Environmental potentials of asphalt mixtures fabricated with red mud and fly ash

Mayara S. Siverio Lima a, M. Hajibabaei a, L. P. Thives b, V. Haritonovs c, A. Buttgereit d, C. Queiroz e and F. Gschösser a

aFaculty of Engineering Sciences, University of Innsbruck, Innsbruck, Austria; bDepartment of civil engineering, University of Santa Catarina, Florianopolis, Brazil; cDepartment of Roads and Bridges, Riga Technical University, Riga, Latvia; dDepartment of Mobility and Civil Engineering, Münster, Germany; eWorld Bank, Washington, DC, USA

ABSTRACT
Several studies evaluated the feasibility of using residues to compose asphalt mixtures. However, the demand for treatments are often neglected in determining the environmental impacts. This study aims to elucidate the decision-making process over the application of residues (e.g., red mud and fly ash) to produce asphalt mixtures. For comparison purposes, limestone and dolomite are used as reference fillers. The cradle-to-gate approach is applied within three scenarios. In the first scenario, the treatment of the residues is included in the modelling, the second excludes treatment, and the third scenario evaluates the environmental impacts of the residues deposited in landfills. To perform the analysis, indicators such as Global Warming Potential, Acidification, and Cumulative Energy Demand are applied. The results show that the treatment provided to the residues strongly influences the environmental impacts of the production of asphalt mixtures and may be crucial to define the feasibility of the residues application.

1. Introduction
The red mud is an alkaline residue generated during the production of aluminum (Tan & Khoo, 2005). Annually, 125 million tons of red mud are produced (2013) and it is estimated that 30 billion metric tons are already accumulated worldwide (Mishra & Gostu, 2017).

The alkaline characteristics of the red mud limits large scale applications and, although several studies investigated the use of red mud in various applications, such as construction materials (Hind et al., 1999), ceramic (Kavas, 2006; Sglavo et al., 2000; Yalçin & Sevinç, 2000; Yang & Xiao, 2008), bricks (Dodoo-Arhin et al., 2013; Feng et al., 2015), tiles (Yang et al., 2009), cement (Liu & Poon, 2016; Manhin et al., 2016) and road pavement layers (Jitsangiam & Nikraz, 2013; Siverio Lima, 2015), the final destination are still industrial landfills (Collazo, Fernández, Isquierdo, et al., 2005; Hildebrando et al., 2013; Kavas, 2006; Yang & Xiao, 2008).

The same occurs with the fly ash, residue generated during the combustion of coal in electrical power plants (Ahmaruzzaman, 2010; Chen et al., 2010). The current worldwide production of fly ash is estimated to be around 400–500 million tons per year and this huge amount of fly ash released
by thermal power plants has become a serious environmental problem since it is considered a highly contaminating material (Ahmaruzzaman, 2010; Nadeem Akhtar & Tarannum, 2019).

Landfills are the main destination for both residues, promoting serious environmental damages due to leaching and gas emissions. Hazardous compounds are often released affecting surrounding areas. Vegetation damage, groundwater pollution, air pollution and greenhouse emissions are some of the consequences of landfills (El-Fadel et al., 1997).

During the last decades, alternative applications for both residues (i.e. red mud and fly ash) have been studied in order to reduce the environmental impacts caused by their storage. Researches from China (Wang et al., 2019; Yao et al., 2020; Zhang et al., 2018), India (Choudhary et al., 2020; Rai et al., 2020), Greece (Kehagia, 2014), Australia (Jitsangiam & Nikraz, 2013) and Brazil (Bezerra et al., 2010; Lima et al., 2017) evaluate the possibilities of using the residues within pavement constructions. In general, the authors highlight that any particular application of residues should be competitive in terms of quality, performance, costs and risks.

Kehagia (2014) evaluated the performance of an unpaved road built with 97% red mud and 3% fly ash, reinforced with bauxite aggregates. According to the author, the pilot road project showed a good performance, and after three years, the pavement had no rutting, surface deformation, nor erosion.

Jitsangian and Nikraz (Jitsangiam & Nikraz, 2013) investigate the use of red mud as a base material for roads using a stabilised mixture of 70% red mud, 25% fly ash, and 5% lime. The results showed a superior performance of the stabilised residues in comparison with conventional materials.

Lima et al. (2016) and Bezerra et al. (2010) evaluated the performance of bituminous mixtures composed with red mud and compared the results with mixtures made with conventional materials. Both studies showed that red mud can improve the resistance of bituminous layers by improving the connection aggregate-binder.

Kehagia (2014) highlighted that potential treatment demands may restrict the suitability of using residues to compose pavements. There are several methods to neutralise residues, for the red mud are mainly used: acids (Rai et al., 2012), seawater (Rai et al., 2012), carbon dioxide (Han et al.; Johnston et al., 2010) and high temperatures (sintering) (Rai et al., 2013). However, none of them is able to inert radioactive and toxic elements, being only for alkalinity neutralisation.

Taking into account that utilising residues in road constructions minimises costs, reduces the area demanded for residues disposal and replaces scarce or expensive natural resources (Ahmaruzzaman, 2010), this study evaluates the environmental potentials of the production of asphalt mixtures fabricated with fly ash and red mud to be applied on road pavements. The reference asphalt materials are produced with limestone and dolomite (EN 12697-35, 2016).

To assess the environmental potentials, the ‘cradle-to gate’ method is used and the residues are modelled in three different scenarios: including treatment before the asphalt production, excluding treatment, and depositing the residues in landfills.

2. Framework of the study

This study evaluates the environmental potentials of the asphalt production of four asphalt mixtures composed of different types of fillers. The reference asphalt mixtures are composed of limestone (LS) and dolomite (DL), while the other two are composed of residues, such as red mud (RM) and fly ash (FA) (EN 12697-30, 2004).

The main goal is to evaluate the impacts of using residues with and without prior treatment. Since landfilling is a common destination for residues, this scenario is included in the analysis.

The input data used in this paper are taken from different sources, and further explained in the following topics:

- The amount of raw materials used to produce the asphalt mixtures was designed in the laboratory to obtain the optimum content of bitumen for each asphalt mixture.
- The heat, energy consumption and some additional data were taken from literature.
As both residues are widely available in Brazil, the distances used in the modelling consider the material suppliers' locations and the asphalt plant within the country.

The dataset used to model the residues (treated, non-treated and in landfills) were taken from the Ecoinvent database.

2.1. Laboratory tests

The asphalt mixtures are composed of 50/70 bitumen, granite aggregates, and fillers such as red mud, fly ash, dolomite and limestone. The mixtures were designed in laboratory using Marshall method, in accordance with the EN 12697-30 and EN 12687-35 standards.

The asphalt mixtures are categorised as Asphalt Concrete (AC) with the largest grain size value 16 mm (AC 16) and meet the requirements to be applied within the surface layer of the road pavements (EN 12697-30, 2004; EN 13108-1, 2016). The amount of filler (7%) to be used within all mixtures was predefined to avoid differences in the analysis. The optimum asphalt content has been defined in order to reach 4% of air voids and varied according to the porosity of the filler used. Table 1 shows the asphalt mixtures analysed in this study.

2.2. Asphalt production

The asphalt plant is characterised as a batch mixing plant and is located in the south of Brazil, produces around 310.000 tons of asphalt mixtures per year, and uses heavy fuel oil for heating. The mineral aggregates and fillers are pre-heated at 110 °C and mixed with the bitumen at an average temperature of 160°C.

As the asphalt producer from Brazil was not able to provide an overall annual amount of heating and energy consumed to produce the asphalt mixtures, these data were taken from the literature (Lima, Hajibabaei, et al., 2020) and are shown in Table 2.

2.3. Geographic boundaries

Brazil is one of the largest contributors to the generation of red mud in the world (Lima & Thives, 2020; Lima, Thives, et al., 2020), and reserves vast areas for the storage of fly ash.

In that sense, this study aims to simulate scenarios considering the geographic boundaries of Brazil. The asphalt plant modelled is located in Biguaçu, state of Santa Catarina (South of Brazil) at the same place as the extraction of aggregates (granite).

Table 1. Asphalt mixtures recipes.

| Mixtures | Filler (kg/kg mixture) | Sand (kg/kg mixture) | Gravel Crushed (kg/kg mixture) | Bitumen (kg/kg mixture) | Density (g/cm³) |
|----------|------------------------|----------------------|-------------------------------|------------------------|-----------------|
| AC 16 (DL) | 0.07 | 0.27 | 0.61 | 0.047 | 2.479 |
| AC 16 (LS) | 0.07 | 0.27 | 0.61 | 0.045 | 2.474 |
| AC 16 (FA) | 0.07 | 0.27 | 0.61 | 0.050 | 2.473 |
| AC 16 (RM) | 0.07 | 0.27 | 0.61 | 0.047 | 2.448 |

Table 2. Energy and operating materials of the asphalt production.

| Energy and operating material | Energy and operating material |
|------------------------------|------------------------------|
| Electricity                  | 16.02 MJ/t                   |
| Heat (heavy fuel oil)        | 280.08 MJ/t                  |
| Diesel (internal transports) | 8.53 MJ/t                    |
The bitumen, the fly ash, the limestone and the dolomite are taken from a company located in Araucaria, State of Paraná (289 km from the asphalt plant) – near the capital Curitiba. On the other hand, the aluminium plant responsible for generating the red mud is located in Poços de Caldas, State of Minas Gerais (923 km to the asphalt plant), as shown in Figure 1.

2.4. Treatment of residues

The use of red mud to compose asphalt mixtures has been studied only in laboratory and, has never been applied on industrial scale (Collazo, Fernández, Izquierdo, et al., 2005; Lima et al., 2016, 2017). This occurs because the red mud is still considered hazardous material and further environmental analysis, such as leaching from the asphalt mixtures, are currently being developed. In addition, none of the treatments tested so far are considered truly effective or require higher costs, due to the complexity of the procedure (D. Cooling, 2007; Lima et al., 2016; Palmer et al., 2010).

To reduce the alkalinity of the red mud and allow the residue to compose asphalt mixtures without causing any future harm, this study simulates a pre-treatment scenario that includes washing the red mud with water in order to remove as much caustic soda (NaO₂) as possible, drying and, crushing the residue. Since the particles of red mud easily agglomerate, the crushing process should facilitate its use as filler.
On the other hand, the most common ways of handling fly ash are landfilling or using it as a secondary material after inertisation. Among many inertisation procedures, the fly ash is often submitted to a thermal treatment (i.e. sintering) to entrap heavy metals and abate organic pollutants (Zacco et al., 2014).

In most cases, the sintering process involves temperatures that reach 900–1,000°C, and creates a more homogeneous, denser product with improved leaching properties (Zacco et al., 2014). Therefore, one of the scenarios analysed within this paper considers that the fly ash is thermally treated (sintering) before the production of asphalt mixtures.

3. Life cycle assessment

3.1. Goal and scope

This study assesses the environmental potential within the production of asphalt mixtures composed with different fillers, such as red mud, fly ash, limestone and dolomite by using the cradle-to-gate approach (ISO 14040, 2019; ISO 14044, 2019). Therefore, all life cycle phases from raw material acquisition to the finished product at the asphalt plant are analysed (i.e. A1–A3) (EN 15643-5, 2016; EN 15804, 2018; ISO 14040, 2019). As long-term service life is out of boundaries, the pavement performance is not taken into account.

As fly ash and red mud are secondary materials generated in another primary process, all environmental burdens prior to their generation are attributed to the primary system, and therefore, not included in this analysis.

Three different scenarios were modelled in order to simulate different realities and compare the environmental potentials of using residues (treated or not) within the production of asphalt mixtures.

- In the first scenario, the residues receive treatment before the production of the asphalt mixtures. The treatment applied to the red mud is washing, drying and crushing while the fly ash receives a thermal treatment (sintering). Both treatments occur at the same site where the residue is generated.
- The second scenario neglects the necessity of treatment, as both residues are considered inert and fully covered by asphalt during the asphalt production. In this scenario, the residues are transported directly from the original site of generation to the asphalt plant.
- The third scenario simulates the storage of the residues in landfills, located near the original site of generation.

The functional unit used in this study is 1 kg of asphalt mixture produced at the asphalt plant. The functional unit applied within the third scenario is 1 kg of residue (red mud or fly ash) deposited in the landfill. The system boundaries and phases considered in the analysis are shown in Figure 2.

The nomenclature used to describe the life cycle stages in Figure 2 is defined by the EN 15804:2018 standard, as presented below: (EN 15804, 2018)

- A1 – raw material supply, including the processing of secondary material input
- A2 – transport of raw material and secondary material to the manufacturer
- A3 – manufacture of the construction products, and all upstream processes from cradle to gate
- C3 – waste processing operations for reuse, recovery, or recycling
- C4 – final disposal of end-of-life construction product

3.2. Life cycle inventory (LCI)

The primary data and some other relevant inputs and outputs connected with the asphalt production, such as energy and heating consumption were obtained from the literature (Gschösser, 2011; Gschösser et al., 2012; Lima, Hajibabaei, et al., 2020).
The asphalt mixtures were designed in the laboratory and the recipes were obtained through tests conducted in accordance with the EN 12697-30 and EN 12687-35 standards.

The transportation distances consider the location of the specific suppliers to the asphalt plant in Brazil. The aggregates extraction site is located near the asphalt plant. The bitumen, fly ash, limestone and dolomite are 289 km distant from the asphalt plant. The distance from the red mud is 923 km (EN 12697-35, 2016).

All materials are transported by a four-axle lorry. The vehicle weighs approximately 14 tons and has a maximum load capacity of 26 tons. The fuel used is diesel and the consumption is about 0.18 l/km when empty and 0.32 l/km when fully loaded (EN 12697-30, 2004; Lima, Hajibabaei, et al., 2020).

All inputs and outputs were modelled using SimaPro 9.0 software (PRé Consultants, 2016) and the Ecoinvent 3.5 database (Ecoinvent Center, 2010). Although the asphalt mixtures were designed with a 50/70 bitumen, SimaPro provides only one average dataset for bitumen, which was used to model all asphalt mixture LCIs. The mineral aggregates were modelled with the dataset for crushed gravel. The reference fillers (i.e. limestone, dolomite) used within the asphalt mixtures were milled limestone and dolomite, for which the corresponding Ecoinvent dataset was applied. The red mud was modelled with the dataset for bauxite digestion residue, and the treated red mud was modelled combining datasets of the untreated red mud and datasets for washing, drying, and crushing, taken from the different operations applied within the limestone and available in the database. The fly ash was modelled using the dataset entitled ‘fly ash and scrubber sludge’ and both treated and untreated options are available in the Ecoinvent, as well as the datasets for the landfilling of the red mud and fly ash.

The LCIs of all analysed asphalt mixtures are listed in Table S1.

### 3.3. Life cycle impact assessment

To assess the environmental impacts of the asphalt production, it is used the Global Warming Potential – GWP (kg CO₂ equivalent) and Acidification (kg SO₂ equivalent) indicators from the Environmental Product Declarations (EPD 2018) (EN 15804, 2018) method and the Non-renewable Cumulative Energy Demand (nr-CED) based on the Cumulative Energy Demand method V1.11 published by Ecoinvent version 2.0 (Ecoinvent Center, 2010), and presented in MJ equivalent.
4. Results

Table 3 shows the environmental impacts (GWP, Acidification, and nr-CED) associated with the production of the asphalt mixtures analysed.

Table 3 shows that the environmental impacts generated by the production of asphalt mixtures composed of limestone and dolomite are nearly equivalent to the impacts of the asphalt mixtures fabricated with red mud (treated and untreated) and the fly ash (untreated) within all indicators analysed. The treatment added to the red mud (washing, drying, and crushing) does not substantially increase the impacts within the production of asphalt mixtures composed with red mud (untreated).

The greatest environmental impact generated in terms of nr-CED, Acidification, and GWP is given by the production of asphalt materials composed of treated fly ash. Therefore, the small amount of filler (7%) chosen to compose the asphalt mixtures does not substantially influence the environmental impacts within the production process, except when a highly impactful procedure is included, such as the thermal treatment given to fly ash.

The landfill of both residues revealed the lowest environmental potentials amongst all the scenarios evaluated. In other words, the storage of residues produces lower environmental impacts than using the residues as filler in the production of asphalt mixtures. This study, however, only provides a cradle-to-gate analysis, and further investigations including all life cycle phases of the pavement should be performed. Also, it is considered the geographical boundaries of Brazil, which increases the distances between suppliers and the asphalt plant and, therefore, the impacts of using the residues within the asphalt production. Thus, the results of the environmental impacts may change for different scenarios.

In Figure 3, the environmental impacts in terms of nr-CED are divided into different categories, considering the impacts of raw materials (i.e. aggregates, bitumen, filler) and processes (i.e. heating, transportation) caused by the mixtures during the asphalt production. Figures 4 and 5 show the same categories considering the acidification and GWP indicators. In Figures 3–5 the results are shown taking the AC FA – T values as basis since is the highest impact obtained.

Figure 3 shows that all asphalt mixtures present nearly 70% of the impact given by the AC 16 FA (T) in terms of CED, nearly 40% in terms of acidification (Figure 4) and 30% in terms of GWP (Figure 5).

The greater values of CED obtained by the AC 16 FA (T) mixture are due to the high consumption of non-renewable energy given by the bitumen and treatment of the fly ash. The use of fossil fuels to produce energy generate higher amounts of carbon dioxide (CO2) as a by-product, which is absorbed by the seawater and soils, lowering the pH and increasing the acidification values.

The bitumen is the main responsible for the environmental burdens associated with the production of asphalt mixtures in terms of nr-CED due to the energy and resources required for its extraction and manufacturing processes. On the other hand, the fly ash has around 30% of the environmental burdens in terms of nr-CED associated with the production of the asphalt mixture AC 16 FA (T) due to the thermal treatment given to the residue.

The thermal treatment given to the fly ash contributes within 60–80% to the environmental impacts regarding the acidification and GWP indicators. The incineration process produces a variety of pollutants and emissions to the atmosphere, dangerous for human health, which expressively increases the

|          | CED – Non renewable (MJ eq/kg) | Acidification (kg SO2 eq/kg) | Global warming (kg CO2 eq/kg) |
|----------|--------------------------------|------------------------------|-------------------------------|
| AC 16 DL | 3.43E+00                       | 4.88E-04                     | 6.65E-02                      |
| AC 16 LS | 3.39E+00                       | 4.72E-04                     | 6.35E-02                      |
| AC 16 FA (T) | 5.08E+00                     | 1.32E-03                     | 2.60E-01                      |
| AC 16 FA (NT) | 3.40E+00                    | 4.70E-04                     | 6.36E-02                      |
| FA (Landfill) | 2.96E-01                     | 6.63E-05                     | 8.19E-03                      |
| AC 16 RM (T) | 3.41E+00                     | 5.05E-04                     | 6.96E-02                      |
| AC 16 RM (NT) | 3.40E+00                    | 5.02E-04                     | 6.90E-02                      |
| RM (Landfill) | 2.95E+00                     | 6.68E-05                     | 8.15E-03                      |
Figure 3. Nr-CED results (%).

Figure 4. Acidification results (%).

Figure 5. GWP results (%).
share of the impact associated with the production of the asphalt mixtures containing treated fly ash, even though the asphalt mixture contains only 7% of the residue.

The modelled treatment for the red mud (i.e. washing, drying, and crushing) does not contribute significantly to the impacts caused by the production of asphalt mixtures composed of treated red mud. The use of red mud, however, increases the impacts due to transportation.

5. Conclusions

This study analyses the environmental impacts associated with the production of asphalt mixtures composed of two residues: red mud and fly ash. For comparison purposes, dolomite and limestone were considered the reference fillers.

To evaluate the benefits of applying these two residues within asphalt mixtures, three scenarios were modelled in SimaPro. The first scenario considered that the residues received treatment before the asphalt production. The treatment applied to the red mud was washing, drying and crushing, while the fly ash received a thermal treatment (sintering). In the second scenario, the necessity of treatment for the residues is neglected, and both residues are considered inert and ready to be applied into asphalt pavements without any harm. The third scenario simulated the disposal of the residues in landfills.

The landfilling process revealed the lowest environmental potentials amongst all the scenarios evaluated. However, it is not possible to state that accumulating residues in landfills generates lower environmental impacts than using the residues to compose road pavements since this study does not consider the energy compensation, in which the energy from the incineration process is recovered and designated to a secondary operation. The inclusion of the recovered energy into the LCA analysis would reduce the total impact attributed to the thermal treatment of residues and, perhaps, be equivalent to landfilling.

In some countries (e.g. China), residues as fly ash cannot be deposited in landfills without prior treatment (Kanhar et al., 2020). In this case, the landfilling process would automatically generate higher environmental impacts than using treated residues for construction purposes.

Also, the input data used to model the treatment process of residues taken directly from Ecoinvent database is a variable that influences the results, since the emissions of heavy metals during the incineration process is affected by several reasons, such as type of incinerator, sources and composition of residues and combustion temperatures (Long et al., 2011; Zhang et al., 2008).

Even though the results show higher environmental burdens associated with thermal treatments, some studies suggest this practice can be considered an environmentally friendly option in comparison with landfilling residues due to the unproductive use of land for long-term periods driven by the storage (Iyer & Scott, 2001; Kanhar et al., 2020).

Except for the asphalt mixture fabricated with treated fly ash, the production of asphalt materials, with and without residues, presented similar environmental burdens of impact. The use of residues to produce asphalt mixtures may reduce the environmental impacts of using primary raw materials. However, the feasibility of using residues to compose road pavements highly depends on the amount used, the treatment chosen, and the distances required to transport the residues.

To fully assess the sustainability of applying residues to compose road pavements, it is essential to consider the economic, environmental, and social impacts caused during the long-term service life. Some studies indicate that residues’ incorporation to compose road pavements reduces long-term financial burdens of maintenance (Iyer & Scott, 2001) and potential environmental impacts. However, the outcomes may vary for each scenario evaluated.

Therefore, future works should consider the performance of residues in the asphalt mixtures, the use of different treatment procedures, and distances from residues generators to asphalt plants within a ‘cradle-to-grave’ analysis, where the construction, maintenance, demolition, and end of life phases of road pavements are included.
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ORCID
Mayara S. Siverio Lima ♦ http://orcid.org/0000-0002-7831-0960
M. Hajibabaei ♦ http://orcid.org/0000-0002-0047-9715
L. P. Thives ♦ http://orcid.org/0000-0002-4782-2496
V. Haritonovs ♦ http://orcid.org/0000-0003-3119-2677
C. Queiroz ♦ http://orcid.org/0000-0002-1898-7053
F. Gschösser ♦ http://orcid.org/0000-0002-1273-658X

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