based domestic waste

3.1 Case study description

The research of this case study is part of the project PLASMARE (www.plasmare.it), funded by the Italian Ministry of the Environment, Land and Sea Protection, grant protocol RINDEC-2017-00196, current Italian Ministry of Ecological Transition.

The project started with the collection of hard plastic with the support of Lanuvio Municipality. The plastics were first subject to a coarse grinding and then to a finer grinding, thus obtaining flakes consisting of different types of polymers with linear dimensions not exceeding 20 mm. The flakes were then ground again to obtain linear dimensions of less than 5 mm. Before recovery treatments were carried out, the post-consumer mixed plastics flakes were characterised in order to assess their quality, composition, etc. The results were obtained using Fourier Transform Infrared Spectroscopy (FTIR). Using this technique, it was possible to identify the different types of polymers present in the material and any additives. Subsequently, the flakes were subject to separation tests to recover the different polymer fractions.

This case study focuses on the following project phases in order to assess the effect of adding mixed, uncharacterized hard polyolefin based waste to polypropylene plastic blends, and to identify the maximum amount of postconsumer material not affecting the overall properties of the secondary raw material, thus indirectly studying the properties of Italian recycled hard plastic. For this purpose, postconsumer (PC) and post-industrial (PI) polyolefin blends were prepared and characterized in order to assess whether their rheological characteristics were compliant with injection molding or thermoforming requirements. Moreover, an LCA analysis was performed to evaluate the environmental benefits of employing high quality recycled polyolefin blends instead of virgin plastics, aiming for a closed-loop system by recovering polymers that do not need to be down-cycled for reuse. Moreover, an LCA analysis was performed to evaluate the environmental benefits of employing high quality recycled polyolefin blends instead of virgin plastics, aiming for a closed-loop system.

3.2 Materials and methods

3.2.1 Raw Materials and Blends

Mixed, non-preselected post-consumer (PC) polyolefins were manually sorted and gathered from the Lanuvio Municipality (Latium, Italy) and coarsely shredded by Revet S.p.A. PC polyolefins were collected at random, without prior selection regarding provenance or type; therefore, their characteristics showed wide variance between samples due, among other factors, to composition, inclusions, impurities and previous use. Postindustrial (PI) plastics, instead, were discarded waste polyolefins resulting from industrial processes. In this case, polypropylene (PP) was preferentially selected. Both starting materials could not directly undergo extrusion due to granule size; as such, they were further shredded by blade mill (Model 32-20/JL-SL, Tria, Cologno Monzese (MI), Italy) with 2 fixed and 3 rotating blades inside a 320 mm X 200 mm cutting chamber ending with a 5 mm mesh, to reduce sizing and, for PC, to obtain a system characterized by homogeneous composition through dry mixing. The mill engine power input was set to 4 kW, resulting in production rates between 40 and 60 kg/h. No further treatment, such as washing or drying, was applied. For additional homogenization, PC granules were extruded by a counter-rotating twin-screw extruder (Model MD30, Bausano, Turin, Italy), producing three filaments that were subsequently blade milled again to obtain homogenous plastic granules.

The twin counter-rotating screw treatment is a partial solution to the problem of PP and PE incompatibility, often leading to lower mechanical properties than those theoretically expected using the additivity laws (Bertin & Robin, 2002). Figure 35 shows a sample of PC polyolefins before and after extrusion. The temperature profile for the extrusion process is shown in Table 23; low temperatures were chosen for the starting zones, up to 230 °C in the middle and back down to 200 °C at the extruder's die. High temperatures were selected to avoid unmolten parts in the resulting filaments. During preliminary extrusions, feeding rate and extrusion rate were regulated to prevent any sudden pressure or shear stress increases. Pressure was measured upstream of the extrusion die and ranged between 70 and 80 bar during the compounding stage. Shear stress, instead, was kept low by regulating operating current, as higher values could lead to process interruption. Finally, extruder screw speed and feeding screw speed were set at 37 and 62 rpm, respectively. Downstream of the extruder, a 500 µm steel mesh was used to filter out coarse impurities. This filter was chosen as a compromise between high productivity and material homogeneity. Nonetheless, the presence of metal powder, non-polyolefin foreign polymers, paper fibres and other impurities could be excluded, despite subsequent DSC analyses that evidenced melting and crystallization mechanisms typical of PE and PP only, while TGA analyses showed the presence of inorganic residues, similarly to the received shredded material.

Figure 35 Comparison of polyolefins before (left) and after (right) homogenization treatment

Table 23 Temperature profile for the extrusion treatment

Zone	Head	Zone 6	Zone 5	Zone 4	Zone 3	Zone 2	Zone 1
Temperature [°C]	200	220	230	230	220	170	100
		Metering			Compression		Feeding

Several blends were mixed with differing percentages of PC and PI polyolefin. These blends, summarized in Table 24, were dry mixed and extruded as described above to ensure homogeneity, and then injected via injection molding (Micro Injection Molding Machine 10cc, DSM, Geleen, Netherlands) at 230 °C with pressure-time profiles for injection and maintenance, respectively, of 9 bar - 2 s and 9 bar - 12 s, and cooling time of 30 s. Besides filaments, three types of specimens were produced: dog bone flat samples for tensile tests (type 1BA, ISO 527 [30]), impact test samples (IZOD methodology, ISO 180 [31]) and cylindrical samples for hardness tests.

Table 24 Produced blends of post-consumer and post-industrial polyolefins

Blend name	PC Percentage	PI Percentage
PC100-PI0	100	0
PC85-PI15	85	15
PC70-PI30	70	30
PC50-PI50	50	50
PC0-PI100	0	100
PC92-PI8	92	8
PC78-PI22	78	22

The last two blends were produced based on results of a preliminary characterization carried out on the first samples and were subjected to the same testing activity.

Each blend was characterized as described in the next paragraphs, in order to identify the PC/PI ratio giving the best functional performance.

3.2.2 Thermal and Thermogravimetric Analysis

The first type of tests was performed with a differential scanning calorimeter (Q200, TA Instruments, New Castle, DE, USA). A dynamic scan procedure was applied to all the samples, based on three cycles: a preliminary dynamic (temperature ramp) heating from -25 °C to 200 °C (10 °C /min), followed by a cooling phase from 200 °C to -25 °C (10 °C/min) and finally a second heating scan, identical to the first one. Between each ramp, the sample was kept at the final temperature for 3 min. The first heating phase had the aim of erasing the previous thermal history of the sample, so that the cooling and second heating phases could highlight transitions only, depending on the tested material.

Thermogravimetric tests were carried out in saturated nitrogen atmosphere with a heating ramp from 25 to 800 °C (20 °C per minute) in a thermogravimetric balance (Extar TGnDTA 6300, Seiko, Tokyo, Japan).

3.2.3 Melt Index Tests

Melt index tests were carried out following UNI EN ISO 1133 (International Organization for Standardization, 2011), employing an extrusion die with a diameter of 2.095 mm (MFI 452, MP Strumenti, Milan, Italy). Each blend was tested at 230 °C with a pressing weight of 2.16 kg. The extruded samples were cut every 10 s and subsequently weighted over a precision balance (ABT220-5DM, Kern, Balingen, Germany) with a resolution of one tenth of a milligram. At least 10 specimens were collected from each blend and weighted.

3.2.4 Mechanical Tests

Three different tests were carried out. Tensile tests according to ISO 527 (International Organization for Standardization, 2012) were carried out by means of a universal electronic dynamometer (Dyno, Easydur, Varese, Italy), equipped with a 30 KN loading cell (Model 30 K, LLOYD Instruments, Berwyn, PA, USA). The initial gauge length (L_0) was set at 50 mm, with an elongation rate of 50 mm per min. Seven tests per blend were carried out up to sample breaking, discarding the highest and lowest results, as well as a test related to specimens broken in a region excessively close to the fixtures.

Impact tests were carried out on 10 samples per blend. Samples were 80 mm by 10 mm in size with a thickness of 4 mm, and were impacted with a 1,2 J pendulum (Model 15J, Noselab, Nova Milanese, Italy). Fracture occurred in every sample during the test.

Finally, samples of each blend, of 50 mm diameter and 4 mm thickness, were subjected to the Shore hardness test (Manual tool, OMAG, Fanano, Italy), which was carried out on both horizontal surfaces of each sample, in different zones (at least 7 per sample), discarding the highest and lowest values.

3.2.5 Vicat Softening Tests

As the melting transitions of plastic blends are not merely characterized by a narrow temperature range, the softening point was measured by means of the Vicat test, measuring the temperature at which the sample was penetrated by a needle-shaped pin to a specific depth in standardized conditions. Tests were carried out with a heating ramp of 50 °C /h, from room temperature up to the temperature at which the probe could penetrate the sample for 1 mm with an applied load of 5 N (Vicat VH 700, DGTS). Each blend was tested 5 times, following ASTM D 1525 (ASTM, 2001).

3.3 Results

3.3.1 Thermal and Thermogravimetric Analysis

Regarding differential scanning calorimetry tests, as a preliminary investigation pure blend (PC100-PI0 and PC0 PI100) were analysed to characterize base materials and allow more objective comparisons between them. Figure 36 (left) shows the thermogram for pure post-consumer plastic. The diagram highlights the different polymers that can be found in this non-preselected plastic; several melting transitions are clearly visible and may be associated with several compounds. In particular, at a temperature of 106 °C there is a short peak that can be attributed to low-density PE, which has a melting temperature between 105 °C and 115 °C. A higher, narrower and clear peak, probably consistent with the melting transition of a high-density PE, occurs at about 125 °C. The highest peak occurs at 164 °C and can be attributed to the PP. There is also another small peak around 50 °C, indicated as “?” in the figure, which may be due to a low melting point compound or to the evaporation or an evolution of unidentified additives. As an alternative, the described phenomenon may be due to previous treatments or processes, because in the second heating phase, this peak was not detected. Figure 36 (right) shows the thermogram for pure post-industrial plastic. Compared to Figure 36 (left), low temperature transitions are much less evident or absent, with only a neat peak at 166 °C for PP. This is in line with previous results reported in Larsen et al. and Brachet et al., where PE and PP blends of differing compositions showed comparable peaks and behaviour (Brachet et al., 2008; Larsen et al., 2021). These results show the complexity of PC plastic, which is mainly composed of PP, HDPE and LDPE, while PI plastic is mostly homopolymer grade and, to a lesser extent, a co-polymer.

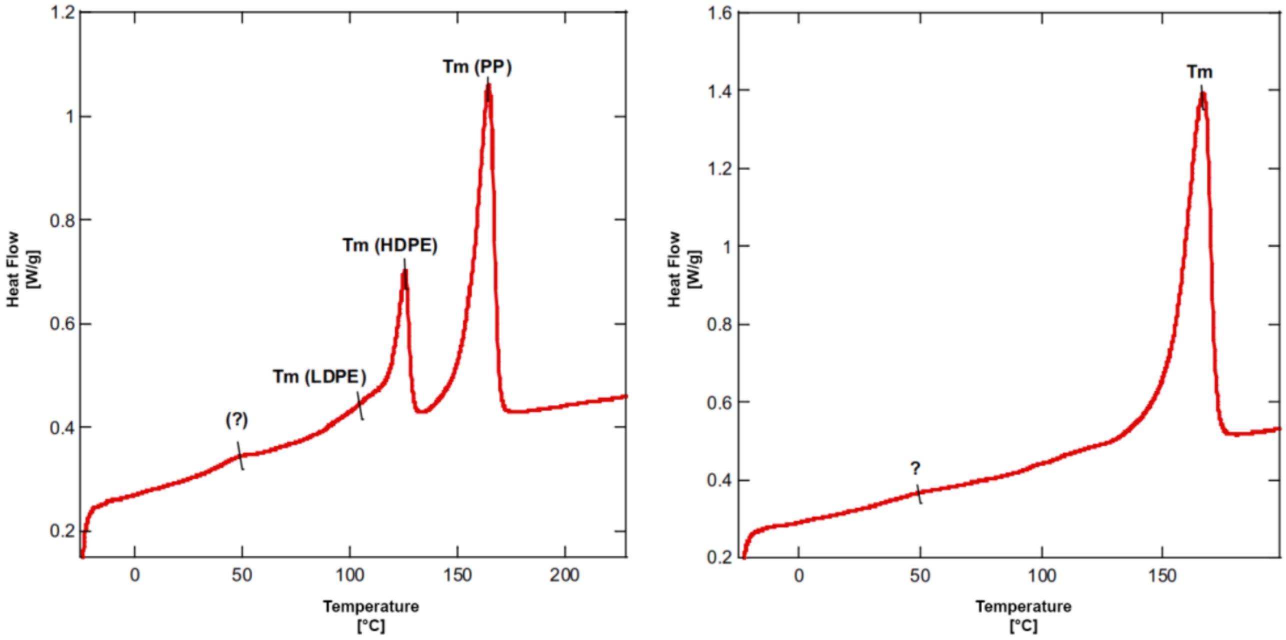


Figure 36 Thermogram of PC100-PI0 blend (left) and of PC0-PI100 blend (right) during the first heating phase

Figure 37 shows thermograms for the first heating phase of all the blends. While it is not impossible to identify a definite temperature trend depending on PC content, there may be a slight increase in PP melting temperature as PI percentage increases, but the described differences are quantitatively in the range of experimental error.

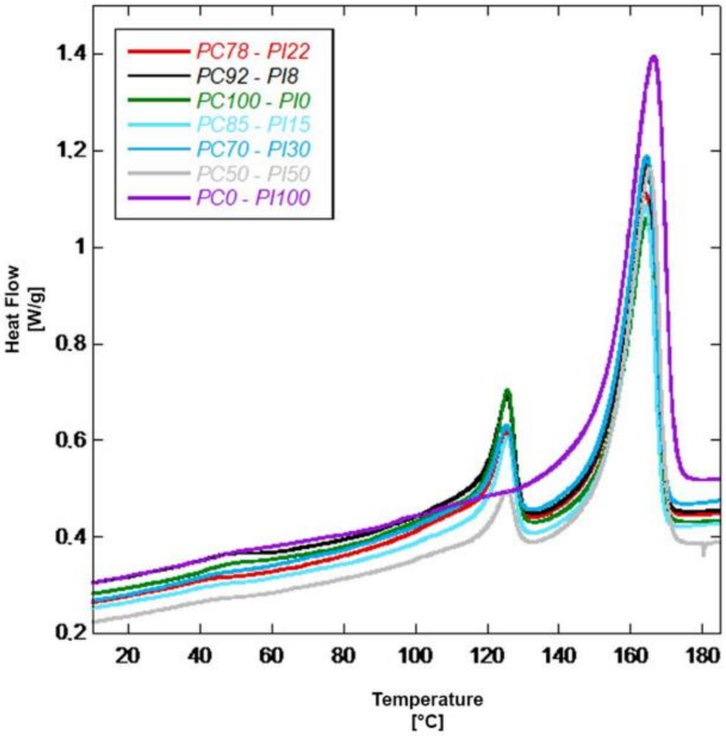


Figure 37 Thermograms of all blends during the first heating phase

Figure 38 shows thermograms for the cooling phase of all the blends. This graph highlights how the recrystallization temperature relative to the polypropylene phase experiences a sharp increase as the percentage of PC material increases, with a maximum for the PC0-PI100 blend. On the contrary, no differences for the recrystallization temperature of PE-based components were measured. A similar behaviour is shown in Aumnate et al. for both PP/LDPE and PP/HDPE blends (Aumnate et al., 2019).

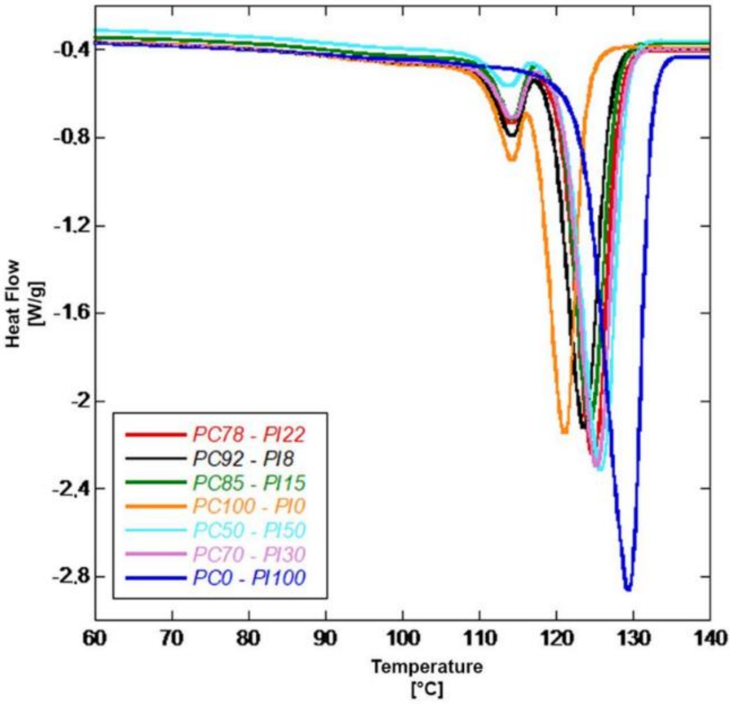


Figure 38 Thermograms of all blends during the cooling phase

The graph for the second heating phase (Figure 39) is very similar to the one obtained in the first heating scan, except for the lack of the slight peak around 50 °C, validating the formulated hypothesis: it is probably the result of a molecular, slightly ordered arrangement, or minor phase, induced by the extrusion process, which is completely removed by the first heating treatment (scan). Table 25 summarizes melting enthalpies for each blend detected in the three scanning phases carried out. Maximum values correspond to pure post-industrial plastic, confirming temperature results shown in the previous graphs.

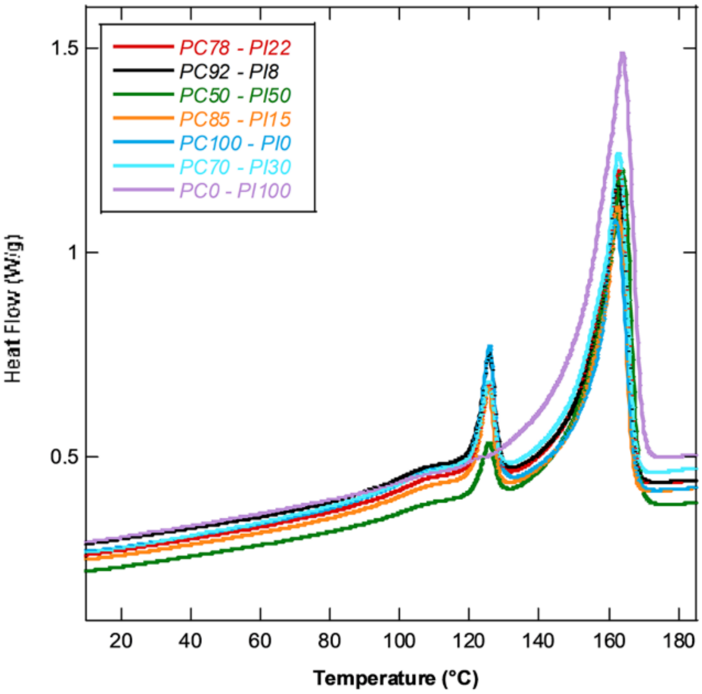


Figure 39 Thermograms of all blends during the second heating phase

Table 25 Enthalpy of fusion for each blend measured in the heating and cooling phases

Blend	First Heating Phase ΔHm [J/g]	Cooling Phase ΔHm [J/g]	Second Heating Phase ΔHm [J/g]
PC100-PI0	68.6 ± 1.6	67.7 ± 2.2	69.7 ± 3.1
PC92-PI8	67.1 ± 2.4	67.1 ± 1.3	61.3 ± 0.6
PC85-PI15	55.6 ± 2.5	68.7 ± 2.4	58.7 ± 1.3
PC78-PI22	65.5 ± 1	66.2 ± 1.4	61.9 ± 1.3
PC70-PI30	60.1 ± 2.8	57.9 ± 3.5	60 ± 3.6
PC50-PI50	57.7 ± 5.5	62.8 ± 3.3	52.6 ± 4.7
PC0-PI100	75.4 ± 1.9	91.1 ± 0.6	84.4 ± 0.1

These values were directly calculated via the Universal Analysis 2000 software, which the measurement apparatus was equipped with. These experimental procedures, together with the TGA measurements, were carried out with the aim to identify the main polymers included in the blend, as well as the related inert additives, and also to provide a process framework in terms of temperatures for potential industrial end-users. Through the DSC analysis, the lower temperature threshold where compound melting occurs was measured, while through the TGA analysis, the higher limit was obtained, to indicate the processing conditions that would prevent any significant degradation phenomena in the investigated material.

As for DTG analysis, Figure 40 shows the trend for all the blends, highlighting an interesting phenomenon around 160 °C; the analysis identified a sudden and unexpected weight loss followed by a quick weight gain to almost the starting values. Such a phenomenon was particularly marked for PC70-PI30 and could be observed for each blend, whereas it did not occur for PC0-PI100. This was possibly due to some unidentified compound, contained in small quantities in random samples of PC plastic, whose thermal degradation led to the formation of gasses that almost immediately or quickly evolved outside of the plastic matrix, causing the crucible to vibrate and shake together with its content, temporarily perturbing the measure.

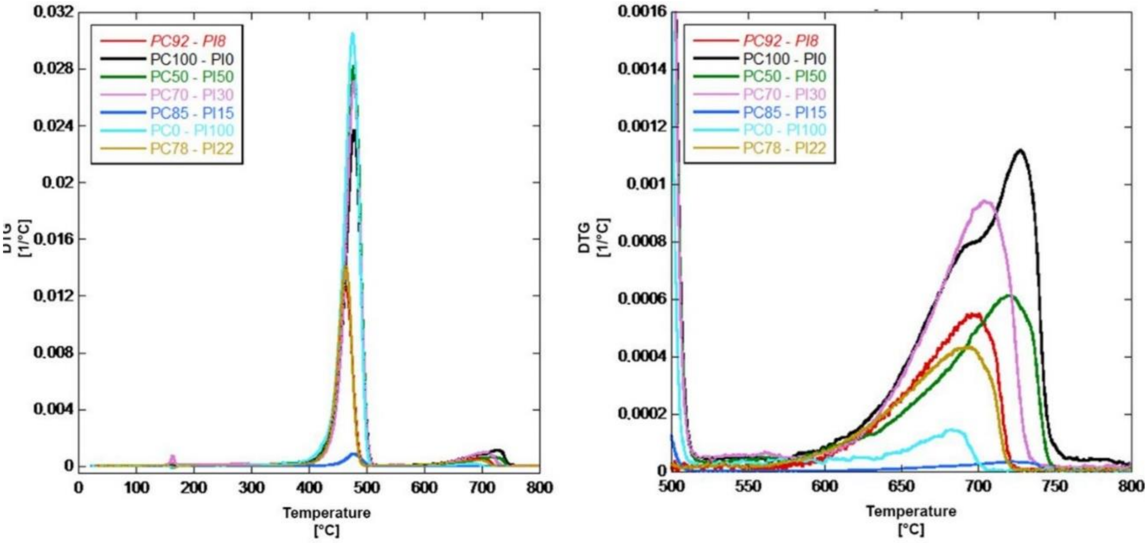


Figure 40 Weight loss rate versus temperature for all blends (left), with a detail for high temperatures (right)

It is evident from Figure 40 that there were two distinct degradation mechanisms, the main one ending around at 500 °C, and the second occurring in the temperature range between 650 and 750 °C. The first mechanism involved the degradation of polyolefins (PP and PE) found in the blends and exhibiting similar degradation behaviors. The second mechanism was instead related to a large amount of inclusions, such as carbon black in very small quantities for PC0-PI100, or calcium carbonate, dyes and inert components for PC plastics. The figure also shows a detail of this degradation mechanism, highlighting how there were actually two degradation rates that may be associated with different inclusion families.

Figure 41 shows the trend of weight loss versus temperature. It shows that no degradation effects occurred below a temperature of 400 °C. Beyond this threshold, a significant weight loss occurred between the range of the mentioned lower limit and about 500 °C, accordingly to the framework depicted by the diagram in Figure 40.

No further weight losses occurred up to a temperature of about 645 ºC, while between this temperature and 715 °C, another degradation reaction occurred, which was attributed to the fillers included in the investigated systems. It is also evident that the amount of residues grew as PC content increased, going from less than 1% for PC0-PI100 to 11% for PC92-PI8, similarly to the system based on the pure PC.

As previously noted, other than information on the polymers and fillers included in the investigated compounds, these analyses could also suggest a process frame for future industrialization. About this, the processing temperature, both in the compounding stage and the injection molding stage, could be chosen within the range from 190 °C to 360 °C, with the aim of obtaining the best compromise between material performance or quality and productivity. The extreme values of the temperature range have to be avoided: the upper limit to prevent degradation induced by shear stresses induced by additional heating, and the lower one to optimize the material flowing ability.

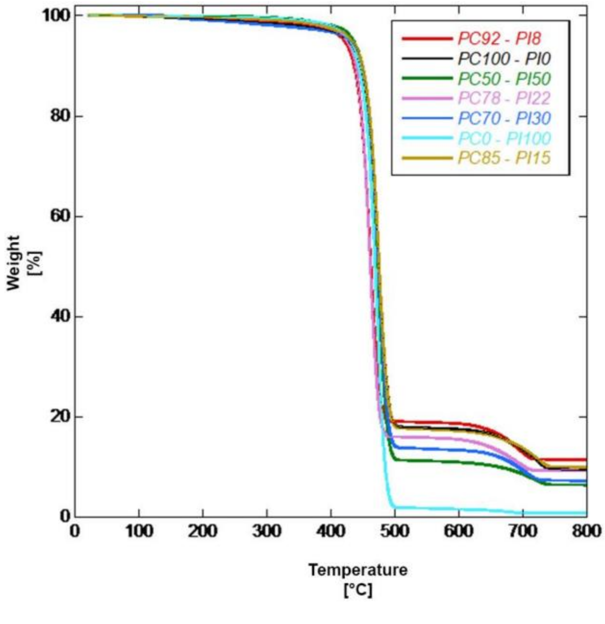


Figure 41 Weight loss versus temperature for all blends

3.3.2 Melt Index Tests

Figure 42 shows results for the melt index tests and highlights how this value is very low for blends characterized by a high percentage of PC, with a minimum for the PC85-PI15 blend. Low values for blends completely or mostly made of PC can partially be attributed to the presence of HDPE as contaminant and partially due to the predominance of relatively long-chain co-polymers. The highest value was obtained for the PC0-PI100 blend (21.5 ± 0.01 g/10 min), which was quite close to the result for PC50-PI50 (21.00 ± 1.60 g/10 min). As evidenced by these results, melt flow index increased as the percentage of PI PP increased, although the obtained trend was not linear. The MFI value roughly increased as the PI percentage increased up to 50% in weight; with higher PI content, a constant value was obtained for MFI, although some deviation from the expected behaviour occurred as the blend containing 18% in weight of PI was unexpectedly characterized by the lowest value of MFI. As this blend was the result of material collected from a different batch in comparison to the one used for the first five blends, these results could have been affected by a less homogeneous composition and, consequently, by the presence of a higher amount of inert materials, fillers and inclusions. This behaviour is quite common in low performance or general purpose hard plastic materials and is also in line with previous literature focused on recycled PC polyolefins, where the likely traces of other polymers with a higher liquefaction temperature can obstruct the capillary (Hubo et al., 2015). Moreover, MFI values for PC polyolefins were lower than values for plastic packaging recycled polyolefins found in the literature; this may have been due to lower requirements in originating items, as bulkier objects require lower melting capabilities during molding, or to a prevalence of blow molded objects.

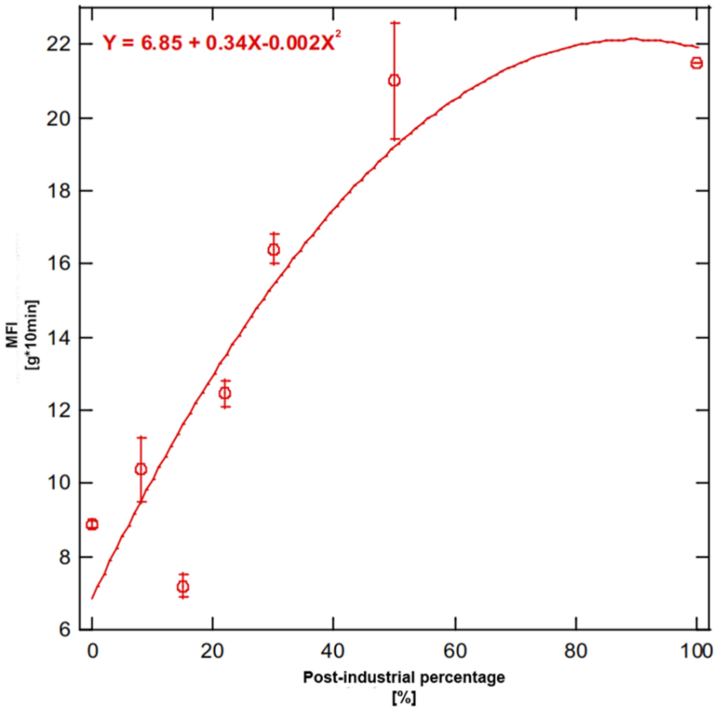


Figure 42 Melt flow index for each blend ordered by post-industrial plastics percentage.

In general, the MFI of the blend proportionally increased with the amount of PI, as the latter was characterized by a lower viscosity.

It can be hypothesized that when the percentage of PI is lower than 50% the two systems are sufficiently miscible and quite finely dispersed phases are formed, as the rule of mixture is approximately matched. This is a proportional rule, which states that when two substances are mixed, the property of the mixture is a ratio of the properties of each substance, weighted by the proportion of each substance in the mixture (Hubo et al., 2016). This law is given in Equation 1 with X_i the property and V_i the volume fraction of the i^{th} constituent material.

$$X = X_1V_1 + X_2V_2$$

Equation 1

The deviation from the linear behavior for MFI for blends with a higher amount of PI was probably due to the formation of two or more coarse and separate phases, related to each system included in the blend (two different kinds of polyethylene, both HDPE and LDPE, as well as polypropylene).

On the basis of these results, it may be safely claimed that blends with at least 22% in PI PP can be recommended for injection molding. Compared to the same test performed on plastic bottles, there was a substantial inversion of the trend, with lower values for pure PC mixed polyolefin and higher values for pure PI PP. In any case, melt flow index values were generally much higher for hard plastic, which is generally injection molded, than for bottle plastics, which are usually subjected to blow molding processes (Curtzwiler et al., 2019).

3.3.3 Mechanical Tests

Three types of mechanical tests were carried out: tensile, impact and Shore hardness test. Table S 1 in Supplementary Materials reports values for tensile properties. Figure 43 shows the averaged results of the ensile tests. From the plots it is evident that post- consumer material showed a clear plastic deformation pattern. The average value of strain at break ε_{br} was 140±90%, and the experienced high standard deviation depends on the fact that samples for PC100-PI0 behaved in very different ways, some breaking at low strain and some at high strain levels, probably due to an incomplete homogenization of polypropylene types even after the whole processing stage of extrusion and injection molding. Figure 35 shows different colours for the starting material, thus different sources and reasonably a different composition. A similarly large standard deviation was reported for PC plastic with non-homogenous, mixed polyolefin composition, underlining the high variability of this type of sample (Luijsterburg & Goossens, 2014). There was a marked reduction in the plastic strain in blends containing post-industrial plastic, up to a complete lack of this plastic feature, as in the system PC0-PI100. A similar, although less marked, behaviour was reported for PP/LDPE and PP/HDPE blends, where maximum strain and plastic behaviour increased as PP content decreased (Aumnate et al., 2019).

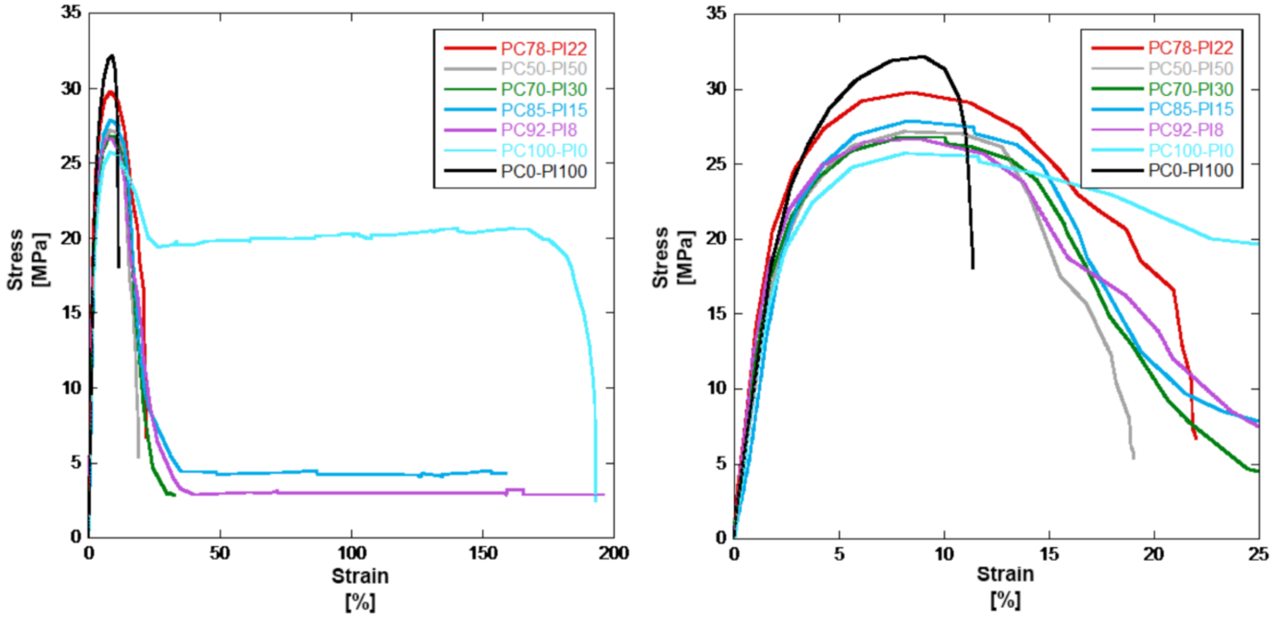


Figure 43 Stress-strain graph for all blends (left) and detail at low strain percentage (right).

Figure 44 shows the strain at break versus post-industrial content, in which the behaviour of post-consumer plastic was confirmed. In this case, standard deviation was high for post-consumer plastic because of the difficulties of obtaining a homogeneous system in terms of composition and, more importantly, in filler content.

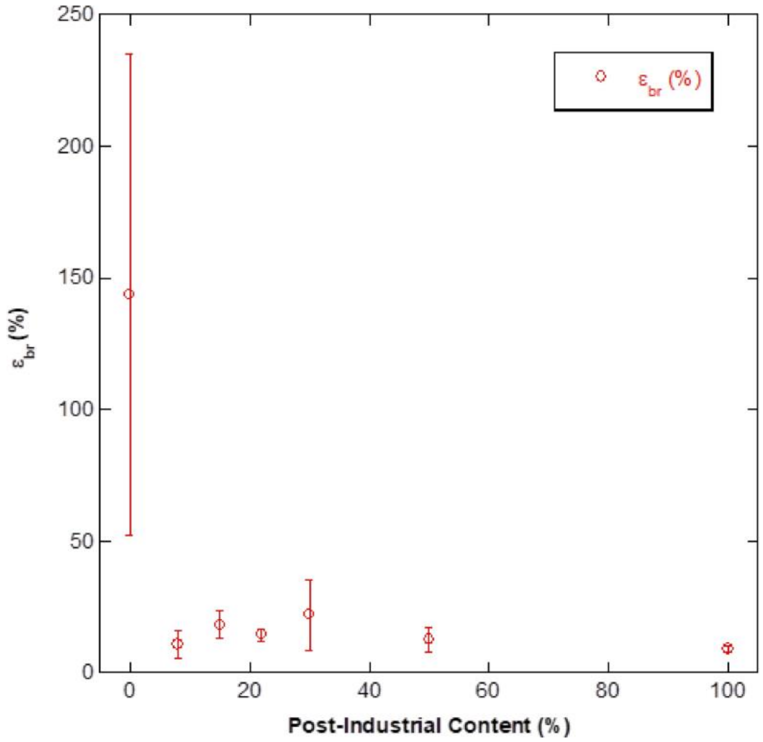


Figure 44 Strain to breaking point versus content of post-industrial plastic

It was also found that the lowest value of yield strength was measured for the system based on the post-consumer plastic, while the highest was measured for pure post- industrial polypropylene. Small differences occurred in the value of the yield strength for the intermediate blends, probably due to the difficulties of obtaining homogenous materials. In any event, it was possible to observe a slight yield strength increase with post-industrial content percentages up to 22% PI content, which subsequently decreased for higher PI content. Values for low PI content blends were below 8,4%, while the pure PI blend reached 8,7%. These results are more or less comparable to those obtained for bottle plastic, as the range was between 5 and 9%. The exception was the much larger standard deviation for hard plastic samples, probably a result of higher inhomogeneity (Curtzwiler et al., 2019).

It can be concluded that in terms of static mechanical properties the blends exhibited similar performance, which was higher than the pure PC system and lower than the PI blend.

Impact strength tests were again characterized by a high standard deviation, partially explaining the apparently random trend found for low PI content plastic. Table S 2 in Supplementary Materials summarizes impact strength results. Analogous results were found in the literature (Gu et al., 2016). Similarly, to the tensile strength, PC0-PI100 and PC78-PI22 showed the best results, at 4,1 KJ/mm^2. This is in line with results from (Curtzwiler et al., 2019), who showed a simi- lar approach with recycled beverage bottle enclosure, concluding that blends containing different amounts of recycled polyolefin may be engineered to obtain desired properties.

The trend for Shore hardness (Figure 45) showed a direct increase in hardness corresponding to an increase in PI content, as a direct consequence of an increase in pure polypropylene content.

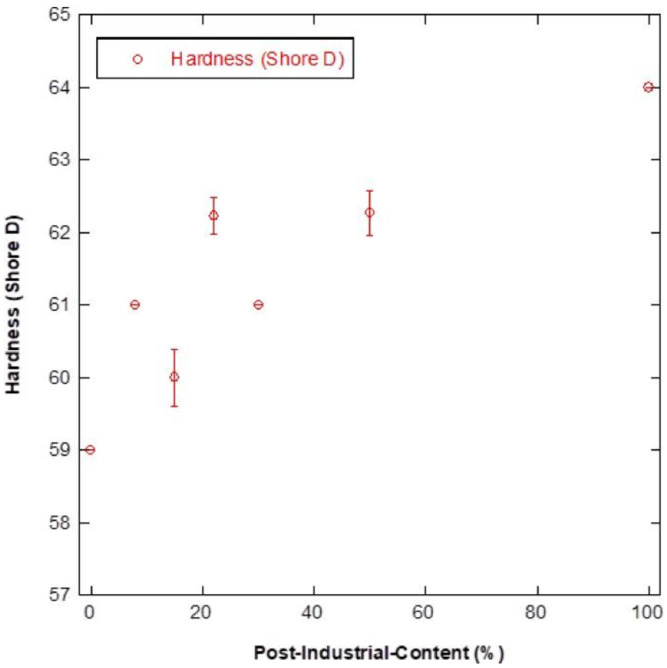


Figure 45 Trend for Shore hardness versus post-industrial content.

3.3.4 Vicat Softening Tests

The Vicat trend (Figure 46) was quite linear, increasing directly as PI plastic content increased, and, consequently, as PE content decreased, except for PC92-PI8, which behaved in an anomalous manner, as the related value for the measured property was quite similar to the value measured for pure PC.

For systems characterized by a low amount of PI (up to 18%), the Vicat softening variation was lower than 3% in comparison to pure PC. On the contrary, for higher weight fractions of PI, that is, beyond the mentioned threshold, more significant variation occurred, as the softening temperature increased by 0,2 °C for each 1% increase in PI content.

The behavior of the material rich in PC polymer could be explained again by the difficulty of obtaining homogeneous material with lab scale batches and apparatus. Probably, this issue can be significantly improved in the case of industrial production, which is based on high production volumes, including the volume reduction and dry mixing stages and the extrusion process.

The presence of non-polyolefin-based inclusions, rubbers and fillers contributes to the difficulty of obtaining a homogeneous system, characterized by constant properties.

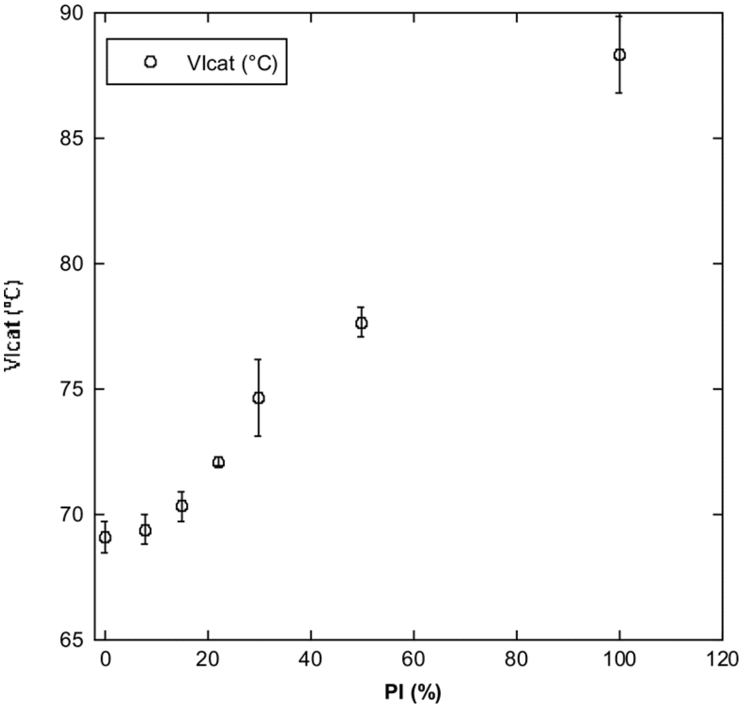


Figure 46 Vicat softening temperature versus post-industrial plastic content

3.4 Life Cycle Assessment Analysis

3.4.1 Scope of application

The goal of the LCA performed during the course of this study was to compare the new recycling process combining post-consumer plastic and post-industrial residues with the production of virgin high-density polyethylene. The functional unit chosen for this comparison was 1 kg of plastic material ready to be used in industrial processes. As this was a preliminary assessment of a yet to be industrialized process, there were no clear geographical boundaries for this analysis. Nonetheless, where possible, European datasets were selected.

The assessed recycling scenario considered the most promising blend according to tests and analyses carried out and described here. The post-consumer plastic production chain was modelled according to experimental processes employed at laboratory scale, with a hypothesized scale-up to industrial scale. Post-industrial plastic, or high-quality plastic residues from the industrial production of plastic objects, is considered equivalent to virgin plastic; plastic residues are generally recovered and re-employed in industrial processes, and as such, their use in the modelled recycling production chain cannot be considered as recycling. Moreover, transport was not considered or modelled, as both production chains might be considered equivalent in that regard. Figure 47 shows the assessed recycling process. Water and energy inputs are not shown in the figure.

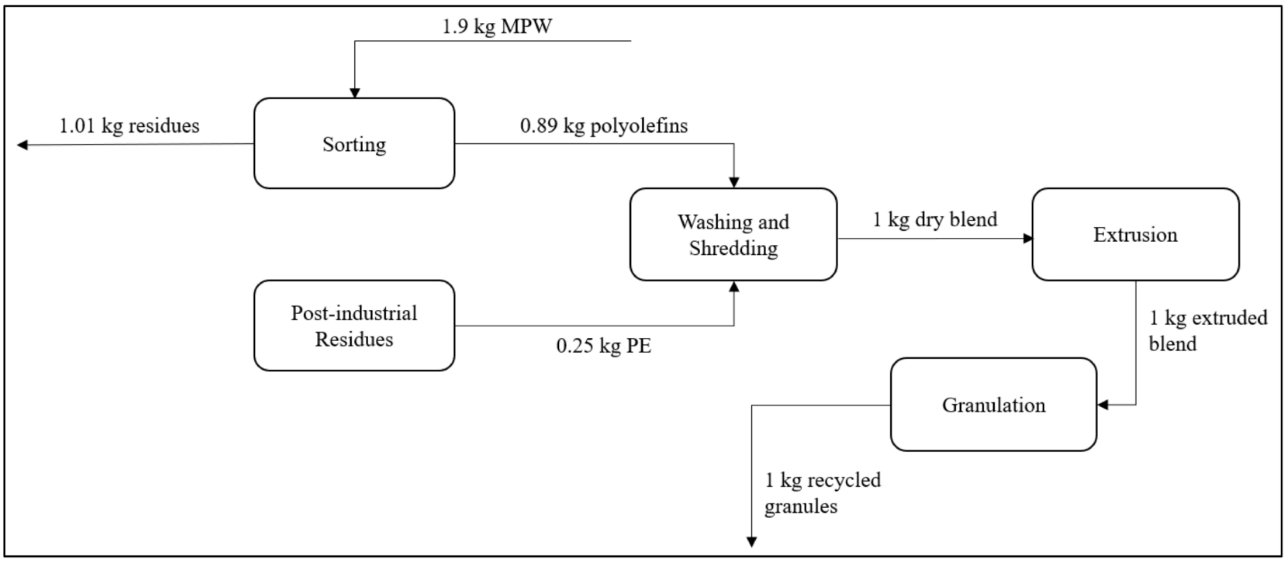


Figure 47 Graphical representation of the recycling process assessed via LCA. Mass flow for plastic materials is shown on arrows connecting each process. MPW: mixed plastic waste

3.4.2 Evaluation methods

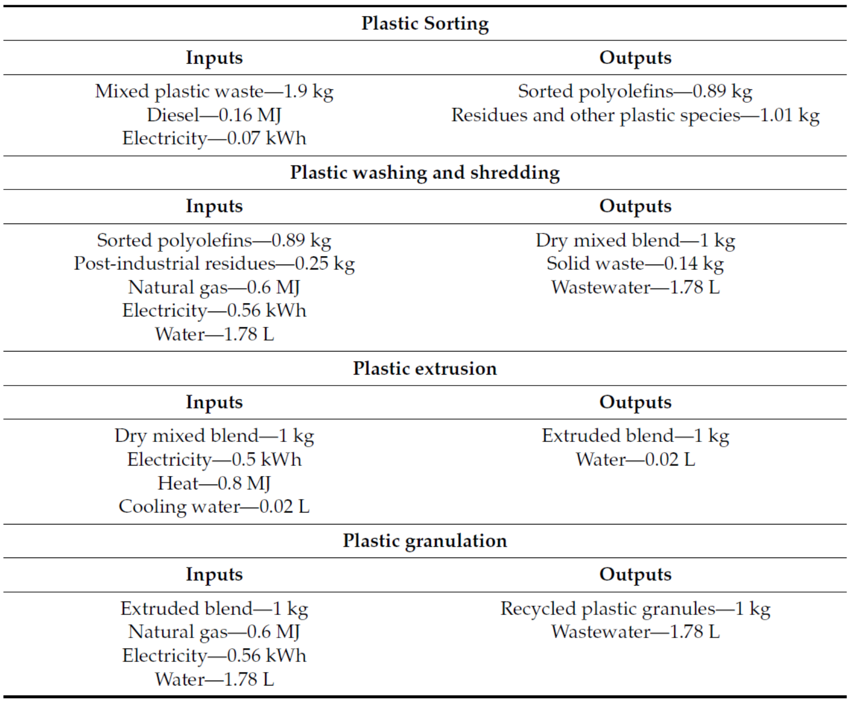
Life cycle impact assessment (LCIA) calculations were performed in OpenLCA 1.10.3.

The ReCiPe (H) 1.13 (National Institute for Public Health and the Environment, 2017) assessment method was selected for its use in previous literature, allowing comparability with other studies, and for its wide applicability. Impact categories considered were climate change (GWP100) in kg CO_2-Eq, particulate matter formation in kg PM_{10}-Eq, fossil depletion in kg oil-EQ, freshwater ecotoxicity in kg 1,4-DCD-EQ, terrestrial acidification in kg SO_2-EQ and water depletion in m^3 water-EQ.

3.4.3 Life Cycle Inventory Analysis

For processes leading to the production of virgin, high-density polypropylene, life cycle inventory data was obtained from the ecoinvent 3.7.1 consequential database (Wernet et al., 2016). European datasets were used where possible, otherwise global datasets were chosen. For processes modeling plastic recycling, data about process efficiency and energy requirements were gathered from previous literature (Arena et al., 2003; Khoo, 2019; Perugini et al., 2005). Extrusion data were adapted from the same ecoinvent 3.7.1 dataset. Table 26 summarizes inputs and outputs of the whole recycling chain.

Table 26 Input and output data for the recycling process.



Plastic Sorting	
Inputs	**Outputs**
Mixed plastic waste—1.9 kg Diesel—0.16 MJ Electricity—0.07 kWh	Sorted polyolefins—0.89 kg Residues and other plastic species—1.01 kg
Plastic washing and shredding	
Inputs	**Outputs**
Sorted polyolefins—0.89 kg Post-industrial residues—0.25 kg Natural gas—0.6 MJ Electricity—0.56 kWh Water—1.78 L	Dry mixed blend—1 kg Solid waste—0.14 kg Wastewater—1.78 L
Plastic extrusion	
Inputs	**Outputs**
Dry mixed blend—1 kg Electricity—0.5 kWh Heat—0.8 MJ Cooling water—0.02 L	Extruded blend—1 kg Water—0.02 L
Plastic granulation	
Inputs	**Outputs**
Extruded blend—1 kg Natural gas—0.6 MJ Electricity—0.56 kWh Water—1.78 L	Recycled plastic granules—1 kg Wastewater—1.78 L

3.4.4 Results

The life cycle assessment analysis was carried out comparing a baseline scenario of virgin polypropylene production from fossil fuels with a recycling process. Blend PC78-PI22 was used as a model, considering that it was the most promising according to the tests described in earlier chapters. Laboratory procedures were scaled-up to industrial level for this simulation. For the use of recycled plastics, the recycling scenario received credits in each impact category equal to the impact avoided, in that same category, by not producing an equivalent amount of virgin plastic. Results for both scenarios are shown in Table 27.

Table 27 LCIA results for both scenarios

Impact category	Unit	Virgin Plastic	Recycled plastic
freshwater ecotoxicity	[kg 1,4-DCB-Eq]	0.0262	0.03
Climate change	[kg CO2-Eq]	18.347	-0.2798
Fossil Deplatation	[kg oil-Eq]	18.295	-0.7874
Particulat matter formatio	[kg PM10-Eq]	0.0018	0.0007
Water deplatation	[m3 water-Eq]	0.0187	-0.0089
Terrestrial acidifcation	[kg SO2-Eq]	0.0049	0.0023

The percentage variation for each category is shown graphically in Figure 48. It is clear that for most impact categories, the recycling scenario was characterized by considerable reductions compared to the baseline scenario. Assuming that plastic use will remain constant, hard plastic recycling could also help reduce climate change and fossil fuel depletion; the avoided production of virgin plastic, in fact, directly translates into a marked reduction in fossil hydrocarbons used for the synthesis of artificial polymers and for energy production. Moreover, water depletion was drastically decreased, as one of the most impactful processes in PP production in this regard was completely avoided (Wernet et al., 2016).

Figure 48 Percentage variation in each impact category for the recycled plastic scenario compared to the virgin plastic scenario. FETPinf: freshwater ecotoxicity; GWP100: climate change; FDP: fossil depletion; PMFP: particulate matter formation; WDP: water depletion; TAP100: terrestrial acidification

Previous literature (Lazarevic et al., 2010; Rigamonti et al., 2009) highlights the problems arising from a reduction in quality due to product use and recycling. In general, when a closed-loop production chain cannot be achieved because of lowered quality of the recycled material, an open-loop production chain should be modelled considering possible uses of recovered materials. As plastic recycling becomes more commonplace and recycled plastic finds new uses, defining which virgin products could be potentially substituted by recycled plastic can be difficult. Thus, to account for the lower quality of recycled plastic, the use of a substitution ratio is suggested for LCA studies focusing on recycling processes. For the Italian market, for example, Rigamonti et al. (2009) suggest a substitution ratio of 1:0.81. In this study, recycled plastic was blended with pure industrial plastic residues: 1 kg of recycled blend actually contained 0.78 kg of recycled plastic. This ratio was a direct consequence of the lower properties of pure recycled plastic and as such, subsumes the substitution ratio within it. This ratio, equal to 1:0.78, is in line with previous studies on the recycling of other plastic species.

4 Case study 3: Recycling of Absorbent Hygiene Products (AHP)

4.1 Case study description

The ReCePiT project was developed starting from process implemented by Fater S.p.A., co-financed by the Lazio Region through the "KETS Enabling Technologies Public Notice" (POR FESR Lazio 2014-2020, Axis I - Research and Innovation and Axis 3 - Competitiveness). In the project, various alternative methods were studied for the use of cellulose recovered from Absorbent Hygiene Products (AHP); innovative thermal insulation panels were produced with this material and the potential as insulation flakes were tested in a laboratory house, also fabrics (viscose, rayon, acetates), air treatment systems (filters) were verified in the project, also the energy purposes were considered. LCA study was carried out to compare the use of insulation panel and energy production from Anaerobic digestion. The aim was to assess the feasibility of reusing this material and gradually contributing to the reduction of waste in a circular economy view.

This case study is focused on two topics of those mentioned above: the use of cellulose for thermal insulation and the evaluation of environmental impact of insulation panel compared to the use for energy purposes.

4.2 Materials and methods

4.2.1 Characterisation of cellulose

As part of the ReCePIT project, the CNR-ISMN deals with the selection of absorbent Hygiene Products (AHP) identified as representative of the various types of AHP used by consumers and their characterization. The characterization of the AHPs before and after the sterilization treatments was carried out in order to evaluate any structural changes and the quality of the cellulose present. In this context, only the results of the sterilized cellulose provided by Fater will be discussed.

The analysis of the cellulose of AHP was carried out using the following analytical techniques: infrared spectroscopy in Fourier transforms with accessory for attenuated total reflectance (FTIR-ATR), X-ray diffraction (XRD) and thermogravimetric analysis (TGA).

The selection techniques to be used to analyse the degree of crystallinity, to obtain compositional and morphological information on cellulose and, according to literature data of cellulose, are the most cellulosic components (Jonoobi et al., 2009). These analyses were carried out on cellulose samples taken from the selected AHPs and separated from the superabsorbent component present.

4.2.2 Laboratory house characteristics and blowing

A laboratory house was built at the CNR - Area della Ricerca Roma-1, consisting of wooden modules (Figure 49).

The realisation of the wooden modules is aimed to energetic performance evaluation of the components of the building envelope following the application of materials insufflated as loose material, in particular cellulose from recycled AHP. The wooden module has an internal cavity for both the perimeter walls and the ceiling.

Figure 49 Laboratory house

The Laboratory House is characterised by a dry prefabricated *blockhaus* typology, with a rectangular plan, consisting of three modules. The central module C1 is the entrance/connection area, allowing access to the two remaining modules M1 and M2, located to the right and left of the central module.

The modules are composed of beams of different sizes, ranging from a minimum length of 72 cm to a maximum of 500 cm. In order to guarantee the maximum stability of the structure, threaded rods with a diameter of 12 mm pass through the holes provided, which are also anchored at the ends to the floor, consisting of a single element in X-lam with a thickness of 10 cm and to the sloping roof beams made of glued laminated timber with a section of 10x12/16 cm.

The external walls, made up of the load-bearing *blockhaus* elements, are completed with the assembly inside them of the cavity for the insulating materials.

The cavity inside the structural wall was created by juxtaposing two rows of wooden mullions with a section of 4x4 cm and 62.5 cm centres. The double frame facilitates the passage of the systems. To close the gap, an Oriented strand board (OSB) panel was placed to create an 8 cm gap.

Blowing was carried out by drilling holes in the OSB panels at a maximum regular distance of 50 cm to allow the loose material to enter the cavity uniformly (Figure 50).

The roof is made with a removable system that guarantees the possibility of inserting prefabricated modules for toilets or kitchens inside. The system consists of a set of beams and planking that can be hooked onto threaded rods with eyebolts and lifted easily. The flat roof made of seamed aluminium sheet metal elements is fixed to the board that defines the ceiling of the modules. The roof has a slight south-facing slope and an eaves gutter on the long side of the modules. The roof slope is ensured by roof beams made of glued laminated timber with a section of 10 x 12/16 cm. Above the beams there is a planking of 2 cm thick tapped fir slats, with additional upper layers consisting of a vapour barrier, 8 cm thick cork panel insulation, a breathable sheet and a second 2.5 cm thick planking supporting the standing seam aluminium sheet.

An OSB panel 12 mm thick is applied to the intrados of the roof beams to create a cavity in which insulating material will be blown.

The walls and ceiling were insulated with appropriate machinery. The room facing East has been insulated, while the room facing West has not been insulated. Both rooms face south and north.

Figure 50 Insufflation procedure

The structure also has 2 windows of 180x114 cm, a French window of 90x219 cm and two internal doors of 70x210cm, equipped with double glazing.

The technical data for windows and French windows are given in Table 28.

Table 28 Characteristics of doors and French windows

Characteristics	Unit	Value
Thermal Transmittance	UW:	1.17
Glass Transmittance	Ug:	1
Acoustic Insulation	dB	36
Light Transm. Light	UL	65
Solar Factor	Sf:	40
Air Perm.	Class	4
Water	Class	E750
Wind	Class	C2

4.2.3 Analysis of microclimate parameters

The main microclimate and air quality parameters were analysed.

Two different measurement campaigns were carried out within the two environments: from 25th to 30th December 2020 and from 31st December to 6th January 2020. In both campaigns the heating systems were kept off.

Two further monitoring campaigns were carried out, with the presence of active heating systems set at 27 ° C. The two campaigns concerned the isolated and non-isolated room and outdoor measurements.

The parameters of temperature and relative humidity, expressed in large Celsius and percentage respectively, were investigated. An AM3220 with integrated digital interface and resolutions of 1% RH and 0.5 °C respectively was used as sensor to monitor RH and temperature.

4.2.4 Analysis of thermal exchanges

The study of thermal exchange is particularly important in order to assess the well-being of an individual in an indoor environment, which is influenced by the amount of energy exchanged through radiation, convection and, to a lesser extent, conduction. Heat transmission is also important to quantifying the energy requirements of buildings for their air conditioning, and is therefore a way of assessing the quality of the built environment in order to reduce energy consumption.

The analysis of the dispersing surfaces with different types of insulating material was carried out in order to compare the results of the bioclimatic parameters between commercial and experimental materials. The evaluation of the energy performance of the structure consists in the application of different materials in order to directly compare two different insulation types. In this case, a module with insufflation of cellulose from AHP was compared with the same module without insulation material and the insufflation of virgin cellulose from paper was compared with the insufflation of cellulose from AHP.

Other study have compared the thermal performance of cellulose from paper come from different country (Kwon & Yarbrough, 2004) or compares cellulose insulation material with the thermal performance of stone wool batts(Nicolajsen, 2005), but this study, for the first time, evaluates the thermal performance of cellulose come from AHP.

Two different evaluations were carried out: transmittance measurement and thermographic analysis. The measurements on the walls without and with cellulose were carried out under the same indoor operating conditions, with a temperature of 27 °C set in the heating system; the performance of the perimeter and partition walls of the two modules was evaluated. In addition, the different performance of the insulation layer applied to the ceiling and made from non-AHP cellulose without applying a heating system was also verified.

The measurement of thermal transmittance (U) was performed using a thermal flowmeter to assess the heat loss through the building envelope. As the thermal transmittance of the building envelope elements decreases, the less heat flows through them. The reference standard used for the measurement of thermal transmittance and thermal resistance was UNI ISO 9869-1:2015 (Thermal insulation - Building elements - In situ measurement of thermal resistance and thermal transmittance).

The measurements were acquired every 10 s by a probe for at least 72 h, the data were downloaded on computer and processed by a dedicated software that directly calculated the K coefficient and the parameters related to thermal insulation, using the "moving average" method or the so-called "black box" method (Figure 51).

The Italian national legislation D.Lgs 311/2006 (Corrective and integrative dispositions to the D.Lgs 19 August 2005, n.192, implementing the directive 2002/91/CE, related to the energy performance in buildings) has fixed, according to the different climatic zones, the thermal transmittance limits both for opaque structures (walls, floors, roofs) and for transparent structures including frames, as reported in the Table 29 and Table 30.

Table 29 Limit values, thermal transmittance U of vertical opaque structures, in W/m²K

Thermal Trasmittance	**U (W/m²K)**		
Climatic zone	From 1st of January 2006	From 1st of January 2008	From 1st of January 2010
A	0.85	0.72	0.62
B	0.64	0.54	0.48
C	0.57	0.46	0.40
D	0.50	0.40	0.36
E	0.46	0.37	0.34
F	0.44	0.35	0.33

Table 30 Limit values, of the thermal transmittance U of horizontal opaque structures or sloping roofing structures expressed in W/m^{2}K

Thermal Trasmittance	**U (W/m^{2}K)**		
Climatic zone	From 1st of January 2006	From 1st of January 2008	From 1st of January 2010
A	0.80	0.42	0.38
B	0.60	0.42	0.38
C	0.55	0.42	0.38
D	0.46	0.35	0.32
E	0.43	0.32	0.30
F	0.41	0.31	0.29

Figure 51 Trasmittance measurment

Thermographic analysis was applied to assess possible thermal points. The typical effects of thermal bridges are a decrease in internal surface temperatures; in the worst cases it can lead to high humidity in some parts of the structure and a significant increase in heat loss. Thermal bridges can be eliminated by inserting an additional insulating material to create a break in the heat flow.

4.3 Results

4.3.1 Analysis of cellulose

The cellulose components after sterilisation treatment obtained by Fater S.p.A were characterized by FTIR-ATR analysis (Figure 52). the values of the LOI, CI and HBI indices fall within the range outlined by the other samples analysed while the TCI value is slightly lower than the average.

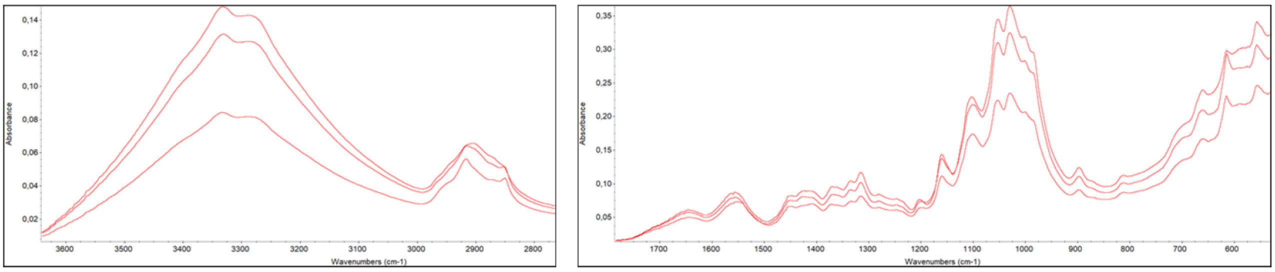


Figure 52 FTIR-ATR spectra of Fater S.p.A samples after sterilization in the wavenumber range between 3650 and 2500 cm-1 (left) and in the wavenumber range between 1800 and 600 cm-1 (right) approximately

The sterilised cellulose samples were also analysed by TGA. The sample of cellulose fibres derived from the AHP recycling process of Fater S.p.A shows a weight loss value of 51.9% between 225 and 425°C, slightly lower than the values obtained for the fibres sampled from the other AHPs selected. This difference is attributable to a lower fraction of cellulose compared to that relating to the non-cellulosic material present.

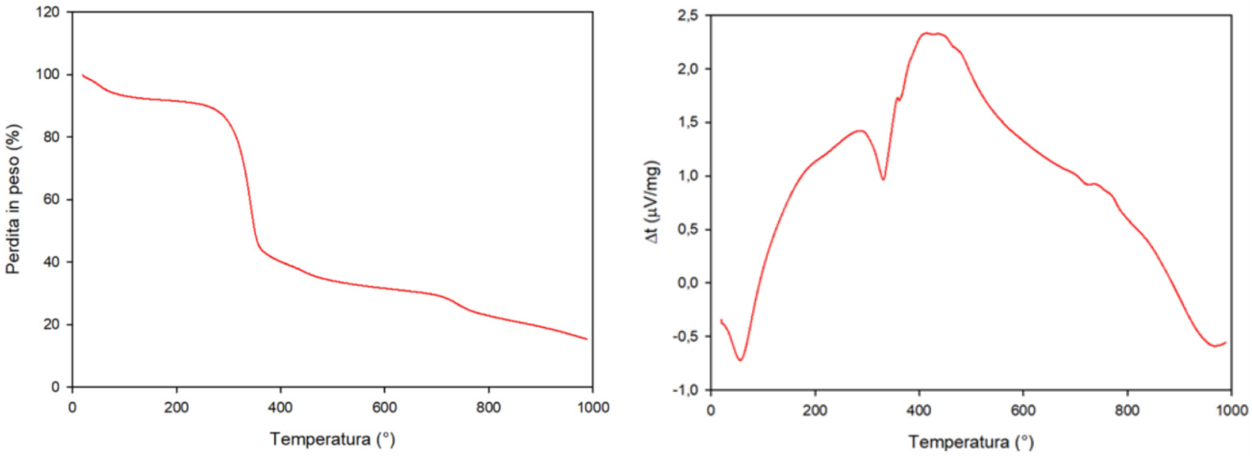


Figure 53 Thermogravimetric curve (left) and corresponding DTA thermogram (right) of the samples taken from the cellulose treated by the Company Fater S.p.A.

The material used and sterilized was also subjected to characterization with X-ray diffraction. The crystallinity index and crystallite size values observed for samples deriving from mixed AHP waste after sterilization (supplied by the Fater company), fall within the range plotted from the values obtained for the AHPs chosen after sterilization treatment. These results indicate that the selected and sterilized AHPs do not differ much from the sterilized AHPs derived from waste. The values obtained for the latter are both above average, suggesting the maintenance of a good crystallinity index and a good size of the cellulose crystallites even after the recovery treatment.

4.3.2 Analysis of microclimate parameters

The measurements for the evaluation of microclimatic parameters were performed to assess the climatic well-being of the laboratory house.

The first measurement assessed the differences of temperature and relative humidity in the two rooms, insulated and non-insulated respectively.

Into the non-insulated room, a considerable relative humidity has been found compared to the insulated room. The temperature recorded in the two rooms had a different trend: in the insulated room, despite the night- day variations, the temperature has been much more constant. In the non-insulated room, the temperature has been generally lower and greater temperature variations between day and night and therefore a greater variation in the internal temperature have been registered (Figure 54, Figure 55).

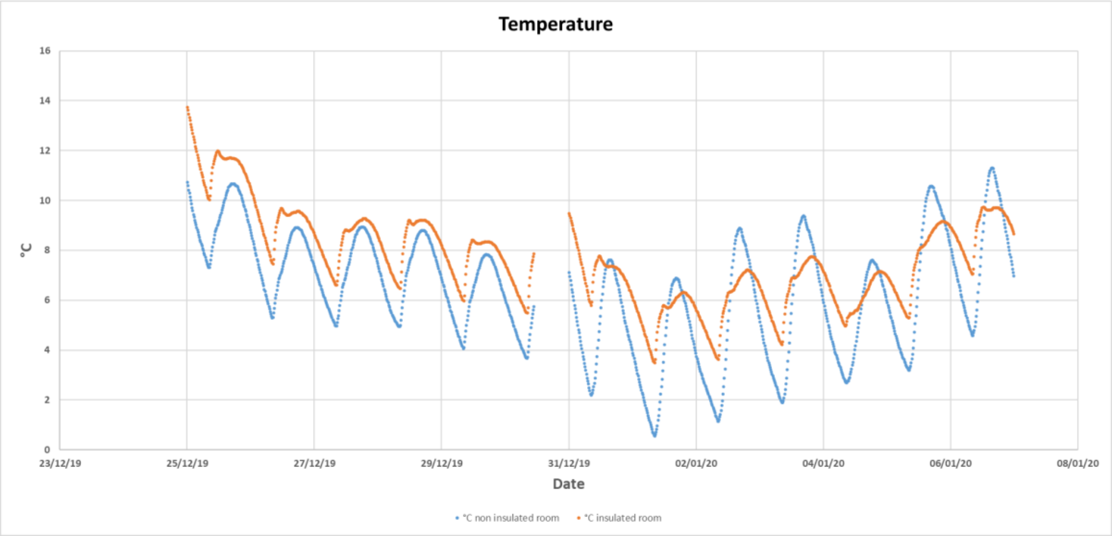


Figure 54 Temperature indoor – insulated and no insulated room

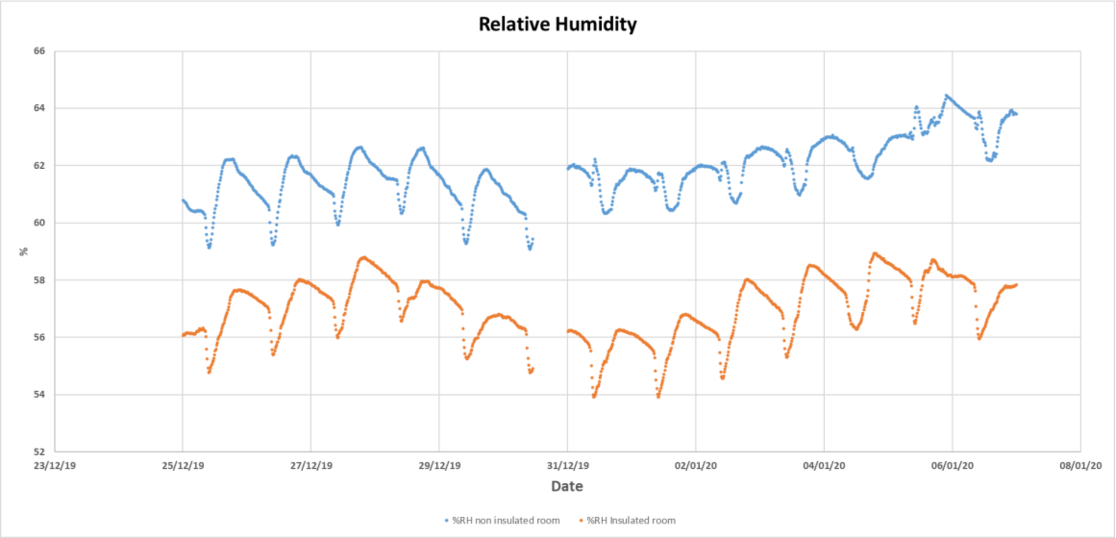


Figure 55 Relative Humidity indoor – insulated and no insulated room

In the second measurement campaign in which the heating systems has been kept on, the temperature variation between inside and outside in the two types of rooms was checked. It was found that although the temperatures reached values below zero degrees in both measurements, only in the insulated room was a constant temperature recorded. Otherwise in the non-insulated room the inside temperature is influenced by the temperature recorded outside (Figure 56, Figure 57).

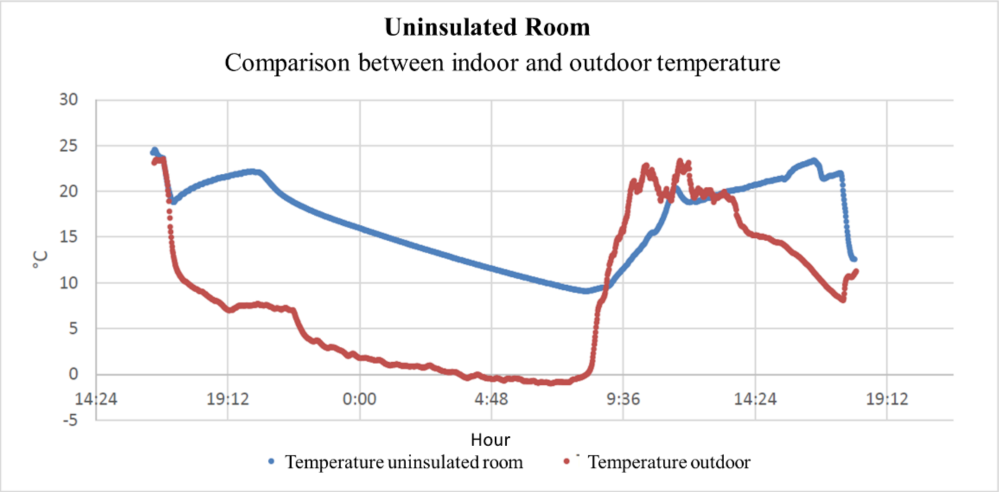


Figure 56 Comparison between indoor and outdoor temperature – Uninsulated room

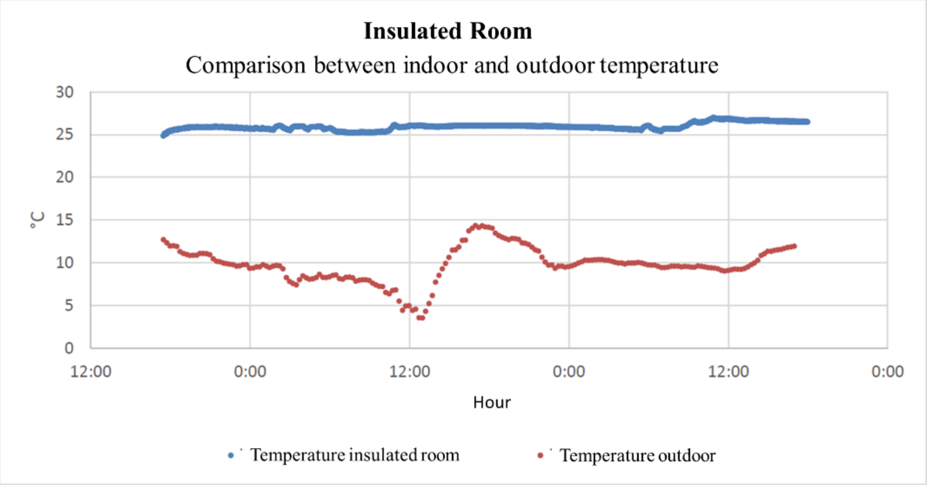


Figure 57 Comparison between indoor and outdoor temperature – Insulated room

4.3.3 Analysis of thermal exchanges

The collected data was processed and the following measurements were compared:

- uninsulated south and east wall with insulated south and west wall,

- internal ceiling in which non-PAH cellulose was blown with the non-insulated internal ceiling.

The following graphs and tables show the temperatures of the internal and external opaque surfaces, the irradiance and transmittance measurements, and a summary table with the average values of the measurements.

In the non-insulated room, measurements were performed on the east and south facing relatives. In the Figure 58, Figure 59, Figure 60 the graphs of the east wall are shown, while the Figure 61, Figure 62, Figure 63 show the results of the south wall.

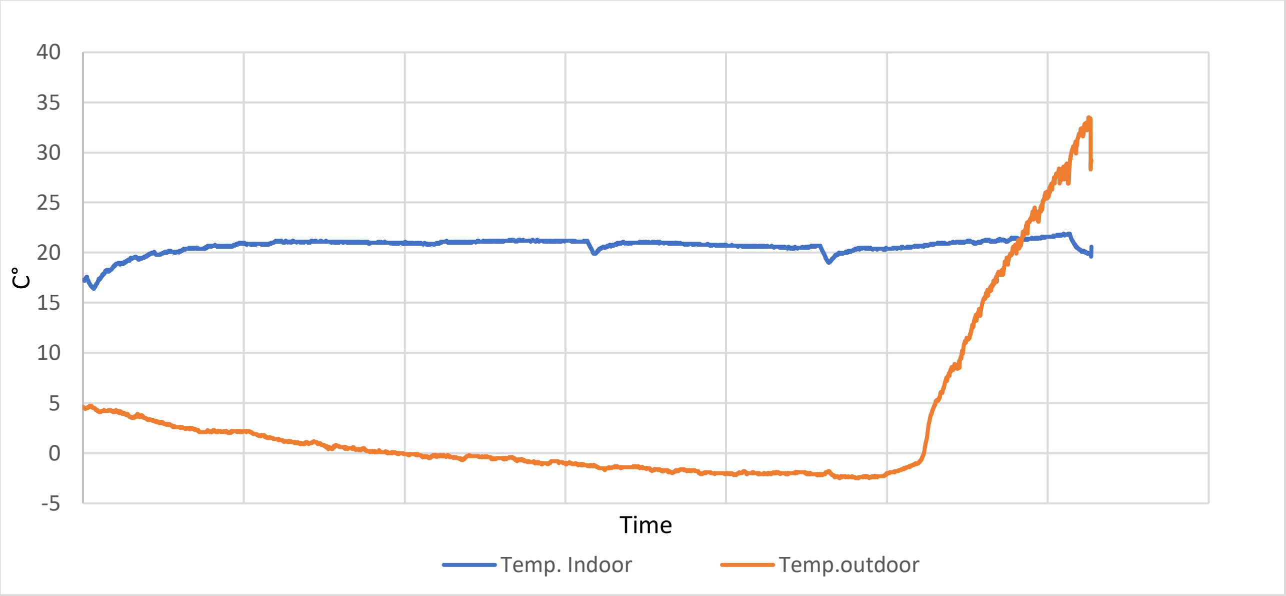


Figure 58 Indoor and outdoor wall temperature – uninsulated east wall

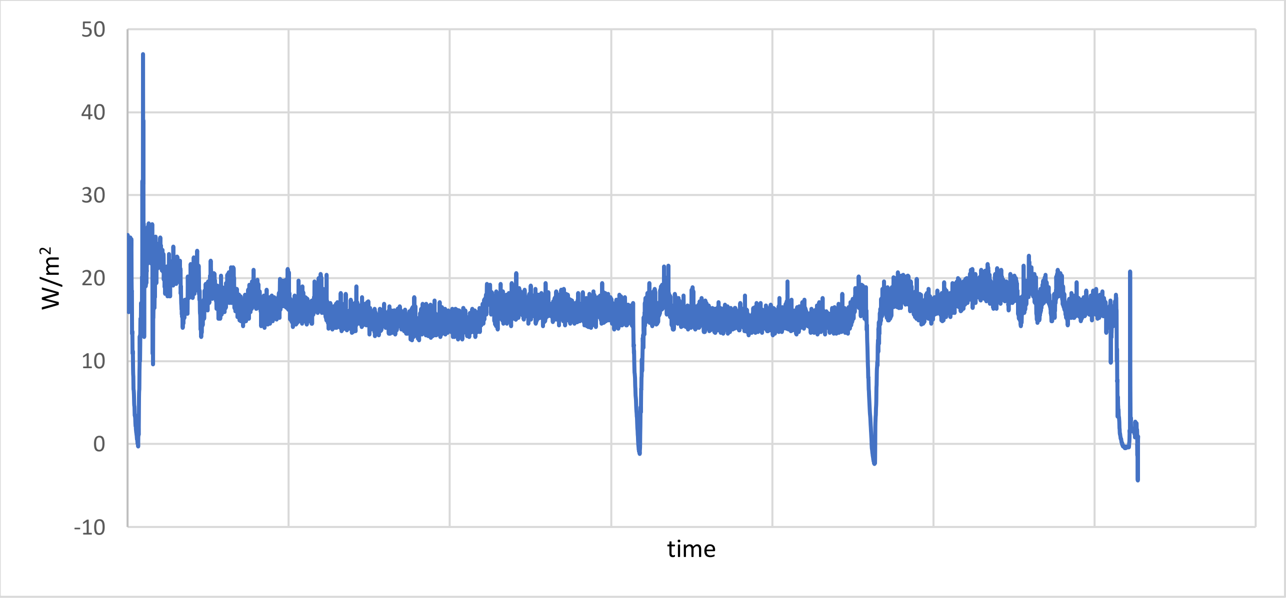


Figure 59 Irradiance measurement – uninsulated east wall

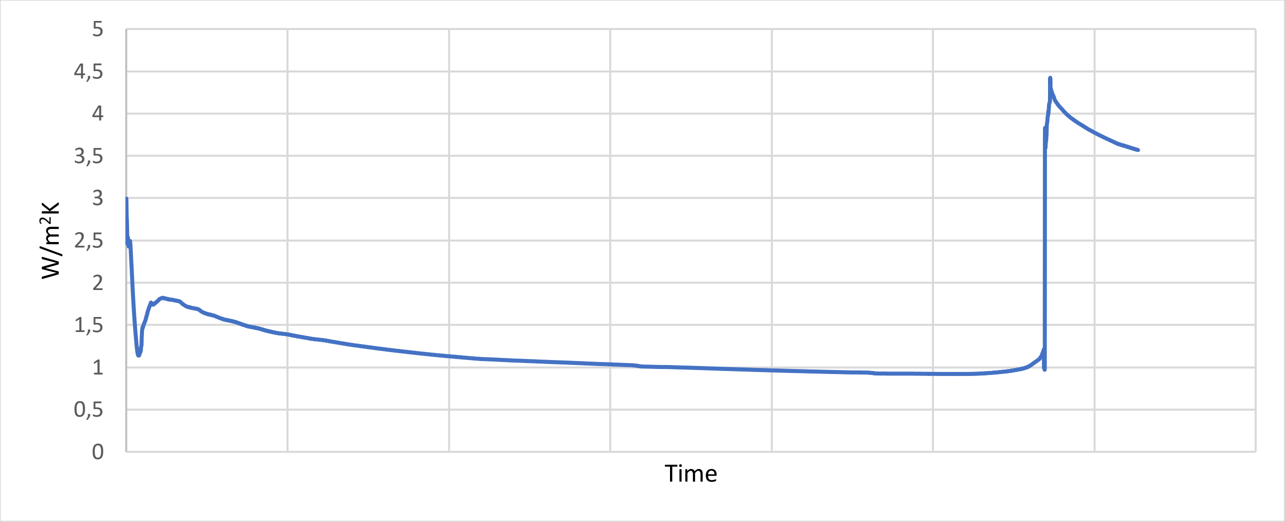


Figure 60 Average of transmittance value – uninsulated east wall

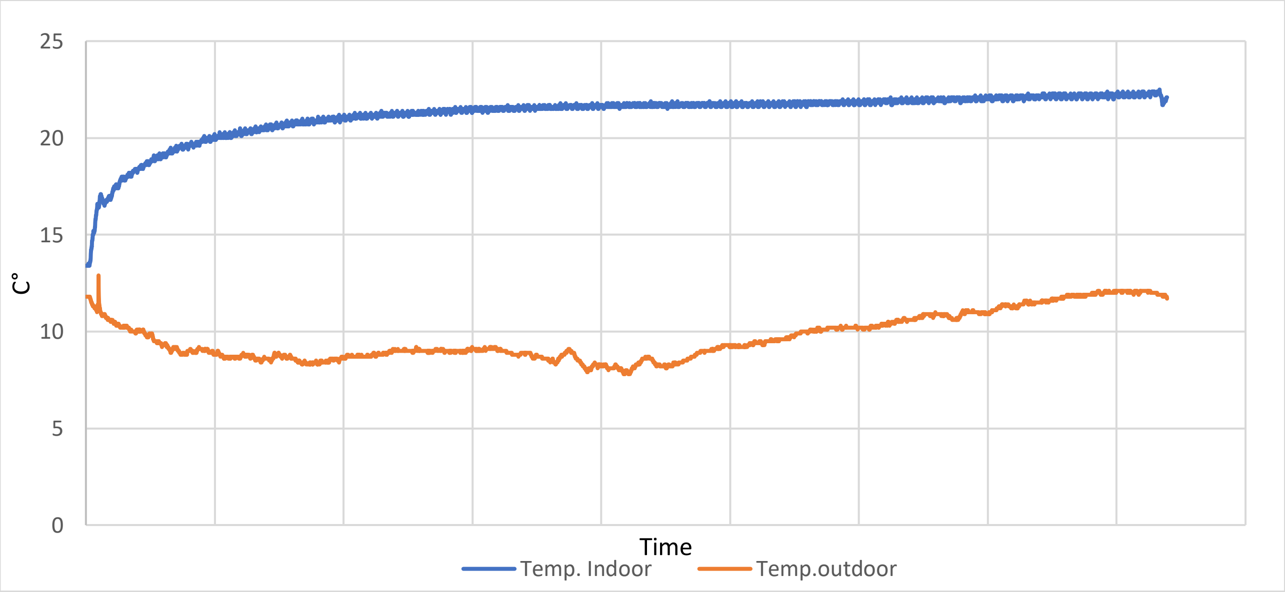


Figure 61 Indoor and outdoor wall temperature – uninsulated south wall

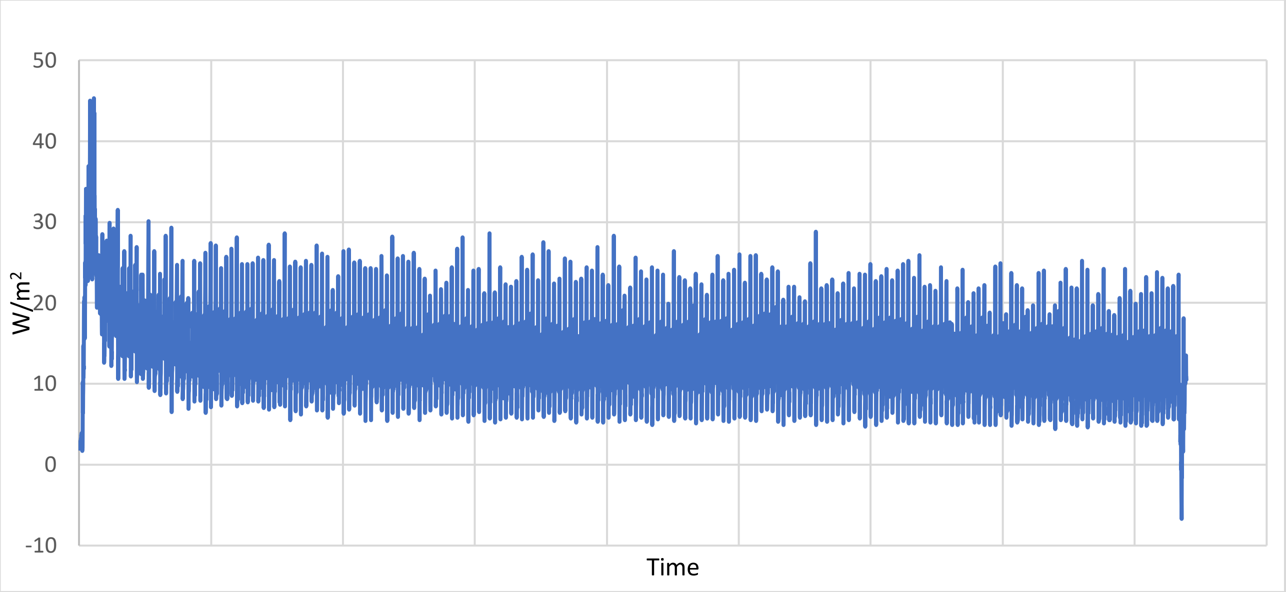


Figure 62 Irradiance measurement – uninsulated south wall

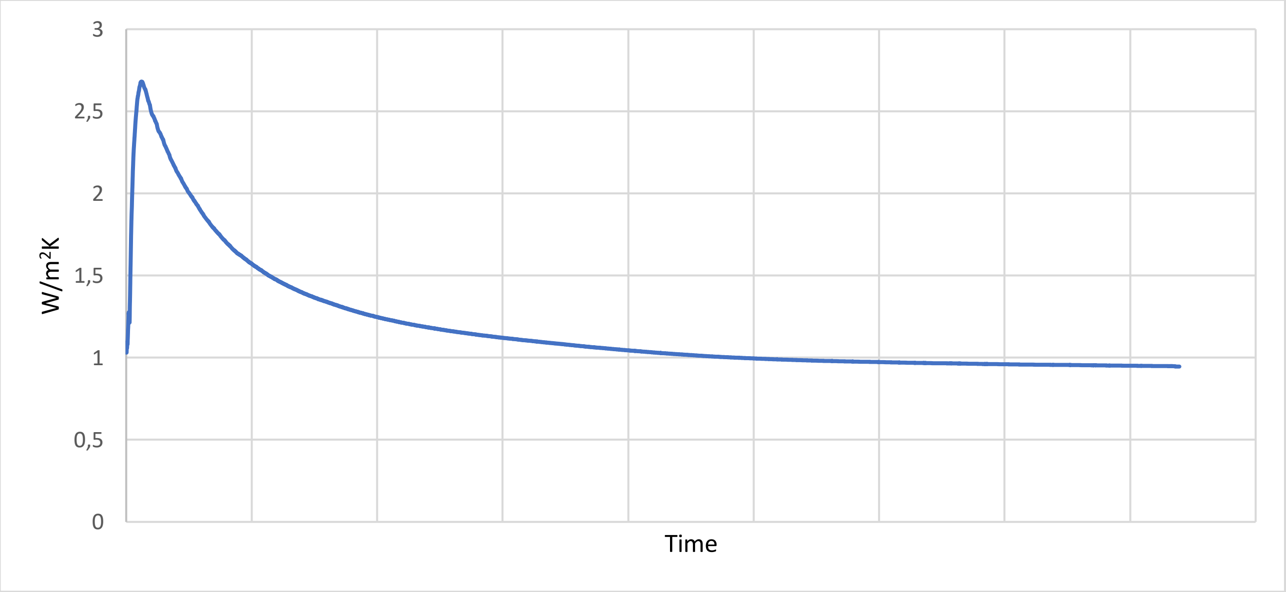


Figure 63 Average of transmittance value – uninsulated south wall

In Table 31 the average values of flow, internal/external wall temperatures and the transmittance value are shown for the south and east walls.

Table 31 Values recorded on south and east walls – uninsulated wall

	Irradiance	**Indoor wall temperature**	**Outdoor wall temperature**	**Transmittance**
	W/m^2	C°	C°	W/m^{2}K
South wall	12.76	21.20	9.69	1.18
East wall	15.75	20.68	3.03	1.40

In the isolated room, measurements on the west and south facing were carried out. Figure 64, Figure 65, Figure 66, show the graphs of the west wall, while Figure 67, Figure 68, Figure 69 show the results of the south wall.

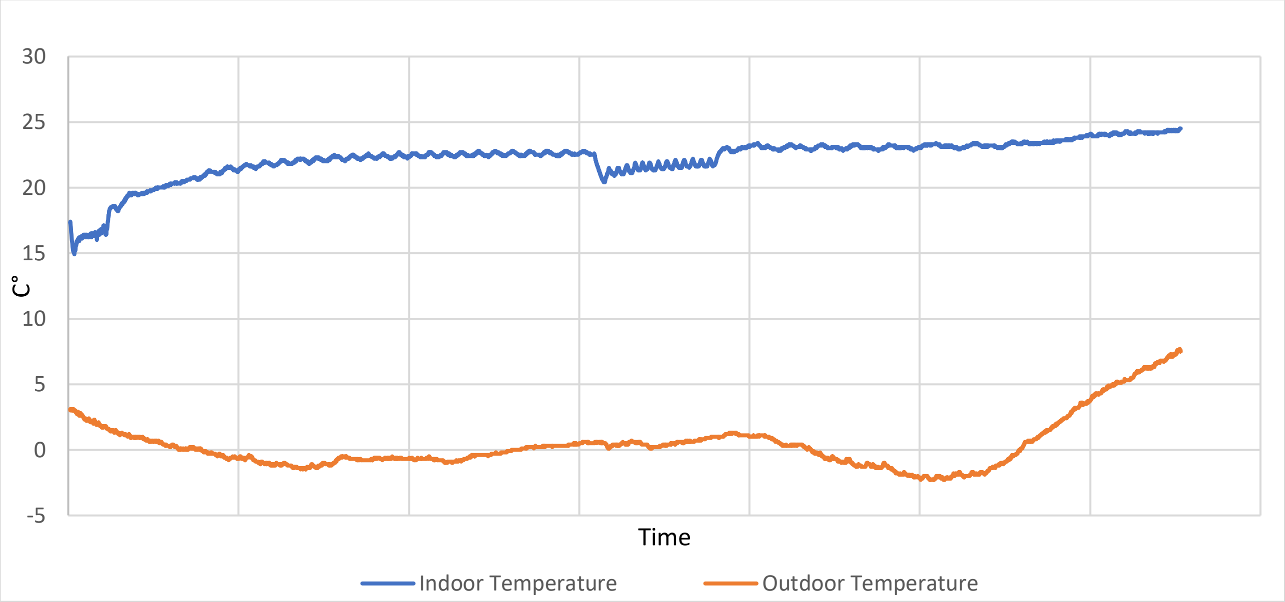


Figure 64 Indoor and outdoor wall temperature – insulated west wall

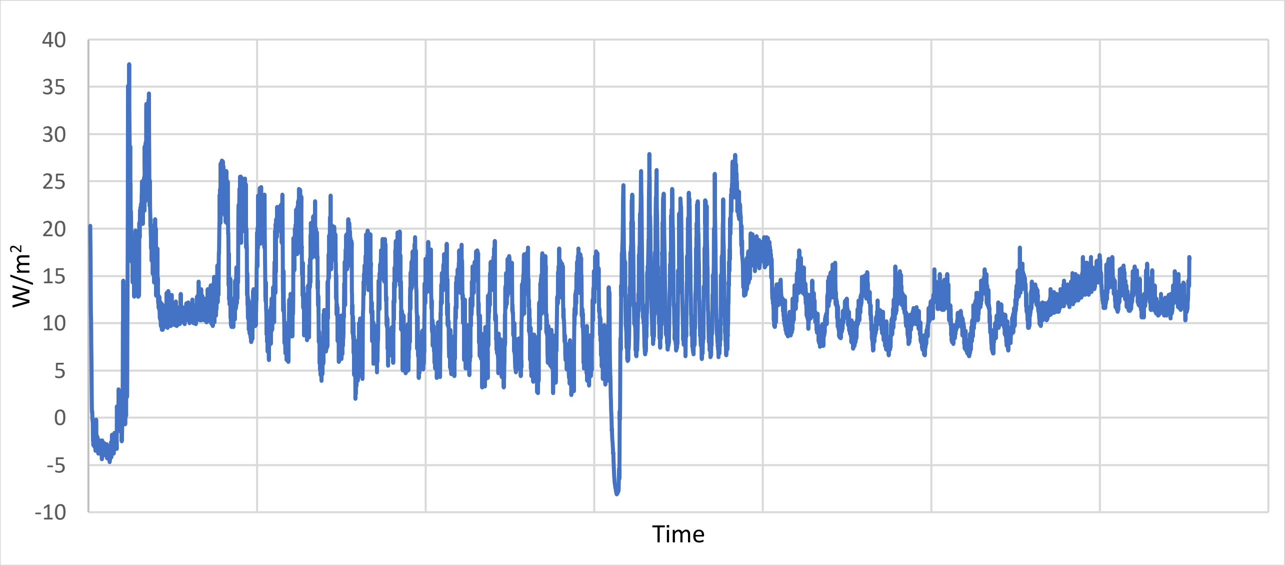


Figure 65 Irradiance measurement – insulated west wall

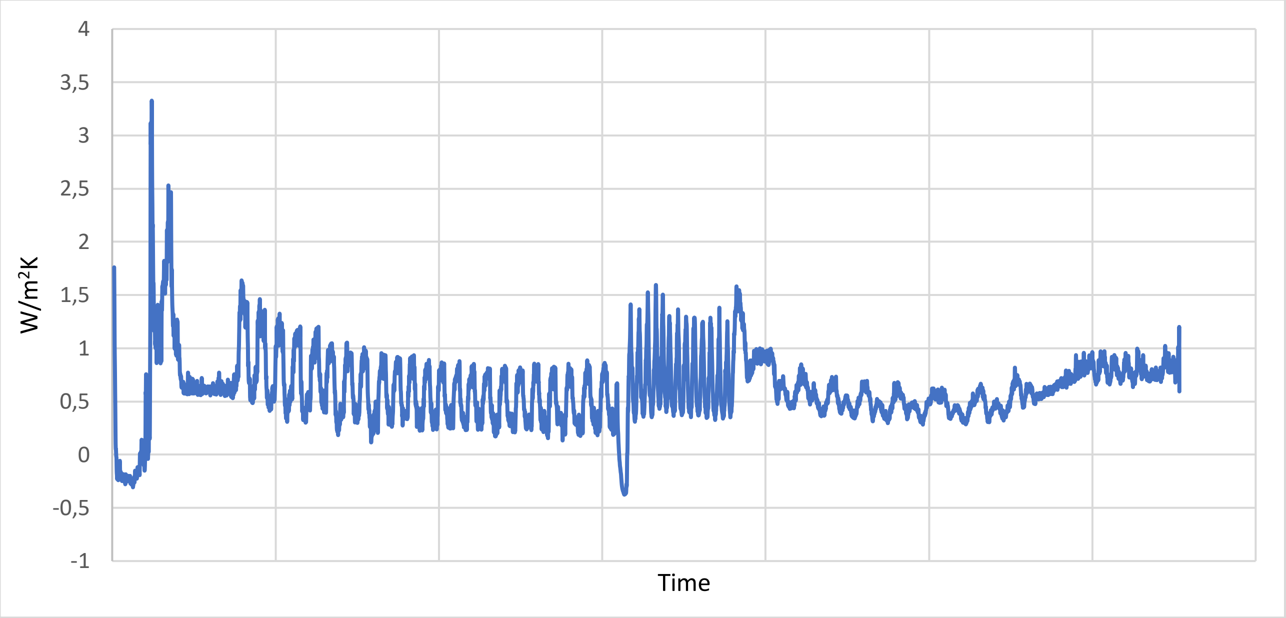


Figure 66 Average of transmittance value – insulated west wall

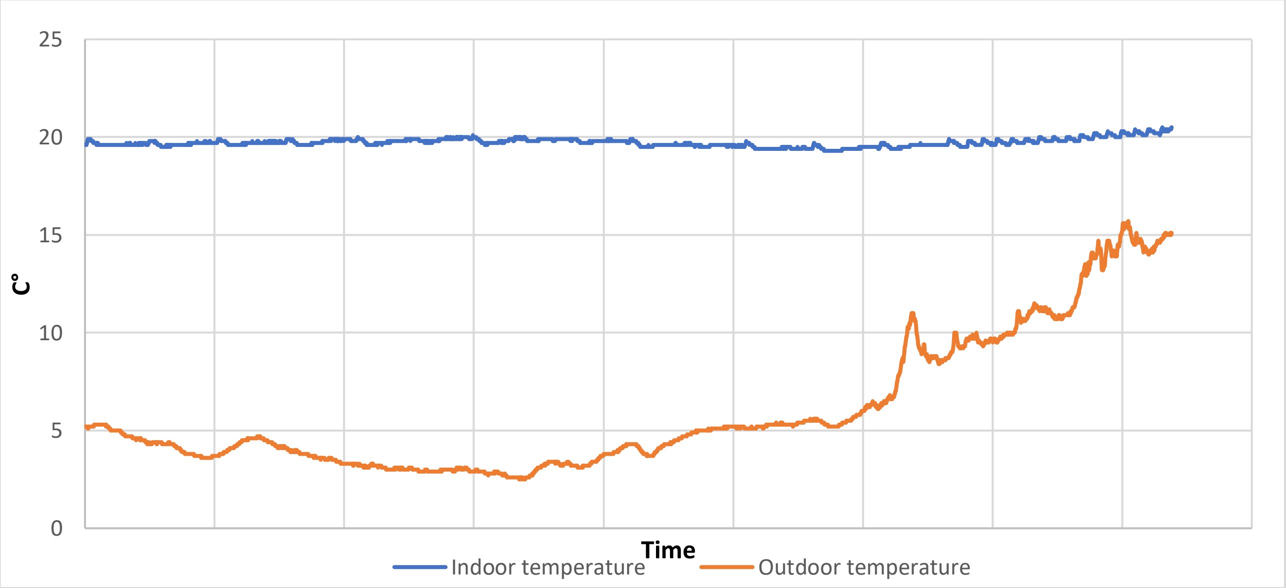


Figure 67 Indoor and outdoor wall temperature – insulated south wall

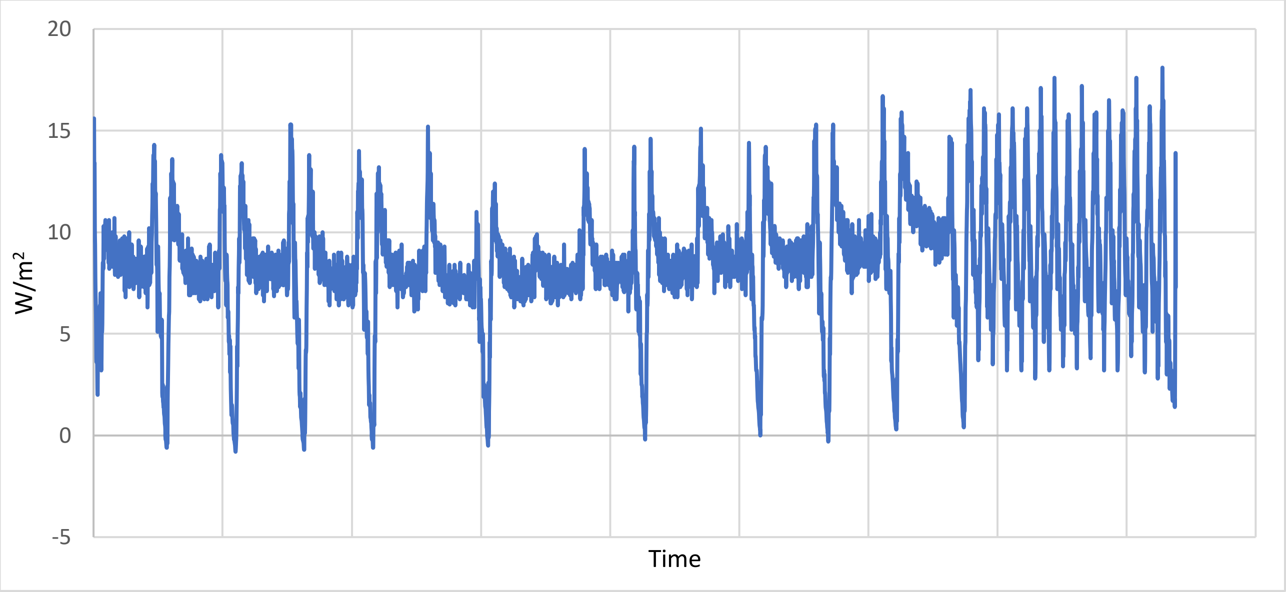


Figure 68 Irradiance measurement – insulated south wall

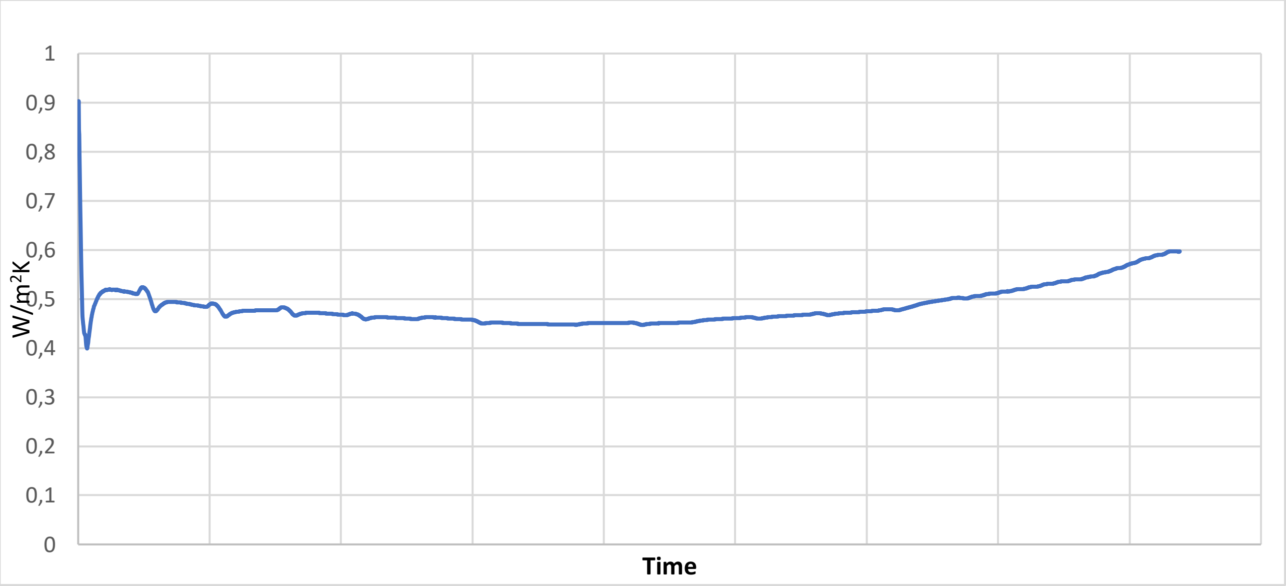


Figure 69 Average of transmittance value – insulated south wall

In Table 32 the average values of flow, internal/external wall temperatures and the transmittance value are shown for the south and west walls.

Table 32 Values recorded on south and west walls – uninsulated wall

	Irradiance	**Indoor wall temperature**	**Outdoor wall temperature**	**Transmittance**
	W/m^2	°C	°C	W/m^{2}K
South wall	8.37	19.72	5.99	0.48
West wall	11.96	22.22	0.40	0.62

The last monitoring campaign was carried out by comparing the measurements made on the insulated ceiling in which cellulose flake fibre not deriving from AHP was blown with the non-insulated ceiling (Figure 70, Figure 71, Figure 72).

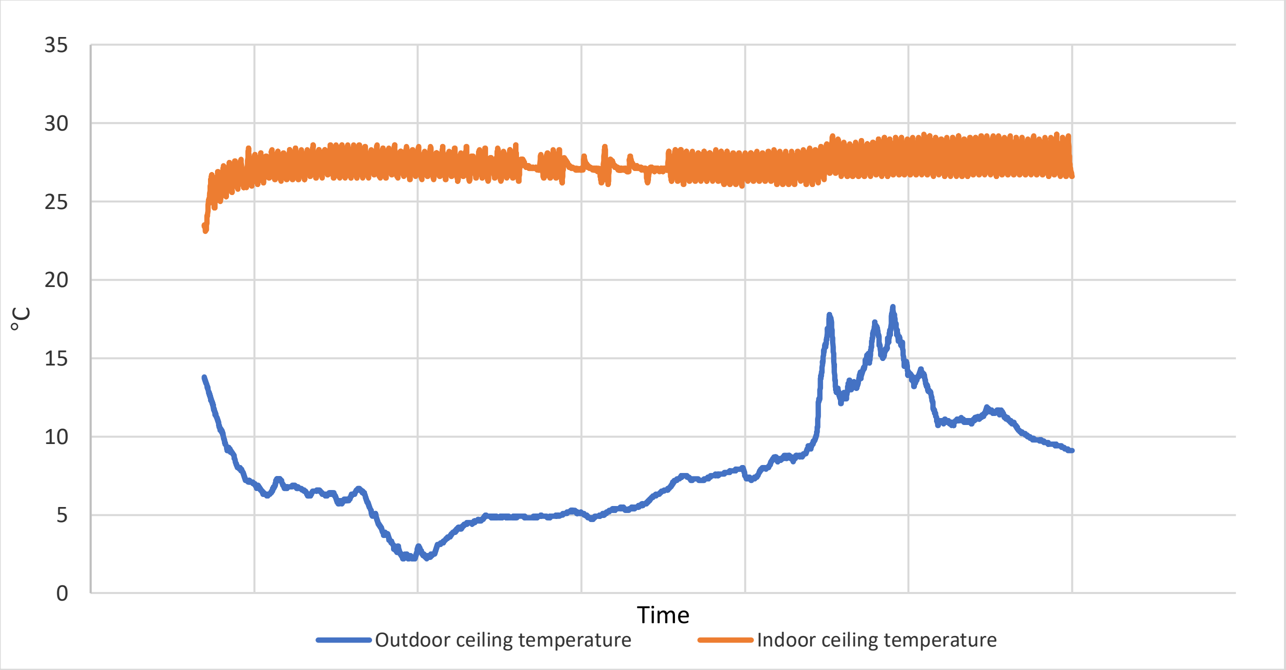


Figure 70 Indoor and outdoor ceiling temperature – insulated ceiling

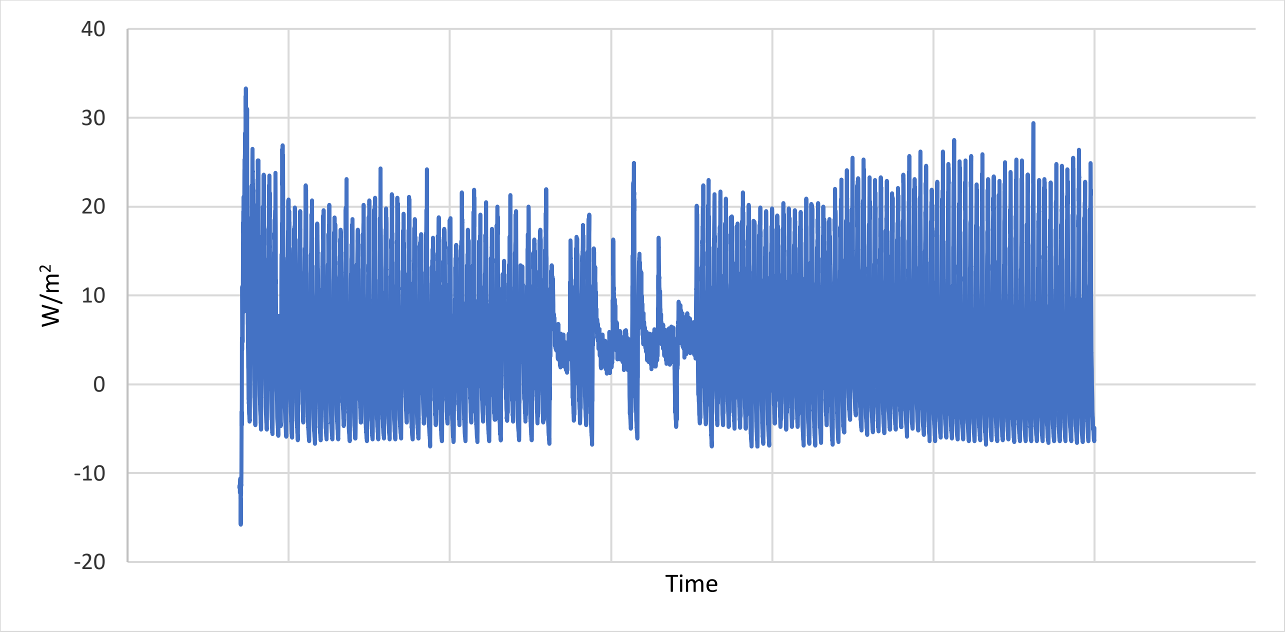


Figure 71 Irradiance measurement– insulated ceiling

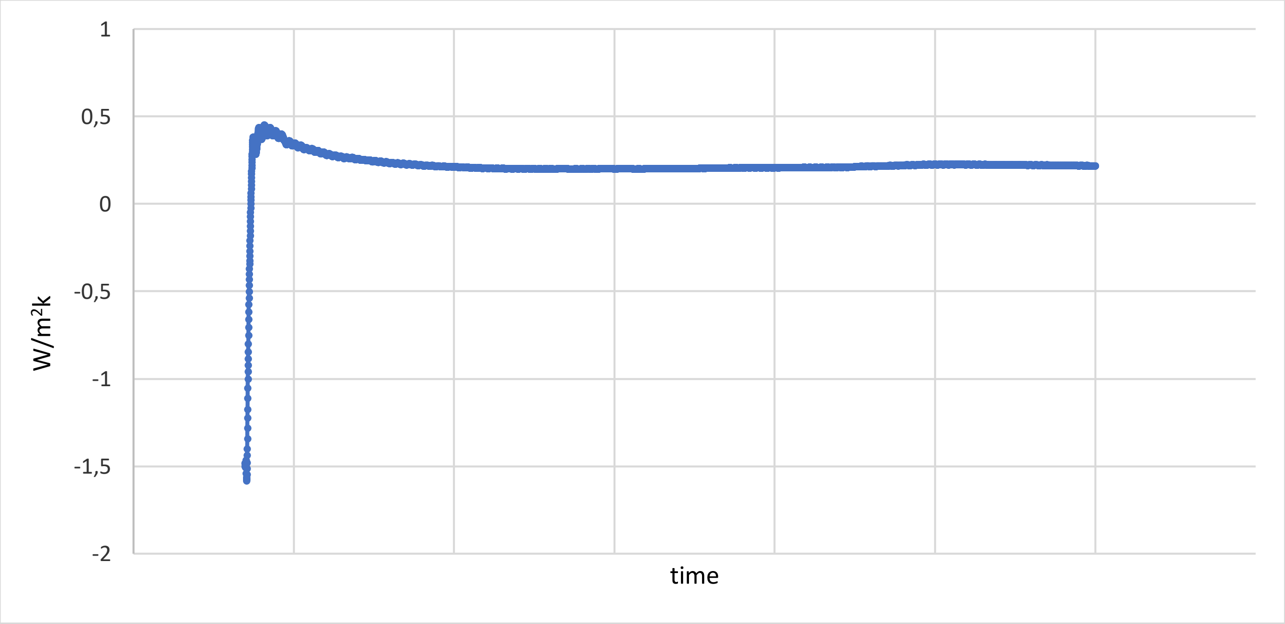


Figure 72 Average of transmittance value – – insulated ceiling

The Table 33 shows the average values of the flux, the internal/external temperatures of the internal ceiling/external cover and the transmittance value for the internal ceiling insulated with non-AHP cellulose. The reference transmittance for the internal ceiling of the non-insulated module room measured is approximately 0.8 W/m^{2}K.

Table 33 Values recorded on insulated ceiling

	Irradiance	**Indoor ceiling temperature**	**Outdoor ceiling temperature**	**Transmittance**
	W/m^2	C°	C°	W/m^{2}K
ceiling	27.52	8.77	4.76	0.21

The measurements on the insulated and uninsulated walls showed a variation of transmittance for both: the south compared with east wall and south compared with west walls. In the uninsulated walls the recorded average transmittance values are greater than 1; while in the insulated walls the recorded average transmittance values are between 0.48 and 0.62 W/m^{2}K. As for the ceiling, the recorded average transmittance value is 0.21 W/m^{2}K.

On the basis of the Italian Legislative Decree 311/2006 (Corrective and integrative dispositions to the Legislative Decree 19 August 2005, n.192, implementing the directive 2002/91/CE, concerning the energy performance in buildings) which has fixed, according to the different climatic zones, the thermal transmittance limit values, the laboratory house is suitable for Zone A and Zone B. For the ceiling, the recorded value is suitable for all climate zones.

The thermographic analysis performed for the measurement of thermal bridges verified that the use of cellulose as an insulating material is suitable in contrast to the non-insulated room results.

For example, in Figure 73 the surfaces adjacent to the ceiling and the wall are compared. From the figure on the right, the minimum temperature in the presence of the existing thermal bridge between the ceiling and adjacent walls is about 2 °C lower than in the insulated room where the thermal bridge is not evident.

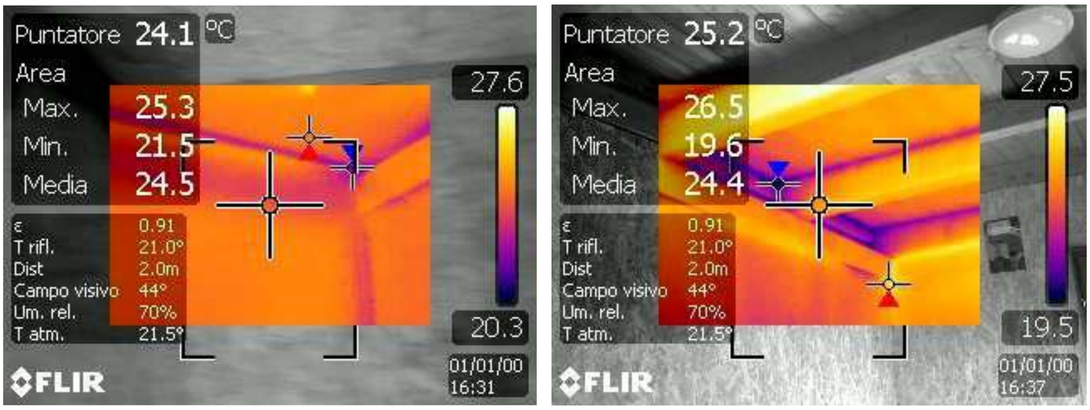


Figure 73 Comparison of thermal bridges between the ceiling and adjacent walls in the insulated and non-insulated room

As indicated by Lopez Hurtado et al., (2016) the properties of cellulose fibre, can vary depending on the mode of production and the method of installation. In fact, there are studies that show a difference in conductivity if there is a variation in the type of newsprint, in particular fibres that have undergone more recycling processes are less performing (higher conductivity values). In the study of Kwon & Yarbrough, (2004) cellulose samples from the USA and Korea were compared. The data found that Korean fibres, which are shorter due to more recycling processes, show a higher value for thermal conductivity, and therefore lower insulation performance than cellulose fibres from the US.

Since cellulose fibres are naturally hygroscopic, moisture absorption can also affect thermal conductivity values. Notwithstanding, as described by Lopez Hurtado et al., (2016), the increase in thermal conductivity could be generally considered negligible. The advantage of using hygroscopic material, such as cellulose insulation, is related to the control of internal relative humidity, in fact several studies have shown its effectiveness (Osanyintola & Simonson, 2006), (Kumar et al., 2020).

Although sustainable insulators are highly combustible and may not have the same thermal properties as other materials, recycled cellulose is a sustainable material, with a low carbon content compared to other materials that are made from non-renewable resources and have a high embodied energy, thus cellulose having a lower impact on the environment compared to other materials.

On the other hand, as indicated by Kumar et al. (2020), optimal insulation materials based on life cycle cost may have a higher carbon emission into the atmosphere due to the higher embodied energy, which means that it will not be environmentally sustainable. So, the use of recycled and natural insulation materials could also help reduce the carbon footprint of building construction and operation.

4.4 Life Cycle Assessment Analysis

4.4.1 Scope of application

Among the different uses of recycled cellulose as indicated in paragraph 4.1 assessment of development insulation panels to improve thermal comfort inside the building and the of cellulose for energy purposes was chosen for this analysis. The objective of the LCA study is the quantification of the environmental impact of the cellulose recovered by Fater S.p.A., used in two distinct processes: insulation panels and biogas production through the anaerobic digestion process.

The life cycle analysis starts with the cellulose recovered after the sanitisation and separation process. This cellulosic fraction, after sterilisation treatment and separation from the organic plastic fraction, is ready for reuse. The boundaries of the system end after the processes of panel production and biogas production by anaerobic digestion.

The functional unit chosen for the case study is 1 kg of cellulose mixed with recovered sorbent.

The cellulose used in the AHP products is combined with materials known for their absorbency properties, such as sodium polyacrylate. When AHPs are recovered, this superabsorbent substance will be found bound to the cellulose in smaller percentages. For this reason, a minimum percentage of this superabsorbent substance (about 3%) has been added to the process included in the calculation software SimaPro. The process related to the thermal insulation panel refers to the realization of a simple model with water. With 1 kg of recovered cellulose, 4 simple panels can be created through a thermo-pressing process. The simplification of the panel produced by mixing cellulose with water has been applied but certain types of coatings are required to resist water absorption, which were not considered in the study.

The biogas produced comes from the anaerobic digestion of the recovered cellulose and the inoculum. For simplicity, an existing process was taken and adapted to the case study. The LCA study is a temporal, i.e. all flows and processes are included as if everything happened at the same time: the comparison of the scenarios of the two possible destinations of the recovered cellulose fraction do not take into account the time needed for the anaerobic digestion, as well as the lifetime of the insulation panels. The study stops at the final production of the two different products. Transport, storage and distribution were neglected, because it was decided to limit the evaluation to the realisation of the final product.

4.4.2 Evaluation methods

Life cycle impact assessment (LCIA) calculations were performed in SimaPro 8.

For the life cycle environmental impact assessment (LCIA), ReCiPe 2016 (H) Midpoint/Endpoint were applied for its use in previous literature (Somers et al., 2021; Velasco Perez et al., 2021) also the Ecological Scarcity 2013, and IPCC 2013 methods have been used.

For each method these impact categories have been considered:

Ecological Scarcity 2013

- *Main air pollutant*
- *GWP100a*
- *Mineral resources*

IPCC 2013

- GWP_{100a}

ReCiPe 2016 Midpoint (H)

- *Terrestrial ecotoxicity*
- *Human non-carcinogenic toxicity*
- *Global warming*

ReCiPe 2016 Endpoint (H)

- *Human Health*

- *Ecosystem*
- *Resources*

4.4.3 Life cycle inventory Analysis

The data used for the LCA study are partly derived from the literature (to identify cellulose mixed with sorbent) (Cordella et al., 2015) (Paolini et al., 2018) and partly from existing databases for the biogas production process by anaerobic digestion during other experimentation of the ReCePiT project.

The Table 34 shows the main inputs and outputs for each process.

Table 34 Main input and output of two processes: Insulation panel production and anaerobic digestion

Insulation panel production			**Anaerobic digestion**		
input			**input**		
Flow	**Unit**	**Amount**	**Flow**	**Unit**	**Amount**
Dry recovered cellulose	kg	0.97	Inoculum	kg	54.54
Sodium polyacrylate	kg	0.03	cellulose	kg	1
Demineralised water	l	3	**output**		
thermoforming, with calendering	kg	1	**Flow**	**Unit**	**Amount**
output			Digestate	kg	52.27
Flow	**Unit**	**Amount**	biogas	kg	0.68
Insulation panel	kg	1	CH4	kg	0.005

4.4.4 Results

The life cycle assessment analysis was carried out comparing insulation panels production and biogas production through the anaerobic digestion process, applying different methods for evaluating the impacts.

On the basis of Ecological Scarcity 2013, the use of recovered cellulose for biogas production is more impactful due to higher releases of pollutants and PM into the atmosphere than the panel making process (Figure 74).

Focusing only on GHG emissions into the atmosphere using the IPCC 2013 method, expressed in CO_2 eq, the panel making process is less impactful (Figure 75).

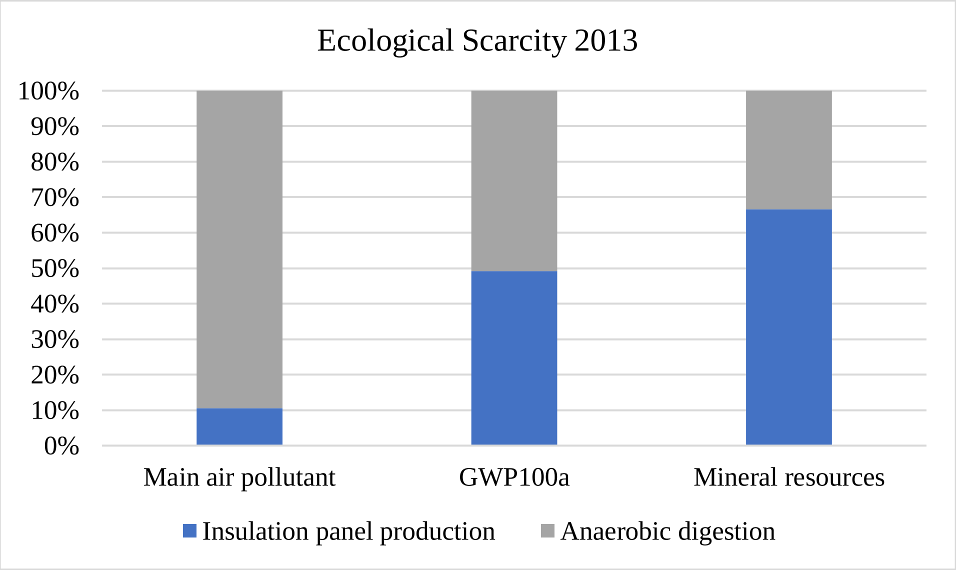


Figure 74 Ecological Scarcity 2013 method: Contribution in each impact category for insulation panel production compared to anaerobic digestion process scenario

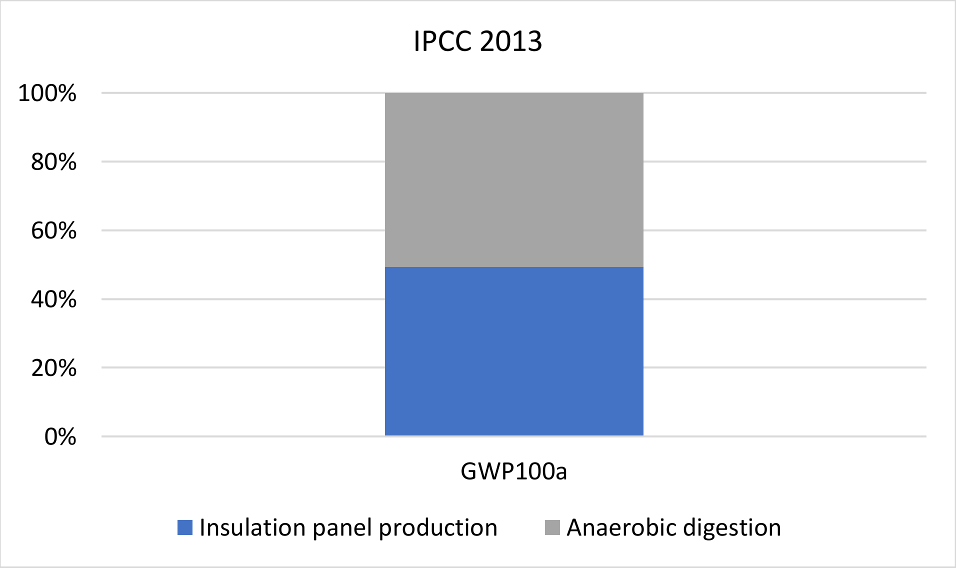


Figure 75 IPCC 2013 method: Contribution in each impact category for insulation panel production compared to anaerobic digestion process scenario

The results of method ReCiPe 2016 Midpoint (H) in the Figure 76 show that the anaerobic digestion of cellulose has a low impact in terrestrial ecotoxicity and human non-carcinogenic toxicity categories, while the insulation panel production causes less impact on the global warming category. Otherwise, the application of

ReCiPe 2016 Endpoint (H) method shows a low impact on the human health and ecosystem for the production of insulation panel (Figure 77).

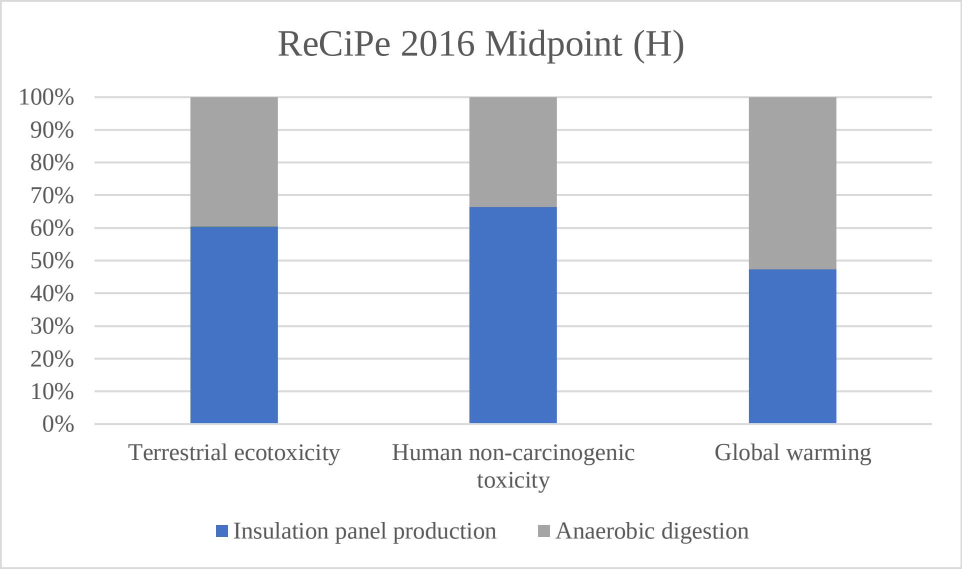


Figure 76 ReCiPe Midpoint (H) method: Contribution in each impact category for insulation panel production compared to anaerobic digestion process scenario

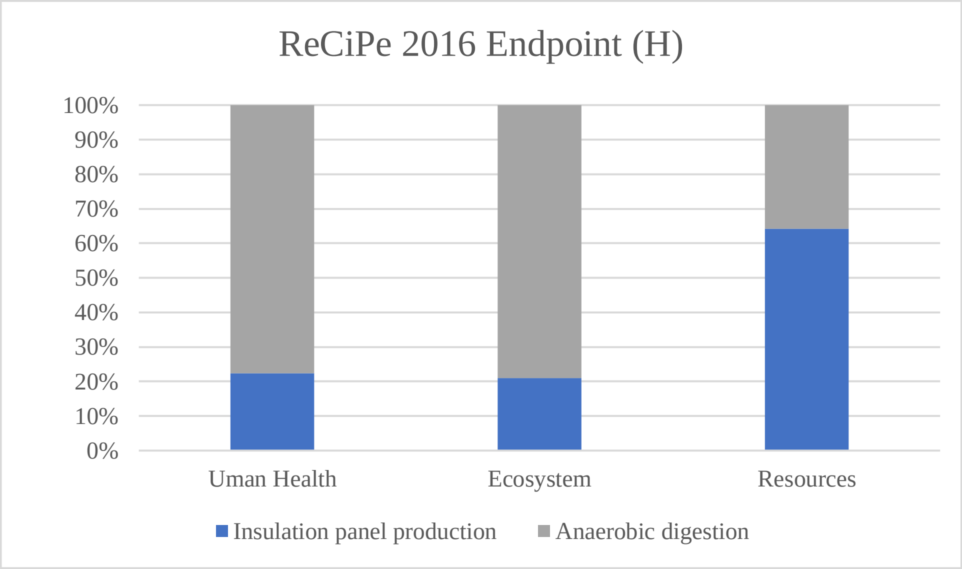


Figure 77 ReCiPe Endpoint (H) method: Contribution in in each impact category for insulation panel production compared to anaerobic digestion process scenario

5 Conclusions

The research carried out over the last three years has assessed some environmental aspects of three types of waste that are currently not recycled or cannot be recycled for non-technological barriers.

The research was developed mainly thanks to public funds obtained through funding from the current Ministry of Ecological Transition and the Lazio Region, only a small private contribution from Zetadì (private) was made. These funds have developed new results with a view to circular economy as envisaged by the objectives set by the European Union.

Indeed, the European Union has now fully embarked on the path towards a circular economy model and has issued a package of directives to support the transition from the linear economy model. This path is explained by the fact that we are now aware of the limited resources and there is widespread concern about the drastic increase in the consumption of raw materials globally, which has occurred in the last century.

The first case study focused on two construction and demolition wastes: Asbestos Containing Materials (ACM) and Man Made Vitreous Fibres (MMVF). Emissions released from the treatment of hazardous and non-hazardous special waste containing asbestos and vitreous fibres were assessed for the first time.

In the first campaign, the most important result regards the potential risk that asbestos fibres are released into the atmosphere during the thermal treatment. Data obtained in this study demonstrate the absence of asbestos fibres in the gaseous emission. Of course, the solid residue obtained in this study was analysed as well, confirming the absence of asbestos fibres in the obtained by-product.

Several other atmospheric pollutants were also investigated for the first time both for the asbestos cement inertisation process and for mineral wool vitrification process. The emissions of the monitored pollutants are very different for the two processes.

In the first case, the laboratory prototype is a controlled process, the composition of the package was known and relatively low concentrations were observed. In contrast, the composition of big bags in the vitrification process is not perfectly known a priori. The presence of residues of plastics, rubbers, glues, etc. could have affected on the emissions. In fact, for the monitoring campaign of the pre-industrial furnace, big bags containing real waste were used, while for the measurements in the laboratory furnace, big bags were specially prepared for the campaign.

Furthermore, during the measurement on the pre-industrial prototype were registered a lot of transients.

The transients registered probably depend on:

- The absence of control in the combustion chamber;
- The loading process of the material in the combustion chamber that has not been homogeneous.

In addition, incomplete combustion probably due to frequent plant transients may have caused higher values of some pollutants such as the high concentrations of pyrene and benzo(g,h,i)perylene recorded.

A burner was installed in the pre-industrial prototype but it was undersized, because in both cases the measurements were carried out in order to assess the emissions as such without the presence of abatement.

For Asbestos-cement treatment, another important finding of this study is the role of the chlorinated coating used during the removal of asbestos cement. This coating is a vinyl resin, consisting of vinyl chloride and vinyl acetate copolymers. Since the presence of polyvinylchloride can strongly increase the emission of chlorinated VOCs and dioxins (PCDD/Fs and dl-PCBs), the use of alternative coatings should be recommended.

The first study also confirmed that there is an influence on emissions based on the input feedstock components. Indeed, the comparison between two blends of waste (asbestos cement with and without polyurethane, samples C and CP) confirmed that the emission of PAHs and non-halogenated VOCs is mainly related to the combustion of carbon-containing components (polyurethane, in this study), rather than to asbestos cement.

More in general, this study highlighted several advantages of the thermal inertisation of asbestos cement compared to landfilling thank to LCA results, in particular the thermal inertisation reduces the human toxicity - non carcinogenic, total human toxicity and impact on human respiratory effects up to 250% less than landfill disposal.

Furthermore, emissions of pollutants into the atmosphere can be further reduced by means of abatement systems. Instead, asbestos landfilling would lead to the release of asbestos fibres whose health impact is higher and would increase in the next years, being a burden on future generations. It is also worth noting that asbestos inertisation plants have a lower soil consumption compared to landfills.

Currently, the decision to move from landfilling to inertisation has already been taken in Europe, and landfills availability is decreasing: thus, the goal of next researches should be focused on the identification and the assessment of the most sustainable inertisation technologies.

With this regard, the potential recycling of the solid residue both for asbestos cement both for mineral wool is another key point, avoiding the consumption of renewable resources and promoting circular economy, which would be a paradigm shift compared to landfilling.

Another very important topic addressed in this PhD was the recycling of domestic hard plastics; a characterization of post-consumer hard plastic and its blends with post-industrial PP waste plastic was provided.

As expected, post-consumer hard plastic is very heterogeneous compared to post-consumer plastic bottles; significant amounts of polypropylene, polyethylene (both high and low density), other plastic polymers and inclusions were found in the studied PC samples. Even after twin screw extrusion and injection molding, a true homogenization could not be achieved, as highlighted by the high standard deviation values for thermogravimetric tests and mechanical tests. Consequently, several mechanical properties of the blends could not be predicted, with the interesting exceptions of the hardness and Vicat tests, for which empirical relations were found.

Such variability may lead to difficulty in the design of custom blends for specific uses. In order to meet the needs of the recycled plastic industry, this study showed that even slight additions of post-industrial plastic can improve the mechanical properties of the blend, e.g., tensile and impact strength: the 78% post-consumer blends showed the most promising results, quite close to pure post-industrial PP, a direct increase in hardness with increases in PI content, and a softening temperature increase of 0.2 °C for each 1% increase in PI content.

This research highlighted some difficulties in treating recovered hard plastic, but also identified some useful approaches to improve the overall quality of the blend. Specifically, twin screw extrusion treatment and blending with pure waste PP may result in an exploitable, valuable secondary raw material. Moreover, an LCA analysis showed the environmental benefits deriving from the industrialization of this process, blending recycled plastic with pure industrial plastic residues to reach performances comparable to virgin plastic while, at the same time, markedly reducing fossil hydrocarbon use, greenhouse gas emissions and water depletion.

This result opens the way to potential applications that are already cost-effective; indeed, the lighter fraction of hard plastic, being mostly PE and PP, can be separated by simple flotation (Turku et al., 2017), thus making available a very large quantity of plastic waste. The mechanical properties of the more promising blends of PC and PI suggest potential uses in the automotive industry (Ajorloo et al., 2021) or for bulky, simple hard plastic items such as baskets, boxes, bins or gardening supplies.

The third case study has taken in account the possibility to recovery the cellulose from Absorbent Hygiene Products (AHP), starting from patent on treatment of this waste implemented by FATER S.p.A. enterprise.

The cellulose is usually made from recycled paper fibres such as unsold newspapers or recovered from waste collection; while in this study the possibility of using cellulose from AHP has been assessed and the insulation properties of cellulose were evaluated.

The energy efficiency of the building and thermo-hygrometric comfort can ensure the full wellbeing of all occupants of any indoor environment. Considering that 30-40% of the world's energy consumption is related to the building and construction sector (Lopez Hurtado et al., 2016b) and the new European directives aim to reduce consumption and reduce environmental impacts, there is a greater need to implement suitable solutions to achieve the objectives.

Cellulose is one of various commercial insulation materials. It is an environmentally friendly with thermal properties similar to other non-renewable insulation materials. In this project, cellulose is obtained from secondary raw materials favouring the circular economy process.

The study evaluated the insulating properties and thermo-hygrometric comfort of cellulose produced by AHP. The monitoring of microclimate parameters shown that the insulated room compared to the uninsulated room registered lower humidity and a constant temperature, also the analysis of thermal exchanges has indicated performances suitable for the regulations in force in Italy.

In addition, the LCA study confirmed a good result of the two scenarios investigated: the insulation panel production process and the anaerobic digestion process. Both methods of reusing cellulose recovered from AHP have a low impact on the environment. By recovering cellulose from AHP materials, resources that can still be reused could be cut out from final disposal. For the same functional unit and approach, biogas production provided slightly more positive environmental impacts than the analysis of the panel making process.

Despite this, as indicated in the previous chapters, the cellulose has some disadvantages that reduce the application as insulation material. However, a good knowledge of the advantages and limitations of this product could ensure its applicability in certain contexts more than others, e.g., in places where only thermal comfort needs to be optimised, thus contributing to the reduction of global warming.

The three case studies have provided good results on the environmental impacts of recycling these categories of waste through the applied technologies; notwithstanding in this context the economic aspect and so the competitiveness compared to traditional solution has not been taken into consideration, e.g., the economic advantages of the inertisation and marketing of the product with respect to the asbestos cement landfills disposal. It is possible to consider an economic advantage obtained from the sale of secondary raw materials, but a more in-depth study is needed to assess the economic benefits in detail.

6 Future perspectives

The three case studies have a good technological basis for their pre-industrialisation or industrialisation. As indicated in the new action plan for the circular economy for a cleaner and more competitive Europe, assessing the possibility of secondary materials to compete with raw materials is important, both in terms of safety, but also in terms of performance, availability and cost.

Therefore, it is essential to proceed in steps and to address possible developments for each case study.

With regard to the first case study, the future prospects are very promising. Zetadì has already developed a pre-industrial prototype for the treatment of mineral wool and is planning an industrial plant for its treatment. Equally important are the prospects for the treatment of asbestos-containing materials. Zetadì has developed a project of an industrial plant on the basis of EP2027943B1 patent in this regard, but since the EIA permit was not granted, the process has been stopped. With regard to KRY·AS, a product obtained from the intertisation of asbestos through this patent, numerous tests have been carried out to verify the safety of the product thanks to the studies conducted by Professor Gualtieri, but today an "end of waste" decree has not yet been approved.

In the near future, the methodology applied on asbestos could be tested on other materials containing asbestos that are currently destined to landfills, such as insulating materials containing asbestos or incoherent materials containing asbestos from reclamation also of disused production plants: dust, sludge, garbage, screeches, excerpts. While in the coming years as regards the methodology tested on waste containing asbestos it could be applied at a higher TRL Technological Readiness Level in the next years for the development of an industrial plant. As already discussed in this study, the environmental aspects were evaluated rather than the economic feasibility of a possible plant and the revenues obtained from the sale of the product, which could be evaluated in the future; furthermore, other relevant aspect to taking in account is the social acceptance of the use and treatment of asbestos containing waste (ACW), and the overcoming current regulatory barriers so that the product is not considered a waste.

Another future perspective will be developed if the project presented by CNR-IIA with the collaboration of UNIMORE will be funded under the POR CALABRIA FESR 2014/2020 action 1.3.2.b "support for the generation of innovative solutions to specific problems of social importance, also through the use of open innovation environments such as living labs". The project will demonstrate the feasibility of removing an asbestos cement surface with a new roof equipped with a photovoltaic system in order to demonstrate the possibility of creating an SEU (Efficient User System) and reusing inert asbestos cement within a local cement supply chain.

The second case study underline those hard plastics are currently not recycled unlike plastic packaging, but the results of possible applications are promising. A further future aspect of the investigation could be focused on the presence of chemical substances in plastics matrix to ensure a safe product such as antioxidants and stabilisers, flame retardants, heat stabilisers, impact modifiers, plasticizers, coupling agents, colourants and pigments, e.g., phthalates, which could be released into the environment. The first step for the assessment of hazardous chemicals could be the identification of investigation analytical procedure suitable for the study of additives and the second step could be cover the selection of classes of substances to be investigated, up to developing material ageing tests to assess the possible release of substances over time.

Relevant regulatory aspects to be overcome are the absence of an end-of-waste decree and so the possibility of using them as a secondary raw material. In the Italian context, the Legislative Office of the Ministry of Ecological Transition is preparing an End of Waste Decree for mixed plastics currently destined for energy recovery, that is not considered a recycle process under the article 205-bis, paragraph 4, letter b, of Legislative Decree 152/06. The path appears to be very long, but it is necessary that plastics will not continue to be used only for energy purposes, but that legislation will favour their use for recycling to produce other products.

On the other hand, with regard to the last issue, the main barrier to the recycling of domestic hard plastic waste not included in packaging waste, and therefore not accepted for example by Corepla Consortium, is represented by their heterogeneity (both in terms of composition and size and hardness) and often from the lack of information about the composition (type of polymer or their mixtures), the possible presence of additives and / or components of different materials (e.g. metal).

In fact, hard plastic does not have an organized and structured recycling system, unlike packaging plastic which has a specific recovery chain manned in Italy by the Corepla Consortium. For example, the ANCI-CONAI agreement provides the possibility for the Municipalities, or third parties delegated by them, to sign an agreement with the CONAI system - Supply chain consortia which commits the Municipalities to carry out the separate collection of packaging waste and confer the collected materials to the Consortia, in accordance with the procedures set out in the specific technical annexes. The Consortia guarantee the collection of the material, the subsequent recycling and the recognition of an economic consideration based on the quantity and quality of the urban waste collected.

Considering that the results of the second case study open up possible applications of recycled hard plastics and there are no cultural problems to overcome in this case study, opening a discussion table on regulatory and management issues, such as the extension of contracts with consortia, could be a way to start a path towards recycling a fraction of waste destined for landfills. Recycling would reduce tonnes of plastic waste; moreover, it would make hard plastics a resource and thus discourage abandonment.

On the other hand, the third and last case study has an excellent technological and non-technological advancement. The sterilisation process has been patented by Fater S.p.A. and the End of Waste decree (DM 15 maggio 2019, n. 62 “Regolamento recante disciplina della cessazione della qualifica di rifiuto da prodotti assorbenti per la persona (PAP), ai sensi dell’articolo 184-ter, comma 2, del decreto legislativo 3 aprile 2006, n. 152”) has been issued allowing the re-use of cellulose as a secondary raw material from AHP treatment.

Subsequent developments may be linked to the use of these materials by Italian and foreign companies for the production of insulating panels or the blowing of flakes as insulation inside cavities. This last process would require the cellulose to be treated for marketing to guarantee the safety of the product, an aspect that was not evaluated during the project. So, further research developments could focus on the use of products such as flame retardants or anti-mould or protect it from rodent and insect attack with low environmental impact, while another aspect to be investigated could be the study of new type of product, cellulose foam, to be obtained from AHP cellulose which could replace products based on petrochemicals such as polystyrene (PS) or polyolefins (PP, polypropylene or PE, polyethylene).

There is still a need to implement a collection chain for AHP and an assessment of the economic impact on the overall chain, despite this product is already marketable and as discussed in the results of the laboratory tests it could be allocated at first time in the insulating materials market.

On the basis of these future technological developments, another aspect to highlight is the economic impact on the possible increase of investments in the circular economy sector which will give the possibility to the creation of new jobs, also supported by national and European funds such as the NexGeneration EU and the national PNRR. Just think that the Italian PNRR has allocated €2.10 billion for the circular economy sector for the next 6 years (2021-2026). Other funds on the circular economy are allocated by the Ministry of Ecological Transition every year for the last 3 years. the call provides for the submission of research projects aimed at the development of technologies for the prevention, recovery, recycling and treatment of waste not included in the categories already served by supply chain consortia, eco-design of products.

Certainly, these funds will be a driving force to support technological development, knowledge enhancement and the overcoming of non-technological barriers and increase the amount of waste avoided or recycled, thanks also supported by National and European reforms.

7 List of Publications

During my PhD some papers have been published thanks to the research carried out with the collaboration of other researchers.

The Publications are listed below:

Tomassetti, L., Di Giuseppe, D., Zoboli, A., Paolini, V., Torre, M., Paris, E., ... & Gualtieri, A. F. (2020). Emission of fibres and atmospheric pollutants from the thermal treatment of asbestos containing waste (ACW). *Journal of Cleaner Production*, *268*, 122179. https://doi.org/10.1016/j.jclepro.2020.122179

Tratzi, P., Giuliani, C., Torre, M., Tomassetti, L., Petrucci, R., Iannoni, A., ... & Di Carlo, G. (2021). Effect of Hard Plastic Waste on the Quality of Recycled Polypropylene Blends. *Recycling*, *6*(3), 58. https://doi.org/10.3390/recycling6030058

Scognamiglio, V., Di Giuseppe, D., Lassinantti Gualtieri, M., Tomassetti, L., & Gualtieri, A. F. (2021). A Systematic Study of the Cryogenic Milling of Chrysotile Asbestos. *Applied Sciences*, *11*(11), 4826. https://doi.org/10.3390/app11114826

Another paper related to asbestos inertisation technology was published shortly before the official start of the doctorate:

Paolini, V., Tomassetti, L., Segreto, M., Borin, D., Liotta, F., Torre, M., & Petracchini, F. (2019). Asbestos treatment technologies. *Journal of Material Cycles and Waste Management*, *21*(2), 205-226. https://doi.org/10.1007/s10163-018-0793-7

8 Acknowledgements

The first case study - monitoring emissions of laboratory prototype - was supported by the Italian Ministry of Environment and Land and Sea Protection, General Direction for Waste and Pollution (MATTM-DGRIN), under the Agreement RINDEC-20016-4 of November 25th 2015, entitled “Development of a novel methodology for asbestos inertisation, emission abatement and analysis of the supply chain” and under the granted project Decr. Dir. Min. Ric-Dec_2017_0000196, 22/12/17, “Tecnologie di trattamento di rifiuti contenenti amianto (RCA) e riciclo per la produzione di piastrelle ceramiche, nell’ottica di una economia circolare”. Sampling of emissions was carried out by Mattia Perilli and Daniele Borin, Ettore Guerriero. Alessandro Gualtieri carried out the sampling of solid residues. The monitoring of pre-industrial prototype is co-financed by ZetaDi s.r.l. Sampling was carried out by Mattia Perilli, Ettore Guerriero, Marco Torre, Laura Tomassetti. Authors express their gratitude to the staff of ZetaDi s.r.l. for their invaluable technical and logistic support in the both monitoring campaign. Many thanks to the authors of paper: Tomassetti, L., Di Giuseppe, D., Zoboli, A., Paolini, V., Torre, M., Paris, E., ... & Gualtieri, A. F. (2020). *Emission of fibres and atmospheric pollutants from the thermal treatment of asbestos containing waste (ACW)*. Journal of Cleaner Production, 268, 122179.

The second case study is part of is part of the project PLASMARE, funded by the Italian Ministry of the Environment, Land and Sea Protection, grant protocol RINDEC-2017-00196. The project was conducted with CNR-ISMN, CNR-IIA and ESPER srl. A special gratitude to the Municipality of Lanuvio for providing post-consumer hard plastic samples, and to Revet S.p.A. for carrying out the plastic shredding. Many thanks to the authors of paper: “Tratzi, P., Giuliani, C., Torre, M., Tomassetti, L., Petrucci, R., Iannoni, A., ... & Di Carlo, G. (2021). *Effect of Hard Plastic Waste on the Quality of Recycled Polypropylene Blends*. Recycling, 6(3),” 58.

Third case study is part of ReCePit project - “Recupero della Cellulosa per Pannelli Isolanti Termici” - co-financed by the Lazio Region through the "KETS Enabling Technologies Public Notice" (POR FESR Lazio 2014-2020, Axis I - Research and Innovation and Axis 3 – Competitiveness). The project was carried out with Azzero CO_2 and Modoolab srl and CNR-ISMN. A greater thank you to all participants in the project.

Special thanks to Professor Alessandro Gualtieri and Professor Gigliola Lusvardi for their support and the opportunity to collaborate together.

Special thanks to Dr. Francesco Petracchini and Dr. Valerio Paolini my mentors.

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10 Supplementary materials

System	**σ_{yield} (MPa)**	**ε_{yield} (%)**	**σ_{break} (Mpa)**	**ε_{break} (%)**	**E (Mpa)**
PC100-PI0	24.8 ± 1.6	8.4 ± 0.6	19.7 ± 1.5	144 ± 91	**1070 ± 18**
PC92-PI8	26.8 ± 1.5	8.4 ± 0.3	25.3 ± 3.3	11 ± 5	**1242 ± 52**
PC85-PI15	27.7 ± 0.4	8.8 ± 0.5	23.6 ± 2	18 ± 5	**965 ± 78**
PC78-PI22	29.4 ± 1.6	8.2 ± 0.2	25.7 ± 1.7	14 ± 3	**1223 ± 36**
PC70-PI30	26.1 ± 0.9	8.5 ± 0.9	22.1 ± 3.4	22 ± 12	**1092 ± 31**
PC50-PI50	26.5 ± 0.7	7.6 ± 0.4	24.7 ± 2.2	12 ± 4	**1056 ± 27**
PC0-PI100	**31.2 ± 1.2**	**8.7 ± 0.7**	**30.7 ± 2**	**9 ± 1**	**1048 ± 28**

Table S 1 Tensile properties for the developed blends

System	**Impact Energy (KJ/mm^2)**
PC100-PI0	**3.6 ± 0.2**
PC92-PI8	**3.3 ± 0.3**
PC85-PI15	**3.4 ± 0.3**
PC78-PI22	**4.1 ± 0.5**
PC70-PI30	**3.4 ± 0.4**
PC50-PI50	**3.5 ± 0.4**
PC0-PI100	**4.1 ± 0.5**

Table S 2 Impact strength of the developed blends