D2.3

CO_2 capture by selected ashes and LCA

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Table of Contents

Table of Figures

Table of Tables

Abbreviations

CCUS	carbon capture, utilization and storage
Gt	gigaton
LCA	life cycle assessment
L/S	liquid to solid
MSWI-BA	municipal solid waste incineration bottom ash
MSWI-FA	municipal solid waste incineration fly ash
ppm	parts per million
RH	relative humidity
SSA	sewage sludge ash
WB-BA	wood biomass bottom ash
WB-FA	wood biomass fly ash

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Objective

The main objective of this deliverable, as defined in Task 2.3 of the AshCycle project, is to assess the carbon storage capacity (sequestration potential) of the ashes considered in this project, i.e. sewage sludge ash (SSA), municipal solid waste incineration bottom/fly ash (MSWI-BA and MSWI-FA), and wood biomass bottom/fly ash (WB-BA and WB-FA).

Ashes containing a high amount of free CaO represent a potential for binding CO_2 into the stable form of carbonates. The amount of free CaO depends on both the type of fuel and the combustion process. Accelerated carbonation makes the ash more stable and therefore easier to reuse. However, various parameters can significantly influence the process, e.g. particle size, RH, temperature, exposure time and CO_2 concentration. Carbon sequestration is also investigated with regard to carbstone concrete products (Task 4.3, cement-free concrete curing in a CO_2 chamber). Such a process could become part of incineration plants and offers two advantages: stabilised ash and lower CO_2 emissions, which can also be achieved by a long-term use of the ash. The results of the potential for temporary carbon storage are assessed as part of the life cycle assessment (global warming potential indicator).

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Introduction

The concentration of CO_2 in the atmosphere has been below 300 ppm for thousands of years [1]. CO_2 emissions from fossil fuels play an important role in the accumulation of CO_2 in the atmosphere, as the burning of fossil fuels, cement production and other industrial processes release more than 36 Gt of CO_2 worldwide every year [2]. The CO_2 concentration has risen steadily since the beginning of the industrial revolution when it was around 280 ppm [3]. In 2014, the CO_2 concentration in the atmosphere exceeded 400 ppm for the first time and is currently increasing by around 1% per year [4,5]. In 2022, the CO_2 concentration in the atmosphere exceeded 420 ppm, and in May 2023, CO_2 emissions reached a record level of 424 ppm [5]. The increase in CO_2 concentration in the atmosphere leads to global warming and consequently to climate changes such as a rise in the average global temperature, increasing air pollution, changes in the pattern and amount of rain and snow fall, changes in relative humidity, the melting of ice in polar regions, a rise of the average sea level, a decline in agricultural production and the extinction of fauna and flora [6,7].

Due to climate change and environmental degradation in Europe and the rest of the world, there is an urgent need to reduce CO_2 and greenhouse gas emissions. To overcome these challenges, the European Commission adopted the European Green Deal in 2020, commiting the European Union to become climate neutral by 2050 [8]. The European Union's initiatives are aimed at various sectors, including the construction industry, transport and renewable energies. In the first phase, net greenhouse gas emissions are expected to be reduced by at least 55% by 2030 compared to 1990 levels [8]. In the technological segment related to construction, several technologies are currently being developed or improved that will mainly contribute to the first phase. These include various recycling methods and industrial symbioses such as alkali-activation and carbonation as well as carbon capture, utilization, and storage (CCUS) technologies. CCUS is an extremely hot topic in EU climate policy because of its potential role in avoiding CO_2 emissions that are difficult to reduce. Although CCUS technologies have been in use since the 1970s, their widespread implementation is still challenged by a range of factors, including political inertia, high costs and their relative novelty in the public discourse [9]. Despite the current energy and cost-intensive technologies for carbon capture and storage, CO_2 mineral sequestration represents a straightforward approach. It primarily involves the reaction of CO_2 with alkaline materials, composed of Ca and Mg rich (hydr)-oxides and silicates, leading to the formation of solid carbonate products and their subsequent storage [2], [10–12]. The exothermic nature of the mineral carbonation reaction can potentially compensate for the energy consumption in CO_2 sequestration and achieve low costs [13,14].

Carbonation is a natural chemical reaction process in which CO_2 initially dissolves in water. As directly binding molecular CO_2 to CaO is very slow (Equation (1)), it dissociates, and reacts with $Ca(OH)_2$ (also with calcium silicate hydrate (C-S-H), calcium aluminate hydrate (CAH), etc.) to form $CaCO_3$ (Equations (2)-(4)). The main product ($CaCO_3$) is thermodynamically stable, meaning it is unlikely that CO_2 will be released under normal conditions [6,11,15].

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$$CaO + CO_2 \rightarrow CaCO_3 \quad (1)$$

$$CaO + H_2O \rightarrow Ca(OH)_2 \quad (2)$$

$$Ca(OH)_2 + CO_2 \rightarrow CaCO_3 + H_2O \quad (3)$$

$$C - S - H + CO_2 \rightarrow 3\,CaCO_3 + 2\,SiO_2 + 3\,H_2O \quad (4)$$

While spontaneous carbonation with atmospheric CO_2 (0.04%) is generally very slow, carbonation can be accelerated by an increased CO_2 concentration or pressure [11, 16,17]. Accelerated carbonation leads to a higher carbonation rate, which is influenced by the carbonation conditions such as the particle size of the selected material, the relative humidity (RH), the liquid to solid (L/S) ratio, the temperature, the contact time, the pressure and the CO_2 concentration [6,18–20]. Although the accelerated carbonation of industrial residues has been the subject of numerous research studies over the last decade [10,13,21–24], there is still a lack of systematic evaluation for the influence of the relevant operating parameters in the scientific literature. In this context, it is essential to find optimal conditions that maximize the potential of the selected material as a carbon sink. In order to mitigate climate change fast enough, accelerated carbonation is highly favourable over natural carbonation.

Particle morphology seems to be important as CO_2 uptake increases when particle size is reduced due to a larger specific area exposed to CO_2 [6,19,25–27]. Water is essential for the reaction kinetics of carbonation, while the CO_2 concentration in the gas has a smaller influence. Some studies have shown that the reaction rate and the conversion rate of $Ca(OH)_2$ increase with increasing RH [18,28,29], where the optimum RH for the carbonation rate is between 60 and 80% [7,30]. Excess moisture can saturate the material and lower its permeability, which can limit the movement of CO_2 into the material and the depth at which carbonation can occur [31]. Increasing the temperature accellerates the reaction kinetics. Carbonates form quickly on the surface, reaching saturation and limiting the further penetration of CO_2 into the material [32].

Waste incineration is steadily increasing throughout Europe, but there are environmental concerns regarding solid residues which are usually landfilled [33]. There are several options for using the received or pre-treated ash, of which CO_2 sequestration by accelerated mineral carbonation is a promising carbon capture and storage technology. However, not much is known about the sequestration potential of waste ashes. The main problem, which is important for wide utilization, is to keep the properties of the heterogeneous ash constant [34]. Ashes with a high content of Ca and Mg compounds, especially ashes from wood biomass, are promising candidates for sequestration [18,34,35]. The high availability of ash as a by-product of solid fuel combustion is therefore an additional advantage. Recently, Tominc and Ducman [18] have shown that a high CO_2 sequestration capacity for WBA can be achieved by a semi-dry mineral carbonation process.

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1. Methodology

As part of the AshCycle project, we analysed ashes from different incineration plants and determined its potential for carbon sequestration [18], as visualized in Figure 1. The methodology was first applied to eight different ashes and then improved by optimising the carbonation conditions, as not all ashes are equally suitable for CO_2 sequestration.

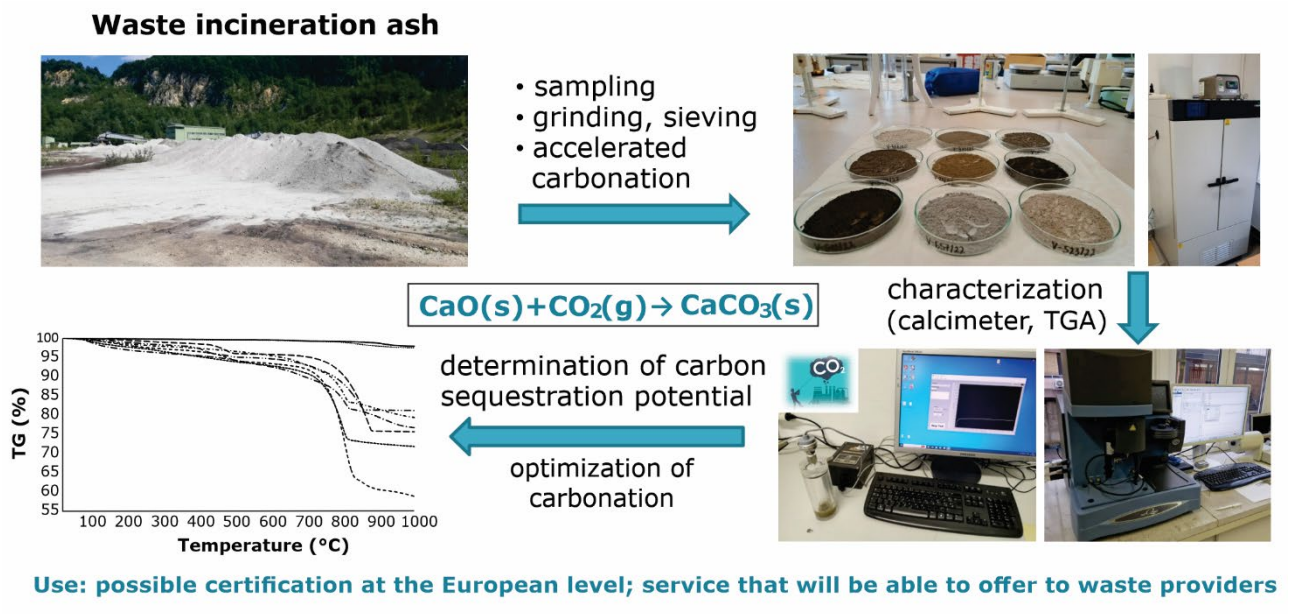

Figure 1: Methodology for evaluating the CO_2 sequestration potential of waste ashes.

1.1. Scope

The methodology was established to determine the carbon sequestration potential of the ashes considered in this project. The evaluation includes sample preparation, characterization and analysis with a pressure calcimeter and a thermogravimetric analyzer.

As a tool, this methodology can also be used to evaluate other waste materials and support the development of construction materials based on the solidification of the material. This enables the long-term use of waste materials and contributes to the reduction of global CO_2 emissions by reducing the need for cement.

1.2. Characterization

1.2.1. Sampling

All samples were homogenized by quartering, packed in a PVC bag, and stored in a plastic container.

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1.2.2. Chemical analysis (XRF)

The ashes were sieved below 125 µm for chemical analyses, dried at 105 °C, and heated at 950 °C to determine their loss on ignition (LOI); a fused bead was then prepared with a mixture of ash and flux (50% lithium tetraborate/50% lithium metaborate) in a 1:10 ratio (0.947 g: 9.47 g) and heated at 1100 °C. The standard deviation of repeatability for LOI is 0.04 mass% according to the standard EN 196-2:2013 [36]. The chemical composition of the ashes was then determined using a ARL PERFORM'X Wavelength Dispersive X-Ray Fluorescence Spectrometer (WDXRF; Thermo Fischer Scientific Inc., Ecublens, Switzerland) with an Rh-target X-ray tube and the UniQuant 5 software (Thermo Fisher Scientific Inc., Walthem, MA, USA). Two measurements were performed for each ash. The average values are listed in the Appendix.

1.2.3. Mineralogical analysis (XRD)

The ashes were sieved below 63 µm and placed in 27 mm holders (in diameter) for mineralogical analyzes performed before and after CO_2 exposure with X-ray diffraction (XRD;Empyrean X-ray Diffractometer, Cu X-ray source; PANalytical, Almelo, The Netherlands) in 0.013° steps from angles of 4–70° under clean room conditions, using the external standard corundum NIST SRM 676a. The mineral content was evaluated using the PANalytical X'Pert High Score Plus diffraction software v.4.8.

1.3. Mineral carbonation

1.3.1. Sample preparation

The obtained ash was first ground and sieved to a particle size of less than 125 µm. Then 20 g of each were placed in a petri dish and exposed in a closed CO_2 carbonation chamber.

1.3.2. Carbonation conditions

Selected ashes were first tested at a RH of 50-55% at a temperature of 20 °C and 4 ± 0.1 vol% CO_2. Then the ashes were tested at an increased RH of 80-85% at a temperature of 20 °C and 4 ± 0.1 vol% CO_2. Samples were taken after 1, 7, 14, 21 and 28 days. To optimize carbonation conditions, selected ashes were treated with 20 ± 0.1 vol% CO_2 for 3 days and with 4 ± 0.1 vol% CO_2 for at least 7 days at a RH of 80% and a temperature of 40 °C.

1.3.3. Determination of the constant mass

Carbonation was complete when a constant mass was reached. The mass is constant if the difference between two weighings is less than 0.1 g.

1.4. Analysis with a pressure calcimeter

1.4.1. General

The obtained and carbonated ashes were analysed with the pressure calcimeter (OFITE Calcimeter, OFI Testing Equipment Inc., Houston, TX, USA, according to ASTM D 4373) with an analytical error of <5%.

The OFITE calcimeter is used to determine the amount of $CaCO_3$ and Ca/Mg carbonate (dolomite) in a sample. $CaCO_3$ reacts with 10% HCl in a closed reaction cell and forms $CaCl_2$, CO_2 and H_2O. The pressure of the released CO_2 is measured with a manometer. The calcimeter is calibrated by reacting HCl with pure $CaCO_3$ before the actual measurements.

1.4.2. Test procedure

Each sample was ground and sieved below 125 µm and dried in an oven at 105 °C for 24 h. Then 1.0 ± 0.01 g of the sample was weighed and added to the reaction cell. The acid cup was filled with 20 mL of 10% HCl and carefully placed in the reaction cell. The vent valve was opened until the pressure was zero, then the vent valve was firmly closed to start the measurement. The reaction cell was swirled for one minute and then held until the reaction was complete, i.e. after 40 minutes. The percentage of $CaCO_3$ and dolomite in the sample was calculated using the software.

1.4.3. Construction of the calibration curve

The volume of a calcimeter reaction cell determines the relationship between the pressure increase and the amount of released CO_2. This relationship is constant for a given reaction cell. The calibration is required before the first test. The software uses a calibration curve to convert the released pressure into a percentage of $CaCO_3$.

Five sets of $CaCO_3$ samples with the following masses were used for calibration: 0.2 g, 0.4 g, 0.6 g, 0.8 g, 1.0 g. All samples were weighed with a margin of error of ±0.01 g. At the end of the calibration, the software displayed the r^2 value as shown in Figure A1.

1.4.4. Calculations

The calculations for the amount of released CO_2 were based on the stoichiometry.

$$CaCO_3 + 2HCl \rightarrow CaCl_2 + CO_2 + H_2O \quad (5)$$

1mol : 2mol ------ 1mol : 1mol : 1mol

$n(CaCO_3)=n(CO_2)=1:1$

$M(CaCO_3)=100.0869$ g/mol; $M(CO_2)=44.01$ g/mol

1 mol of $CaCO_3$ releases 44.01 g of CO_2.

$$CaMg(CO_3)_2 + 4HCl \rightarrow MgCl_2 + CaCl_2 + 2CO_2 + 2H_2O \quad (6)$$

$n(CaMg(CO_3)_2))=n(CO_2)=1:2$

$$M(CaMg(CO_3)_2)) = 184.4008 \text{ g/mol}$$

1 mol of $CaMg(CO_3)_2$ releases 88.02 g of CO_2.

1.5. Thermogravimetric analysis (TGA)

1.5.1. General

A thermogravimetric analysis (TGA) was performed on carbonated ashes using a TGA Q5000IR thermal analyzer (TA Instruments, New Castle, Delaware, USA).

The TGA measures the mass change of a sample when it is subjected to a temperature program in a controlled atmosphere [37]. Due to possible hydration reactions contributing to the weight gain, TGA is a suitable method to quantify the increase in $CaCO_3$ content during carbonation and provides quantitative information on the extent of carbonation [16,19].

1.5.2. Test procedure

TGA was performed from 25 to 1000 °C, with a heating rate of 10 K min^{-1}. Prior to the measurements, the ashes were dried at 105 °C and sieved to a grain size below 63 µm. To prevent oxidation during the measurement, the sample chamber was filled with N_2 with a flow rate of 25 mL min^{-1}. Ash batches were placed in 100 µL Al_2O_3 crucibles.

Using the TGA, we measured the weight loss in the temperature range of decomposition of the carbonate mineral (between 550-950 °C), with an analytical error of <1 %. The results were analyzed using TA Universal Analysis 2000 v.4.5A (TA Instruments, New Castle, Delaware, USA).

2. Results

The results of the assessment of the sequestration potential of the individual ashes are given separately for each country of origin. The ashes were given a systematic name: #country.type.number. Details of the selected ashes were provided in Deliverable 2.1: Ash characterization and categorization.

The ashes were exposed to accelerated carbonation conditions in a closed carbonation chamber at elevated CO_2 concentration and different humidities and temperatures until maximum CO_2 uptake. Maximum CO_2 uptake was achieved at room temperature and elevated humidity (80%) after 28 days of carbonation. By optimizing the carbonation conditions, maximum CO_2 uptake was achieved after 7-14 days of carbonation at elevated temperature (40 °C) and humidity (80%) for the selected ashes.

The highest sequestration potential was found for ashes from wood biomass from Slovenia, Croatia and the Netherlands as well as from co-combustion of wood waste and paper sludge from Slovenia (#SI.CC.MA.2), while SSA or MSWI ashes showed a lower sequestration potential.

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2.1. Slovenia

In Table 1 CO_2 content (in wt%) according to TGA and measured with a pressure calcimeter is compared. The maximum CO_2 content of both methods in each ash is shown in bold. As we can see, the results in ashes from wood biomass are very comparable, in the case of ash #SI.WA.BA.1 the difference between the determination by TGA and the calcimeter is 1.4%. As shown in Figure 2a, the weight loss in the temperature range of decomposition of carbonate minerals was between 550-950 °C, while Figure 2b shows that we detect different phases in this temperature range, as the difference between the calcimetric measurement and TGA is up to 22.5%. If we narrow down the weight loss in the temperature range of 400-700 °C, we obtain a comparable result to the calcimetric measurement (9.7 wt%).

Due to these differences between the TGA and the calcimetric determination, the CO_2 sequestration capacity of all ashes was quantified for further LCA calculations using the calcimetric method. With TGA, it is necessary to limit the temperature range for each ash individually. Of the Slovenian ashes, the WB-BA and the co-combustion mixed ash achieved the highest sequestration potential with a CO_2 content of 30.2 wt% (#SI.WA.BA.1) and 27.6 wt% (SI.CC.MA.2).

Table 1: TGA and calcimetric measurements for wood and co-combustion ashes from Slovenia.

Sample ID	CO_2 conc. (%)	T (°C)	RH (%)	Time (days)	TGA (weight losses-%)		Calculations (TGA)		Calculations (calcimeter)		
					0-150 °C	550-950 °C	% dry matter	% CO_2/dry matter	% $CaCO_3$	% Dolomite	% CO_2
#SI.WA. FA.1	4	20	50	28	2.9	16.8	97.1	17.3	27.1	4.4	14.0
	4	20	80	28	2.4	17.6	97.6	**18.0**	33.0	3.1	**16.0**
	4	40	80	7	2.0	20.3	98.0	20.7	26.1	4.7	13.7
	4	40	80	14	2.0	21.0	98.0	21.5	27.0	5.2	14.4
#SI.WA. BA.1	4	20	50	28	3.2	26.1	96.8	26.9	52.0	0.5	23.1
	4	20	80	28	4.6	23.2	95.4	24.4	67.7	1.0	30.2
	4	40	80	7	2.8	27.9	97.2	28.7	66.7	1.1	29.8
	4	40	80	14	2.4	28.1	97.6	**28.8**	67.8	0.8	**30.2**
	20	40	80	3	3.4	26.8	96.6	27.7	58.1	0.7	25.8
#SI.CC. FA.2	4	20	50	28	1.6	11.8	98.4	12.0	13.8	0.3	6.2
	4	20	80	28	2.0	15.3	98.0	**15.6**	23.5	0.0	**10.3**
	4	40	80	7	1.3	12.7	98.7	12.9	19.2	0.3	8.6
	4	40	80	14	1.1	14.6	98.9	14.8	20.6	0.2	9.1
#SI.CC. MA.2	4	20	50	28	0.3	20.2	99.7	20.2	44.1	1.8	20.2
	4	20	80	28	1.3	28.8	98.7	29.2	59.9	1.6	27.1
	4	40	80	7	0.6	27.3	99.4	27.4	59.3	2.9	27.4
	4	40	80	14	1.5	27.0	98.5	**27.4**	61.3	1.3	**27.6**
	20	40	80	3	0.5	23.9	99.5	24.1	50.9	2.4	23.5
#SI.CC. FA.3	4	20	50	28	2.7	13.5	97.3	13.9	11.3	0.3	5.1
	4	20	80	28	2.9	15.8	97.1	**16.3**	22.1	0.0	**9.7**
	4	40	80	7	2.2	14.1	97.8	14.5	17.4	1.3	8.3

	4	40	80	14	2.2	15.8	97.8	16.2	19.1	0.7	8.7
	20	40	80	3	1.9	13.6	98.1	13.8	15.5	0.5	7.0
#SI.CC.BA.3	4	20	50	28	0.3	12.0	99.7	12.0	23.7	3.4	12.0
	4	20	80	28	0.7	13.7	99.3	13.8	30.5	1.3	14.0
	4	40	80	7	1.0	12.2	99.0	12.3	31.1	2.3	14.8
	4	40	80	14	1.0	12.7	99.0	**12.8**	34.4	1.7	**15.9**
#SI.CC.FA.4	4	20	50	28	2.5	31.5	97.5	**32.3**	19.5	2.6	**9.8**
#SI.CC.BA.4	4	20	50	28	1.6	11.4	98.4	11.5	29.4	6.2	15.9
	4	20	80	28	1.7	12.9	98.3	13.2	28.0	15.8	19.9
	4	40	80	7	1.9	10.8	98.1	**11.0**	26.5	17.8	**20.2**
	4	40	80	14	1.8	10.8	98.2	11.0	27.9	16.4	20.1

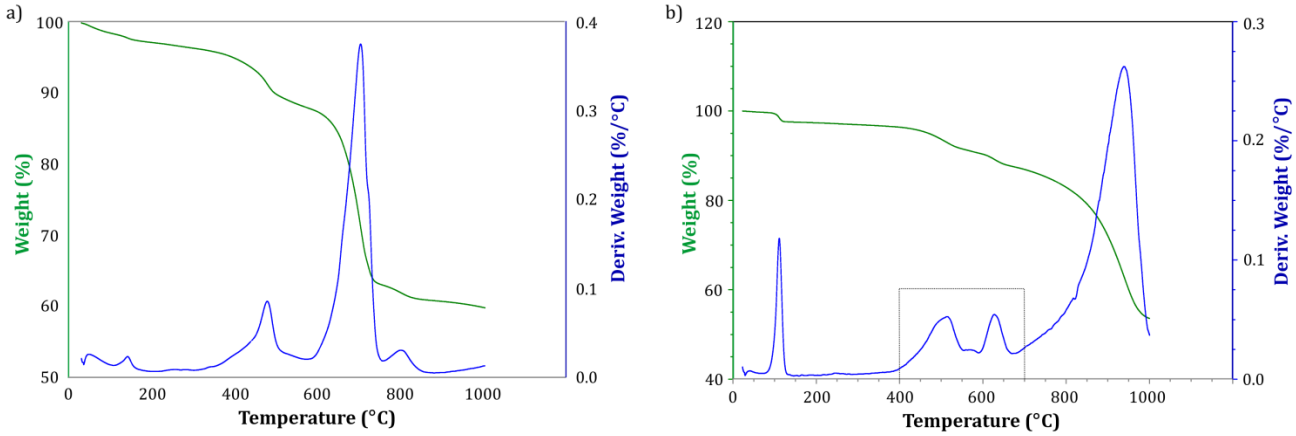

Figure 2: Thermogravimetric analysis of selected sample **a)**#SI.WA.BA.1 with the maximum CO_2 content after 14 days of carbonation in the temperature range from 550 to 950 °C and **b)** #SI.CC.FA.4 after 28 days of carbonation in the temperature range from 400 to 700 °C.

2.2. Croatia

Table 2 compares the CO2 content according to TGA and measured with a pressure calcimeter in the ashes from Croatia. As we can see, the results for ashes from wood biomass are again very comparable, only for SSA there are higher deviations. Seven ashes from wood biomass showed a high CO2 sequestration potential (with bound CO2 above 20 wt%), with the highest value for #SI.WA.11 being 30.4 wt%.

Table 2: TGA and calcimetric measurements for WBA and SSA from Croatia.

Sample ID	CO_2 conc. (%)	T (°C)	RH (%)	Time (days)	TGA (weight losses-%)		Calculations (TGA)		Calculations (calcimeter)		
					0-150 °C	550-950 °C	% dry matter	% CO_2/dry matter	% $CaCO_3$	% Dolomite	% CO_2
#HR.WA.1	4	20	50	28	2.4	19.5	97.6	19.9	38.4	0.3	17.0
	4	20	80	28	2.1	20.3	97.9	20.7	48.2	0.0	21.2
	4	40	80	7	2.4	21.1	97.6	21.6	47.0	1.7	21.5
	4	40	80	14	1.9	20.4	98.1	**20.8**	49.8	0.4	**22.1**
	20	40	80	3	2.0	19.0	98.0	19.4	46.3	0.6	20.6
#HR.	4	40	50	28	0.3	17.9	99.7	18.0	45.4	0.0	19.9

WA.2	4	20	80	28	0.9	24.0	99.1	**24.2**	61.9	0.0	**27.2**
#HR. WA.3	4	20	50	28	0.5	13.7	99.5	13.8	24.9	0.0	11.0
	4	20	80	28	0.6	22.4	99.4	**22.5**	45.1	0.0	**19.8**
#HR. WA.4	4	20	80	28	0.5	24.2	99.5	**24.3**	59.4	2.1	**27.1**
#HR. WA.6	4	20	80	28	0.7	16.9	99.3	**17.0**	35.8	3.1	**17.2**
#HR. WA.7	4	20	80	28	0.3	7.9	99.7	**8.0**	17.1	1.8	**8.4**
#HR. WA.8	4	20	80	28	0.7	23.8	99.3	**23.9**	53.2	2.6	**24.6**
#HR. WA.9	4	20	50	28	0.5	18.2	99.5	18.3	35.6	0.0	15.6
	4	20	80	28	0.8	26.6	99.2	**26.8**	55.7	0.0	**24.5**
#HR. WA.11	4	20	50	28	3.3	25.6	96.7	26.5	51.8	0.5	23.0
	4	20	80	28	5.2	26.9	94.8	**28.4**	69.2	0.0	**30.4**
#HR. WA.15	4	20	80	28	0.6	24.7	99.4	**24.8**	56.0	1.0	**25.1**
#HR. WA.16	4	20	80	28	0.7	17.7	99.3	**17.8**	40.6	0.8	**18.2**
#HR. SSA.1	4	20	80	28	1.7	16.9	98.3	**17.1**	10.4	0.8	**4.9**
	4	40	80	7	1.6	15.8	98.5	16.1	9.5	1.0	4.7
	4	40	80	14	1.7	15.4	98.3	15.7	9.9	0.8	4.7
	20	40	80	3	1.8	11.1	98.2	11.3	9.0	0.8	4.3
#HR. SSA.2	4	20	80	28	0.7	5.6	99.3	**5.7**	7.0	0.8	**3.4**
#HR. SSA.3	4	20	80	28	0.6	2.4	99.4	**2.4**	2.0	0.0	**0.9**
#HR. SSA.4	4	20	80	28	0.9	2.7	99.2	**2.7**	1.7	0.3	**0.9**

2.3. Denmark

MSWI ashes and sewage sludge ashes from Denmark did not show a high CO_2 sequestration potential, with the highest value of 5.9 wt% CO_2 for #DK.MSWI-FA.1.

Table 3: TGA and calcimetric measurements for MSWI ashes and SSA from Denmark.

Sample ID	CO_2 conc. (%)	T (°C)	RH (%)	Time (days)	TGA (weight losses-%)		Calculations (TGA)		Calculations (calcimeter)		
					0-150 °C	550-950 °C	% dry matter	% CO_2/dry matter	% $CaCO_3$	% Dolomite	% CO_2
#DK.MSWI-FA.1	4	20	80	28	1.5	13.3	98.5	**13.5**	12.6	0.8	**5.9**
	4	40	80	7	1.8	11.8	98.2	12.0	10.8	1.6	5.5
	4	40	80	14	1.6	11.5	98.4	11.7	11.6	1.2	5.6
	20	40	80	3	1.8	14.3	98.2	14.5	12.0	1.2	5.8
#DK.SSA-FA.1	4	20	80	28	0.3	0.5	99.7	**0.5**	0.6	0.0	**0.2**
#DK.SSA-FA.2	4	20	80	28	0.2	0.8	99.8	**0.8**	0.8	0.0	**0.4**
#DK.MSWI-FA.2	4	20	80	28	0.4	4.9	99.6	4.9	7.8	1.0	3.9
	4	40	80	7	0.9	9.2	99.1	9.3	6.6	1.7	3.7
	4	40	80	14	1.0	8.1	99.0	**8.2**	8.4	1.0	**4.2**
#DK.MSWI-FA.3	4	20	80	28	0.6	2.7	99.4	**2.7**	3.9	0.3	**1.8**
#DK.MSWI-FA.4	4	20	80	28	0.5	3.1	99.5	**3.1**	3.4	0.5	**1.7**

2.4. Finland

The highest CO_2 content (11.6 wt%) of the ashes from Finland was found in the co-combustion ash (#FI.CC.APC.8), while other ashes showed a lower potential for CO_2 sequestration.

Table 4: TGA and calcimetric measurements for co-combustion ashes from Finland.

Sample ID	CO_2 conc. (%)	T (°C)	RH (%)	Time (days)	TGA (weight losses-%)		Calculations (TGA)		Calculations (calcimeter)		
					0-150 °C	550-950 °C	% dry matter	% CO_2/dry matter	% $CaCO_3$	% Dolomite	% CO_2
#FI.CC. APC.8	4	20	80	28	10.2	13.8	89.9	**15.4**	26.3	0.0	**11.6**
	4	40	80	7	9.1	13.8	90.9	15.2	24.1	1.4	11.3
	4	40	80	14	9.2	14.2	90.8	15.6	25.2	0.7	11.4
	20	40	80	3	7.7	13.2	92.3	14.3	25.7	0.7	11.6
#FI.CC. FA.8	4	20	80	28	0.7	5.2	99.3	**5.2**	9.2	1.6	**4.8**
	4	40	80	7	0.7	4.9	99.3	4.9	8.0	2.0	4.5
	4	40	80	14	0.8	4.7	99.2	4.7	8.2	1.9	4.5
#FI.CC. BA.8	4	20	80	28	0.0	0.5	100.0	**0.5**	0.8	0.0	**0.4**

2.5. The Netherlands

Biomass ashes from the Netherlands showed a high sequestration potential with high CO_2 content: 17.2 wt% for #NL.B.FA.1 and 17.3 wt% for #NL.WA.FA.4.

Table 5: TGA and calcimetric measurements for biomass ashes from the Netherlands.

Sample ID	CO_2 conc. (%)	T (°C)	RH (%)	Time (days)	TGA (weight losses-%)		Calculations (TGA)		Calculations (calcimeter)		
					0-150 °C	550-950 °C	% dry matter	% CO_2/dry matter	% $CaCO_3$	% Dolomite	% CO_2
#NL.B. FA.1	4	20	80	28	2.1	20.8	97.9	**21.2**	37.8	1.3	**17.2**
	4	40	80	7	1.9	19.6	98.1	20.0	36.2	1.3	16.5
	4	40	80	14	1.5	19.9	98.5	20.2	36.3	1.2	16.5
	20	40	80	3	1.9	19.6	98.2	19.9	36.5	1.1	16.5
#NL.B. FA.2	4	20	80	28	1.0	6.9	99.0	**7.0**	14.0	1.0	**6.7**
	4	40	80	7	0.5	5.9	99.5	5.9	11.7	1.2	5.7
	4	40	80	14	0.9	6.2	99.1	6.3	13.0	0.9	6.2
#NL.WA. FA.4	4	20	80	28.0	1.4	29.5	98.6	29.9	38.1	0.5	17.0
	4	40	80	7.0	1.1	27.2	98.9	27.5	37.8	1.2	17.2
	4	40	80	14.0	1.7	29.2	98.3	**29.7**	38.3	1.0	**17.3**

2.6. Belgium

Tables 6 and 7 show the TGA and calcimetric measurements for the ashes from Belgium: from the company Orbix and University of Ghent. The highest sequestration potential was determined for the MSWI ash (#BE.MSWI-FA.IV4).

Table 6: TGA and calcimetric measurements for the ashes received from Orbix.

Sample ID	CO_2 conc. (%)	T (°C)	RH (%)	Time (days)	TGA (weight losses-%)		Calculations (TGA)		Calculations (calcimeter)		
					0-150 °C	550-950 °C	% dry matter	% CO_2/dry matter	% $CaCO_3$	% Dolomite	% CO_2
#BE.ORB.1	4	40	80	7	1.3	9.0	98.7	9.1	17.7	8.3	11.8
	4	40	80	14	1.5	9.5	98.5	**9.7**	20.7	5.8	**11.9**
#BE.ORB.2	4	40	80	7	1.1	5.5	98.9	5.6	8.9	5.0	6.3
	4	40	80	14	1.2	6.1	98.8	**6.2**	10.2	3.6	**6.2**
#BE.ORB.3	4	40	80	7	1.2	6.3	98.8	6.4	17.1	1.3	8.1
	4	40	80	14	1.1	6.5	98.9	**6.5**	18.5	1.3	**8.7**

Table 7: TGA and calcimetric measurements for co-combustion and MSWI ashes from Belgium.

Sample ID	CO_2 conc. (%)	T (°C)	RH (%)	Time (days)	TGA (weight losses-%)		Calculations (TGA)		Calculations (calcimeter)		
					0-150 °C	550-950 °C	% dry matter	% CO_2/dry matter	% $CaCO_3$	% Dolomite	% CO_2
#BE.CC-FA.SL3	4	40	80	7	2.9	5.4	97.1	5.6	12.2	0.7	5.7
	4	40	80	14	3.0	5.2	97.0	**5.4**	12.6	1.0	**6.0**
#BE.CC.BA.SL-VAL3	4	40	80	7	1.1	5.9	98.9	6.0	15.8	0.5	7.1
	4	40	80	14	1.1	5.9	98.9	**5.9**	15.9	0.7	**7.3**
#BE.MSWI-BA.IV-VAL3	4	40	80	7	2.7	7.4	97.3	7.6	20.2	1.3	9.5
	4	40	80	14	2.4	7.2	97.6	7.4	20.6	1.1	9.6
#BE.MSWI-FA.IV4	4	40	80	7	2.2	16.2	97.8	16.6	34.0	3.9	16.8
	4	40	80	14	2.1	16.0	97.9	**16.3**	35.6	3.5	**17.3**

3. RESULTS FOR LCA

Table 8 contains data on the maximum CO_2 sequestration capacity for all ashes, expressed in g/kg, so that they can be used for further LCA calculations.

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Table 8: Maximum CO_2 sequestration capacity for each analyzed ash.

Country	Sample ID	Maximum CO_2 sequestration capacity (g/kg)
Slovenia	#SI.WA.FA.1	160
	#SI.WA.BA.1	302
	#SI.CC.FA.2	103
	#SI.CC.MA.2	276
	#SI.CC.FA.3	97
	#SI.CC.BA.3	159
	#SI.CC.FA.4	98
	#SI.CC.BA.4	202
Croatia	#HR.WA.1	221
	#HR.WA.2	272
	#HR.WA.3	198
	#HR.WA.4	271
	#HR.WA.6	172
	#HR.WA.7	84
	#HR.WA.8	246
	#HR.WA.9	245
	#HR.WA.11	304
	#HR.WA.15	251
	#HR.WA.16	182
	#HR.SSA.1	49
	#HR.SSA.2	34
	#HR.SSA.3	9
	#HR.SSA.4	9
Denmark	#DK.MSWI-FA.1	59
	#DK.SSA-FA.1	2
	#DK.SSA-FA.2	4
	#DK.MSWI-FA.2	42
	#DK.MSWI-FA.3	18
	#DK.MSWI-FA.4	17
Finland	#FI.CC.APC.8	114
	#FI.CC.FA.8	45
	#FI.CC.BA.8	4
The Netherlands	#NL.B.FA.1	172
	#NL.B.FA.2	67
	#NL.WA.FA.4	173
Belgium	#BE.ORB.1	119
	#BE.ORB.2	62
	#BE.ORB.3	87
	#BE.CC-FA.SL3	60
	#BE.CC.BA.SL-VAL3	73
	#BE.MSWI-BA.IV-VAL3	96
	#BE.MSWI-FA.IV4	173

4. SUMMARY AND CONCLUSIONS

The ashes were selected from various partners from different countries. In total, the selection included 16 wood ashes, 11 co-combustion ashes, 6 sewage sludge ashes, 6 MSWI ashes and 3 others. During this task we were able to establish the methodology to assess the sequestration potential of waste ashes. This methodology includes:

- Sampling (homogenized by quartening)
- Grinding and sieving (below 125 µm)
- Exposure of the ash to accelerated carbonation
- Analysis with a pressure calcimeter and thermogravimetric analysis.

Based on calculations, the amount of CO_2 was quantified using the calcimetric method, since other phases can also be detected with TGA in the temperature range from 550 to 950 °C. We identified the ashes with the highest sequestration potential of about 300 g/kg (#SI.WA.BA.1 and #HR.WA.11) and prepared the data for all ashes for further LCA calculations.

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Appendix

The calibration curve for the calcimetric measurements is shown in Figure A1. Based on the chemical and mineralogical composition of the ash it is possible to predict which ash has a sequestration potential. The mean values of the primary oxides measured by XRF and the loss on ignition (LOI) at 950 °C are given in the Tables A1-A7. The XRD analysis enables to monitor the carbonation process and the relative content of e.g. calcite/lime as shown in Figure A2.

Figure A1: Calibration curve for calcimetric measurement

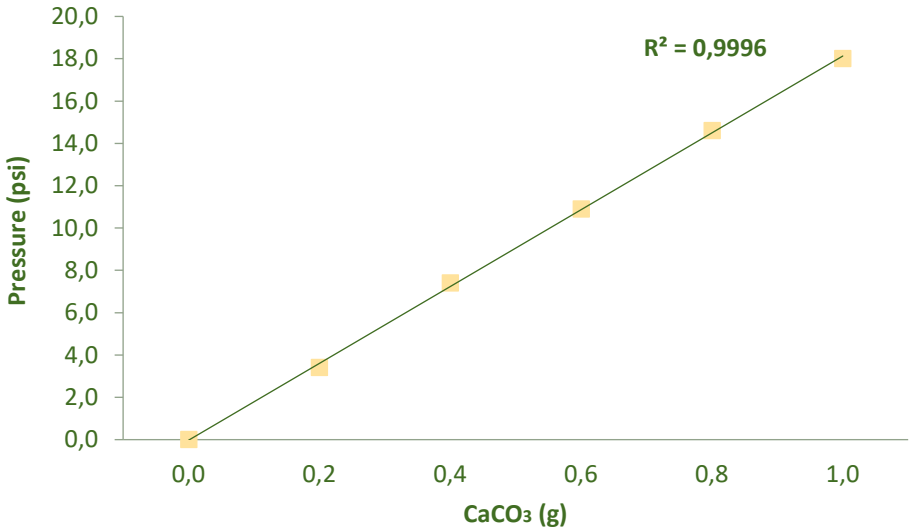

Table A1: XRF results for ashes from Slovenia.

	#SI.WA. FA.1	#SI.WA. BA.1	#SI.CC. FA.2	#SI.CC. MA.2	#SI.CC. FA.3	#SI.CC. BA.3	#SI.CC. FA.4	#SI.CC. BA.4
LOI 950°C	29.91	26.09	11.71	14.55	16.98	12.79	22.31	7.21
Na_2O	0.56	0.48	0.74	0.41	0.28	0.57	21.92	2.28
MgO	3.78	5.88	5.99	2.12	8.21	10.25	1.60	3.46
Al_2O_3	5.62	3.35	11.27	11.08	10.87	7.43	6.85	20.05
SiO_2	19.04	5.64	22.90	14.45	27.78	31.17	8.19	25.91
P2O5	2.24	2.82	0.28	0.26	0.56	1.00	0.89	2.45
SO_3	1.00	0.32	1.70	0.20	1.69	0.07	3.85	0.86
K_2O	5.65	8.14	0.99	0.25	2.16	3.21	2.48	0.55
CaO	29.27	44.95	34.01	55.40	19.77	28.98	13.50	28.62
TiO_2	0.37	0.07	0.73	0.22	0.52	0.32	0.97	1.85
V_2O_5	0.01	0.01	0.02	/	0.02	0.02	0.01	0.03
Cr_2O_3	0.01	0.01	0.02	/	0.01	0.02	0.04	0.11
MnO	0.43	0.93	0.15	0.03	0.27	0.31	0.06	0.21
Fe_2O_3	1.70	0.68	8.53	0.56	10.44	3.56	1.21	3.37
Co_3O_4	/	/	0.01	/	0.02	/	/	/
NiO	0.00	0.01	0.01	/	0.01	0.01	/	0.02
CuO	0.01	0.02	0.04	0.06	0.01	0.01	0.10	0.71
ZnO	0.05	0.09	0.27	0.03	0.04	0.02	1.04	1.02
As_2O_3	0.07	0.08	0.08	0.07	0.08	0.10	0.16	0.17

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Rb_2O	0.02	0.02	0.01	0.01	0.01	0.01	0.01	0.01
SrO	0.06	0.09	0.08	0.09	0.07	0.05	0.02	0.05
ZrO_2	0.02	0.00	0.02	0.02	0.02	0.02	0.01	0.05
BaO	0.12	0.29	0.19	0.03	0.17	0.07	0.07	0.23
PbO	0.02	/	0.03	0.02	0.01	0.02	0.09	0.06
Cl	/	/	/	/	/	/	14.45	0.56
SUM	99.96	99.95	99.76	99.85	99.97	99.97	99.85	99.83

Table A2: XRF results for WBA from Croatia.

	#HR. WA.1	#HR. WA.2	#HR. WA.3	#HR. WA.4	#HR. WA.6	#HR. WA.7	#HR. WA.8	#HR. WA.9	#HR. WA.11	#HR. WA.15	#HR. WA.16
LOI 950°C	20.81	20.49	12.54	13.49	7.53	5.17	15.56	11.89	20.99	14.51	8.25
Na_2O	0.70	1.99	0.38	0.68	0.64	0.48	0.51	/	0.27	0.68	0.88
MgO	3.05	2.84	3.56	3.12	3.05	4.43	4.72	2.43	5.48	2.69	4.53
Al_2O_3	3.83	0.31	2.75	3.38	6.06	9.14	1.86	1.51	0.96	2.50	3.64
SiO_2	15.42	1.03	9.22	14.66	20.71	36.61	5.94	6.86	2.67	9.72	12.14
P_2O_5	2.17	2.01	3.68	2.60	3.38	2.30	3.16	1.40	1.99	2.06	4.03
SO_3	3.44	10.00	0.97	0.50	2.61	0.57	6.66	1.38	5.90	1.24	12.20
K_2O	9.82	19.06	4.34	8.13	8.57	8.74	14.15	3.08	18.98	5.54	8.93
CaO	37.51	40.97	59.86	50.71	40.63	25.30	44.79	69.89	41.12	58.18	41.40
TiO_2	0.33	0.04	0.24	0.22	0.46	0.58	0.16	0.12	0.08	0.18	0.28
Cr_2O_3	0.01	0.02	0.01	0.03	0.04	0.02	0.01	0.01	0.03	0.01	0.02
MnO	0.52	0.23	0.40	0.38	3.20	0.50	0.69	0.28	0.40	0.87	0.67
Fe_2O_3	2.01	0.36	1.66	1.84	2.68	5.95	1.18	0.86	0.64	1.38	1.94
CuO	0.02	0.03	0.03	0.02	0.05	0.01	0.03	0.03	0.04	0.02	0.02
ZnO	0.05	0.04	0.01	/	0.06	0.03	0.10	0.01	0.10	0.03	0.17
SrO	0.09	0.10	0.16	0.13	0.09	0.05	0.11	0.15	0.06	0.12	0.10
BaO	0.13	0.13	0.21	0.13	0.14	0.14	0.22	0.09	0.11	0.30	0.11
Cl	0.12	0.41	0.03	/	0.14	0.01	0.22	0.03	0.24	/	0.73
SUM	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00

Table A3: XRF results for SSA ashes from Croatia.

	#HR.SSA.1	#HR.SSA.2	#HR.SSA.3	#HR.SSA.4
LOI 950°C	19.44	31.84	13.21	13.81
Na_2O	0.64	0.80	0.66	1.25
MgO	1.72	2.45	3.05	2.38
Al_2O_3	10.28	6.81	13.60	13.52
SiO_2	26.90	21.27	38.36	41.45
P_2O_5	10.98	7.12	8.54	6.79
SO_3	3.71	4.65	2.26	1.63
K_2O	1.14	1.34	2.28	1.94
CaO	18.46	17.48	9.65	7.38
TiO_2	0.55	0.64	0.75	0.72
Cr_2O_3	0.06	0.06	0.05	0.08
MnO	0.09	0.08	0.16	0.24
Fe_2O_3	5.73	4.84	6.89	8.54
CuO	0.03	0.07	0.31	0.03

ZnO	0.13	0.15	0.11	0.03
SrO	0.03	0.02	0.03	0.11
BaO	0.07	0.09	0.09	0.12
Cl	0.07	0.33	0.03	0.03
SUM	100.00	100.00	100.00	100.00

Table A4: XRF results for ashes from Denmark.

	#DK.MSWI-FA.1	#DK.SSA-FA.1	#DK.SSA-FA.2	#DK.MSWI-FA.2	#DK.MSWI-FA.3	#DK.MSWI-FA.4
LOI 950ºC	9.55	1.17	1.35	3.58	4.62	7.99
Na_2O	3.98	0.96	1.22	5.38	10.14	13.42
MgO	2.76	3.48	4.55	3.36	3.04	2.58
Al_2O_3	10.83	10.49	6.84	9.07	7.28	5.59
SiO_2	21.07	30.67	26.52	24.31	17.86	14.84
P_2O_5	1.36	22.49	25.98	1.86	1.57	1.38
SO_3	7.29	0.24	0.12	10.81	10.73	11.23
K_2O	3.49	2.01	1.88	2.51	5.45	6.58
CaO	24.87	14.92	14.86	26.14	21.17	17.60
TiO_2	1.82	0.74	0.80	2.38	1.78	1.48
V_2O_5	0.02	0.02	0.02	0.02	0.02	0.01
Cr_2O_3	0.11	0.01	0.02	0.05	0.03	0.03
MnO	0.11	0.10	0.09	0.19	0.19	0.17
Fe_2O_3	6.24	11.76	14.70	2.09	1.63	1.43
Co_3O_4	0.01	0.01	0.01	/	/	/
NiO	0.02	0.01	0.01	0.01	0.01	0.01
CuO	0.08	0.08	0.10	0.04	0.08	0.11
ZnO	1.92	0.31	0.40	1.31	1.94	2.25
As_2O_3	0.09	0.07	0.09	0.14	0.15	0.10
Rb_2O	0.01	0.01	0.01	0.01	0.02	0.02
SrO	0.07	0.26	0.23	0.06	0.05	0.04
ZrO_2	0.03	0.03	0.02	0.04	0.03	0.02
SnO_2	0.11	/	/	0.03	0.05	0.07
Sb_2O_3	0.08	/	/	0.05	0.06	0.06
Cs_2O	0.01	/	0.01	/	0.02	0.02
BaO	0.15	0.12	0.13	0.36	0.28	0.26
La_2O_3	/	/	0.01	/	/	/
PbO	0.01	0.01	0.01	0.01	/	/
Cl	3.93	/	/	4.16	10.30	12.72
SUM	99.98	99.96	99.96	99.96	99.96	99.99

Table A5: XRF results for ashes from Finland.

	#FI.CC.APC.8	#FI.CC.FA.8	#FI.CC.BA.8
LOI 950ºC	16.30	2.79	0.06
Na_2O	1.53	3.74	3.65
MgO	2.49	3.85	2.32
Al_2O_3	8.51	14.97	11.57
SiO_2	15.57	31.11	62.62
P_2O_5	1.38	2.50	0.80

SO_3	9.11	6.10	0.17
K_2O	0.94	2.70	2.79
CaO	32.13	19.40	10.32
TiO_2	0.52	1.37	0.69
V_2O_5	0.02	0.03	0.02
Cr_2O_3	0.03	0.08	0.11
MnO	0.30	0.52	0.29
Fe_2O_3	6.45	9.65	3.63
Co_3O_4	0.01	0.01	/
NiO	0.01	0.02	0.01
CuO	0.01	0.16	0.19
ZnO	0.09	0.25	0.31
As_2O_3	0.08	0.12	0.10
Rb_2O	0.01	0.02	0.01
SrO	0.04	0.08	0.06
ZrO_2	0.01	0.03	0.06
BaO	0.08	0.19	0.21
La_2O_3	/	0.02	0.01
Cl	4.38	0.31	/
PbO	/	/	0.01
SUM	99.96	99.96	99.99

Table A6: XRF results for ashes from The Netherlands.

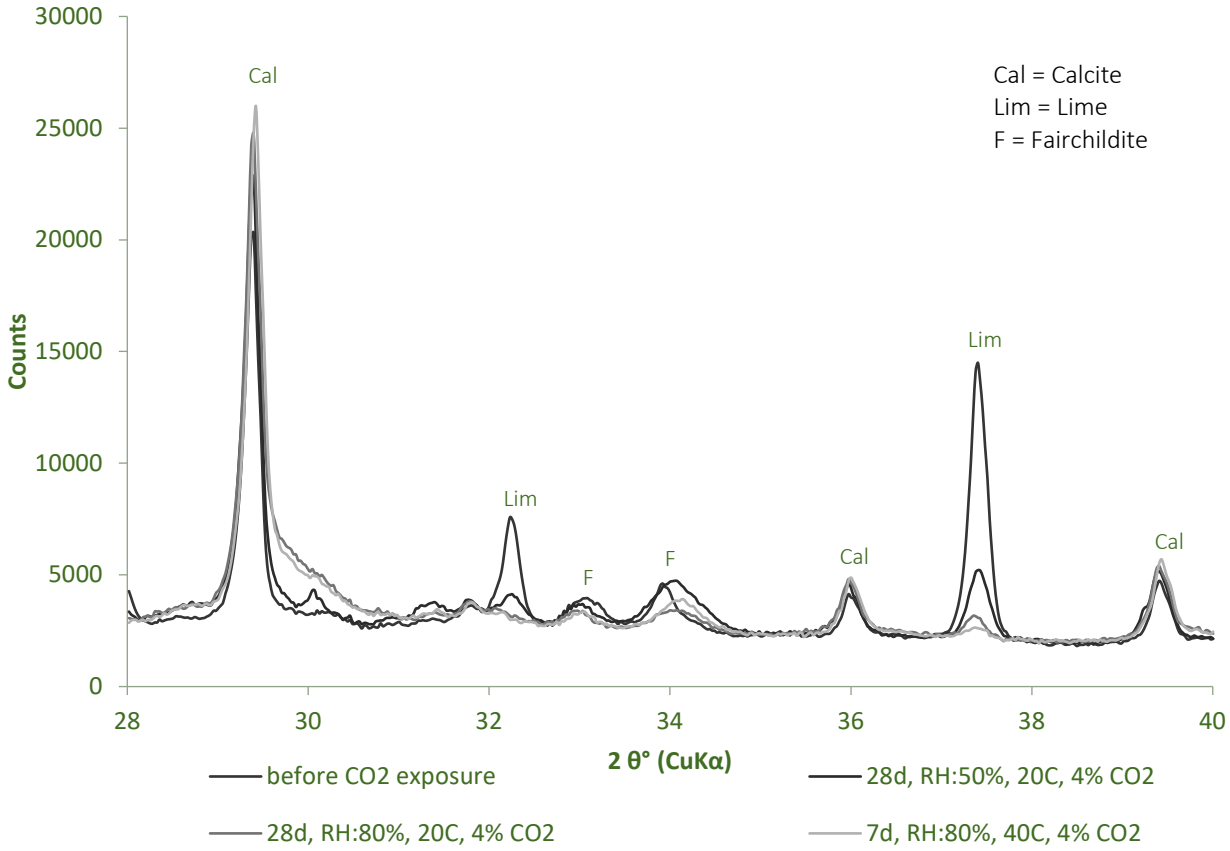
	#NL.B.FA.1	#NL.B.FA.2	#NL.WA.FA.4
LOI 950°C	13.85	3.94	19.90
Na_2O	1.48	1.90	2.95
MgO	3.17	3.67	2.42
Al_2O_3	5.63	7.54	3.69
SiO_2	25.51	45.20	14.80
P_2O_5	2.58	2.08	0.95
SO_3	3.88	1.62	5.86
K_2O	4.68	1.75	1.98
CaO	33.38	22.25	36.63
TiO_2	1.48	4.10	1.43
V_2O_5	0.01	0.03	0.01
Cr_2O_3	0.04	0.11	0.07
MnO	0.27	0.45	0.28
Fe_2O_3	1.86	3.45	1.59
Co_3O_4	/	0.01	/
NiO	0.01	0.02	/
CuO	0.03	0.08	0.04
ZnO	0.49	0.62	3.25
As_2O_3	0.08	0.10	0.09
Rb_2O	0.01	0.01	0.00
SrO	0.09	0.09	0.07
ZrO_2	0.03	0.08	0.02
Cs_2O	/	/	0.03

BaO	0.19	0.72	0.22
SnO_2	/	/	/
Ta_2O_5	0.01	/	/
PbO	0.12	0.10	0.07
Cl	1.10	0.07	3.63
SUM	99.97	99.96	99.96

Table A7: XRF results for ashes from Belgium.

	#BE.ORB.1	#BE.ORB.2	#BE.ORB.3	#BE.CC-FA.SL3	#BE.CC. BA.SL-VAL3	#BE.MSWI-BA. IV-VAL3	#BE.MSWI-FA. IV4
LOI 950ºC	4.75	3.78	3.30	8.65	11.93	21.73	9.61
Na_2O	3.15	3.78	0.24	1.71	2.27	2.57	2.06
MgO	3.29	3.95	12.92	3.17	1.87	2.70	4.11
Al_2O_3	10.97	3.38	8.66	10.43	10.19	8.80	10.98
SiO_2	39.02	9.69	26.93	31.09	52.73	32.12	16.12
P_2O_5	2.78	32.10	/	2.79	0.24	1.55	3.58
SO_3	3.33	2.35	0.24	8.50	1.20	1.43	6.44
K_2O	1.46	9.46	/	0.99	1.39	0.72	0.74
CaO	24.79	2.39	42.10	24.89	13.10	20.29	38.10
TiO_2	2.23	25.71	1.03	1.40	0.49	0.86	2.87
V_2O_5	0.02	2.35	0.07	0.03	0.01	0.01	0.02
Cr_2O_3	0.10	0.02	2.33	0.04	0.04	0.06	0.12
MnO	0.10	0.12	1.07	0.10	0.12	0.23	0.12
Fe_2O_3	2.54	0.11	0.76	4.91	3.84	5.59	1.88
Co_3O_4	0.00	2.49	/	0.00	/	0.01	0.01
NiO	0.09	/	0.08	0.01	0.01	0.02	0.01
CuO	0.04	0.08	/	0.08	0.08	0.19	0.01
ZnO	0.43	0.05	0.00	0.50	0.11	0.69	0.53
As_2O_3	0.06	1.10	0.06	0.12	0.12	0.10	0.11
Rb_2O	0.00	0.08	0.00	0.01	0.01	0.01	0.01
SrO	0.07	0.01	0.03	0.12	0.03	0.07	0.08
ZrO_2	0.04	0.08	0.03	0.05	0.09	0.03	0.04
Cs_2O	/	0.08	/	/	/	0.01	/
BaO	0.18	/	/	0.27	0.09	0.14	0.17
SnO_2	/	0.18	/	0.01	/	0.01	0.01
Nb_2O_5	0.00	0.02	0.14	/	0.00	0.01	/
PbO	0.00	0.00	/	/	0.02	0.03	/
Cl	0.56	/	/	0.07	/	/	2.23
PtO_2	/	/	/	0.02	0.01	0.01	/
Au	/	/	/	0.01	0.01	0.01	0.00
SUM	99.99	99.99	99.99	99.98	99.99	99.97	99.94

Figure A2: X-ray diffraction patterns of selected sample #SI.WA.BA.1.



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