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Recovered Fly Ashes as an Anthropogenic Raw Material

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Abstract: Poland is a country where the commercial power industry mainly uses coal to produce energy. As a result of energy production processes, by-products of combustion are generated, primarily fly ashes. In Poland, these are mostly obtained from conventional coal combustion boilers. Fly ashes from coal combustion account for 1.2% of all industrial waste generated in Poland. In addition, fly ashes are produced by fluidized-bed boilers. These are classified as a mixture of fly ashes and solid calcium-based reaction waste from flue-gas desulphurization, and constitute almost 2% of Polish industrial waste. This paper describes the amounts of fly ashes generated in Poland and considers activities related to their recovery and disposal. The high recovery levels of fly ashes (about 90%) and fluidized ashes (about 98%) mean that these waste products can also be considered anthropogenic raw materials. The use of these materials in the cement industry is an example of industrial symbiosis. Such usage benefits not only the economy but also the environment and, therefore, society as a whole. To describe the use of recovered fly ashes in cement plants, the authors use the anthropogenicity index, which characterizes the level of technological advancement and the substitutability of primary raw materials for secondary raw materials.

Keywords: fly ash; recovery; by-product; waste; anthropogenic raw material; industrial symbiosis; case study

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1. Introduction

The production of energy from solid fuels is associated with the formation of combustion by-products. In spite of continuing activities related to the limitation of solid fuel usage, in many countries, especially in the current geopolitical situation, solid fuels remain the basic energy resource for many economic sectors, especially the generation of power. The combustion of solid fuels such as coal for power generation is negatively associated with CO_2 emissions. However, coal power generation also produces the by-product of fly ash, which is a valuable raw material in many industries.

Hard coal and lignite are the basic fuels used in the power industry in Poland (Figure 1). However, biomass is also used as a solid fuel. Therefore, this is classified as a renewable resource, and CO_2 emissions from biomass in the ETS system are considered to be zero, in accordance with the Commission Implementing Regulation (EU) 2022/388 of 8 March 2022 amending Implementing Regulation (EU) 2018/2066 on the monitoring and reporting of greenhouse gas emissions pursuant to Directive 2003/87/EC of the European Parliament and of the Council (Text with EEA relevance).

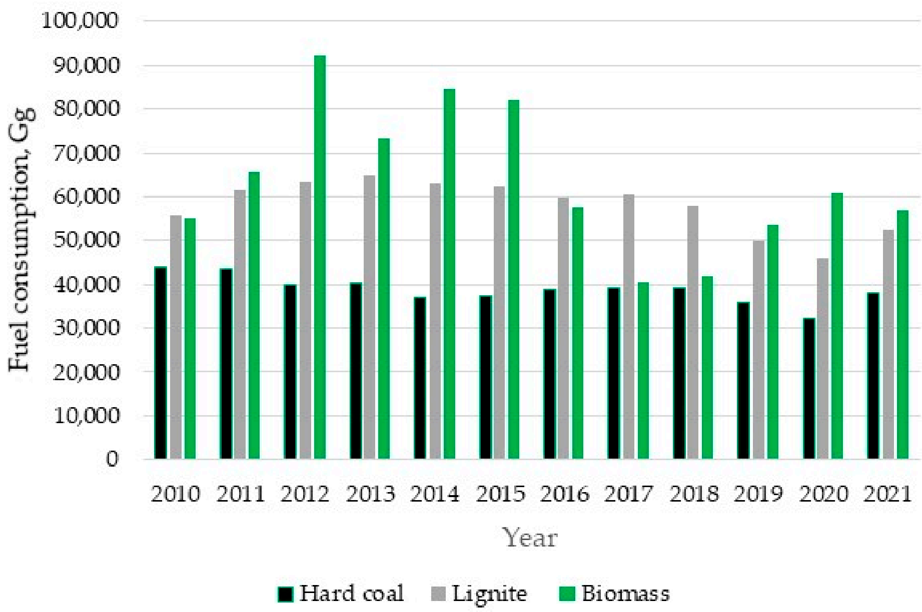

Figure 1. The use of solid fuels in the power industry in Poland, based on [1].

In the European Union, fly ashes produced by the combustion of solid fuels in the energy sector are classified as wastes from thermal processes in accordance with the 2014/955/EU: Commission Decision of 18 December 2014 amending Decision 2000/532/EC on the list of waste pursuant to Directive 2008/98/EC of the European Parliament and of the Council Text with EEA relevance in group 10. Section 10 01 of this list sets out different types of fly ashes from power stations and other combustion plants, and includes the following:

- 10 01 02 coal fly ash;
- 10 01 03 fly ash from peat and untreated wood;
- 10 01 16* fly ash from co-incineration containing hazardous substances;
- 10 01 17 fly ash from co-incineration other than those mentioned in 10 01 16;
- 10 01 82 compounds of fly ash and calcium-based reaction waste from flue-gas desulphurization in solid form (dry and semi-dry methods of desulphurization and fluidized-bed combustion).

Fly ashes are a form of waste characterized by high variability of properties; these determine the possible directions of their application, which result primarily from the type of fuel used, the type of boiler involved, and combustion conditions.

For many years, fly ashes have been used in various industries around the world, such as the construction and ceramic industries. They are also widely used for the purpose of soil amelioration. In addition, research has been conducted on innovative directions of application; these include catalysis, environmental protection, depth separation, zeolite synthesis, CO_2 sequestration, and the recovery of valuable metals [2,3]. However, the most common direction for the use of fly ashes from the power industry is the production of building materials.

The use of fly ashes in the production of building materials, including cement, is an implementation of the idea of industrial symbiosis, which is defined as a process in which waste or by-products from one industry become raw materials for another industry [4–6].

The key benefits of industrial symbiosis include the following [7]:

- Reducing the environmental impact of waste through recovery, reuse, and recycling;
- Creating economic value from waste;
- Reduction in GHG emissions from waste transport and raw material extraction;
- Reduction in dependence on fossil resources;
- Reductions in NO_x, SO_2, and CO_2.

The use of fly ashes and fluidized ashes (FBC fly ash) in cement production as a substitute for cement clinker can reduce the use of natural resources, lower the cost of the raw material, and significantly reduce energy consumption, with a consequent reduction in greenhouse gas emissions [8].

The article presents an analysis of the production and use of fly ashes in Poland, with particular emphasis on cement production as a functioning example of industrial symbiosis.

The presented data indicates that fly ashes are an anthropogenic raw material in the production of cement in Poland. In 2018, 1,545,028 Mg of fly ashes were used in the production of clinker in the EU-28 countries, where almost 30% (459,859 Mg) were in Poland [9].

The analysis was based on statistical data from Statistics Poland [10,11], Agencja Rynek Energii Spółka Akcyjna (ARE SA) [1], and the Polish Cement Association [12].

2. Recovery and Disposal of Fly Ashes in Poland

According to the directive's provisions mentioned above, waste should be subject to recovery processes if it cannot be avoided. If this is not possible for technological, economic, or environmental reasons, it must be disposed of. For the operators of plants where waste is generated, such waste is best considered a by-product.

In the power industry in Poland, as a result of flue-gas cleaning, the main types of waste obtained as by-products are as follows: ashes from coal combustion in conventional boilers; ashes from biomass combustion; and mixtures of fly-ash and solid waste originating from limestone treatment, which are classified as fluidized ashes.

2.1. Coal Fly Ash

Fly ash is one of the residues formed during the combustion of solid fuels in the energy production process. Fly ashes are captured from flue gases in dust collection devices such as electrostatic precipitators or bag filters. Fly ash from conventional boilers is classified on the basis of its chemical composition as either silica (FA) or calcareous (HCFA), depending mainly on the type of coal used. These ashes are mostly composed of glassy balls with some crystalline matter and unburnt carbon [13]. This classification group includes combustion ashes from both hard and brown coal.

Analysis of fly ash data is difficult because of the change in its status from waste to by-product. The reported amount of waste produced has decreased every year, even though the number of ashes generated has remained at the same level (Figure 2). Recovery waste is waste used by the producer or transferred to other recipients for recovery processes (Figure 2). In 2010, 3980.1 Gg of fly ash was generated. By 2012, this amount had increased to more than 4500 Gg. After 2014, when 4489.1 Gg of fly ash was produced, the annual amount gradually declined until 2020, when just 1305.8 Gg of fly ash was generated, before rising again slightly in 2021 to a figure of 1324.3 Gg [10,11].

Fly ashes from hard coal (10 01 02) have been used for many years in various industries in Poland, including the production of building materials (mainly cement and concrete), road building, and mining.

In the years 2010–2012, fly ash recovery was mainly used for the following industrial purposes [1,14,15]:

- Production of building materials (2010–1146.9 Gg; 2011–1284.6 Gg; 2012–1560.1 Gg);
- Cement production (2010–1864.3 Gg; 2011–1601.5 Gg; 2012–1360.5 Gg);
- Road construction (2010–81.2 Gg; 2011–295.0 Gg; 2012–191.8 Gg);
- Mining (2010–728.0 Gg; 2011–757.2 Gg; 2012–698.6 Gg).

During this period, the amount of recovered fly ash represented about 90% of the total generated. Only in the years 2014 and 2015 did the amount of waste subjected to recovery decrease—though dramatically—to 0.1% and 3.3%, respectively. In addition, a fall in the proportion of recovered fly ash to a level below 90% was reported in 2021, when the figure was 84.6%. Fly ash has now been recovered for many years, and less and less of this material ends up in landfills (Figure 2). In 2010, it was 6.1% of the total; in 2021, it was 0.1%.

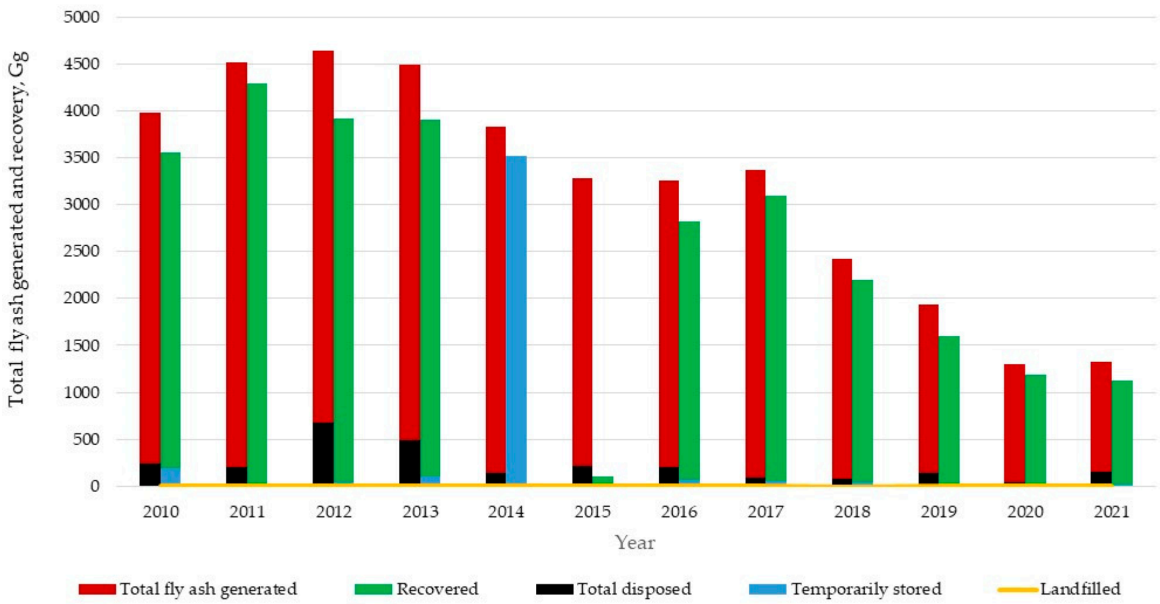

Figure 2. Coal fly ash (10 01 02) recovery and disposal in Poland, Gg/year, based on [10,11].

The Directives 2008/98/EC of The European Parliament and of The Council of 19 November 2008 on waste and repealing certain Directives [16] provide for the possibility of considering a substance or object resulting from a production process, the primary purpose of which is not the production of this substance or object, not as waste, but as a by-product. This can only be done after demonstrating compliance with the requirements for use that must be ensured. If a substance or product is to be considered a by-product, its use must not require further processing other than normal industrial practice. In addition, the substance or object must be produced as an integral part of the production process; and its use must be lawful.

All these requirements can be met in the case of conventional fly ash produced by the combustion of hard coal in Poland. Furthermore, these provisions allow power plants—and combined heat and power plants—to recognize fly ashes as a by-product and not as waste classified as 10 01 02. By such means, totals of fly ash with the code 10 01 02 have fallen in the statistical data. Examples of Polish power plants that now classify fly ashes as by-products are the Opole Power Plant [17] and the Dolna Odra Power Plant [18].

In EU countries, the use of fly ashes in cement and concrete is regulated by the European Standards EN 197-1:2011 Cement—Part 1: Composition, specifications and conformity criteria for common cements; and EN 450-1:2012—Fly ash for concrete—Part 1: Fly ash for concrete—Part 1: Definition, specifications and conformity criteria [13]. According to the European Standard EN 197-1: 2012, they are used for the production of CEM II Portland-composite cement (CEM II/A-V, CEM II/B-V, CEM II/A-W, CEM II/B-W, CEM II/A-M, CEM II/B-M), CEM IV Pozzolanic cement (CEM IV/A, CEM IV/B), and CEM V composite cement (CEM V/A, CEM V/B).

In the cement industry in Poland, fly ashes are used in the production of CEM II, CEM IV, and CEM V. In 2019, the Polish cement industry used 1,863,224.0 Mg of fly ash as an anthropogenic raw material, which represented approximately 96% of the fly ash produced (Figure 3) [12].

Fly ashes from the combustion of hard coal (10 01 02) have been used in the production of cement for many years in various countries, including Poland, Germany, Italy, Austria, the UK, India, and Brazil [9,12,17,19–25]. For example, in the cement industry in Italy, the total amount of coal combustion ashes (10 01 02) used in 2021 was 238,711.14 Mg [25]; in Germany, the corresponding figure was 280,000 Mg [21].

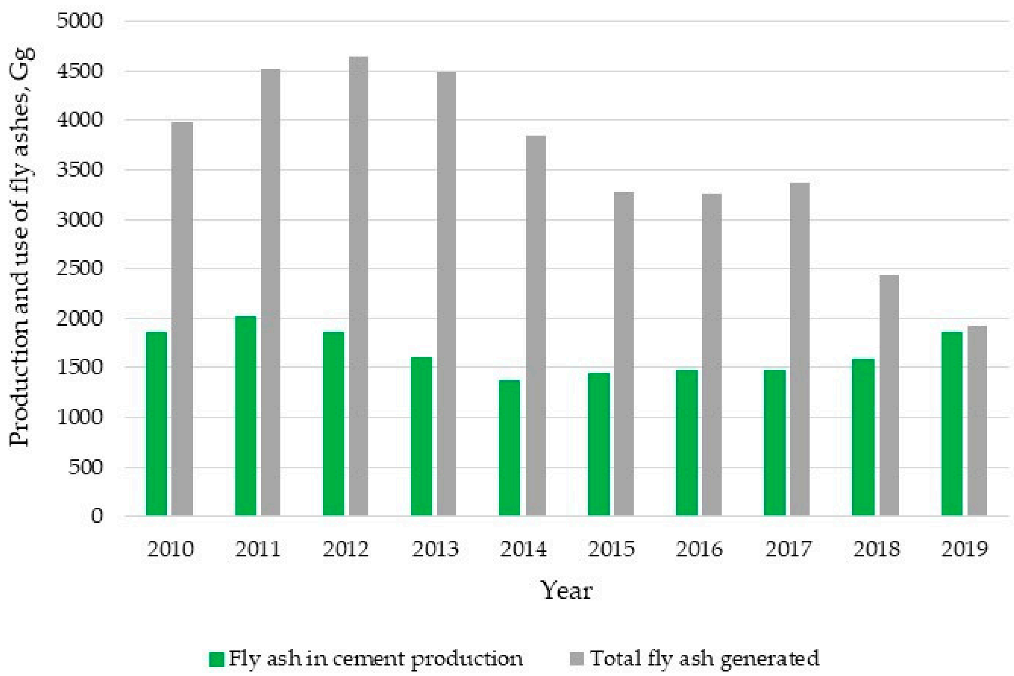

Figure 3. Use of fly ash as additives in clinker and cement production in 2006–2019 in Poland, in Gg, based on [10–12].

2.2. Biomass Fly Ash

A completely different situation can be appreciated in the case of fly ashes from biomass combustion, which are mainly landfilled (Figure 4). These wastes are difficult to recover because of their variable composition and properties [26–28]. Furthermore, these wastes are often characterized by high chlorine, potassium, and phosphorus levels, which greatly limit the possibility of their recovery [29].

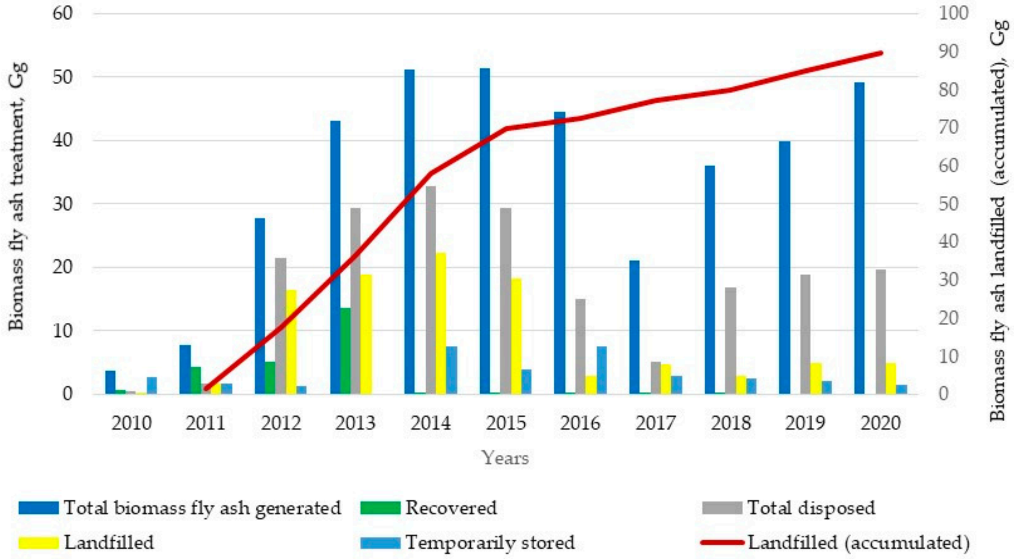

Figure 4. Biomass fly ash (10 01 03) generation, recovery, and disposal in Poland, in Gg, based on [10,11].

The most frequently indicated directions of using ashes from biomass combustion are for improvement in the quality of soils and for fertilization. However, research findings indicate that the use of fly ashes from biomass combustion needs further and much more detailed study, including long-term monitoring and a more precise understanding of potential problems related to such issues as the availability and unavailability of nutrients, the presence of dangerous trace elements, pH changes, interference with microbes, additional salinity problems, dust emissions, ash swelling, and clogging of soil pores [26]. Instabilities in the properties of biomass waste, in particular chemical parameters, mean that this type of waste should always be tested before it is introduced into soil [30].

Biomass waste is also now being tested for use in the production of cement [31,32], and in 2021 the cement industry in Italy used a total of 5301.45 Mg of fly ashes from biomass combustion (10 01 03) [25].

2.3. Fly Ashes from Fluidized-Bed Boilers

Fly ashes from fluidized-bed boilers form a separate group of ashes, which are different from conventional ashes and consist primarily of irregular grains subjected to dehydration or dehydroxylation, and minerals forming primary gangue, as well as fine-grained quartz, anhydrite, desulphurization products, and unreacted sorbents, e.g., $CaCO_3$, free CaO and unburned carbon. There are no spherical grain forms in these ashes.

In waste reporting, fly ashes from fluidized-bed boilers are classified as waste with the code 10 01 82—mixtures of fly ash and solid waste from flue gas desulphurization lime methods (dry and semi-dry methods of exhaust gas desulphurization and incineration in fluidized bed). As with conventional fly ashes, until 2013, recovery was indicated as the method of dealing with them; from 2015, the basic method has been recovery. As a result, the recovery levels of this waste group have remained very high level, at approximately 98%, with figures for individual years as follows: 2021—98%, 2020—98.8%, 2019—97%, 2018—99.5%, 2017—98.4%, 2016—92.8%, 2013—100%, 2012—99.9%, 2011—100%, 2010—100% [10]. However, very low recovery rates were recorded in 2015 and 2014, 1.2% and 0.2%, respectively, when waste was temporarily stored (Figure 5) [10,11].

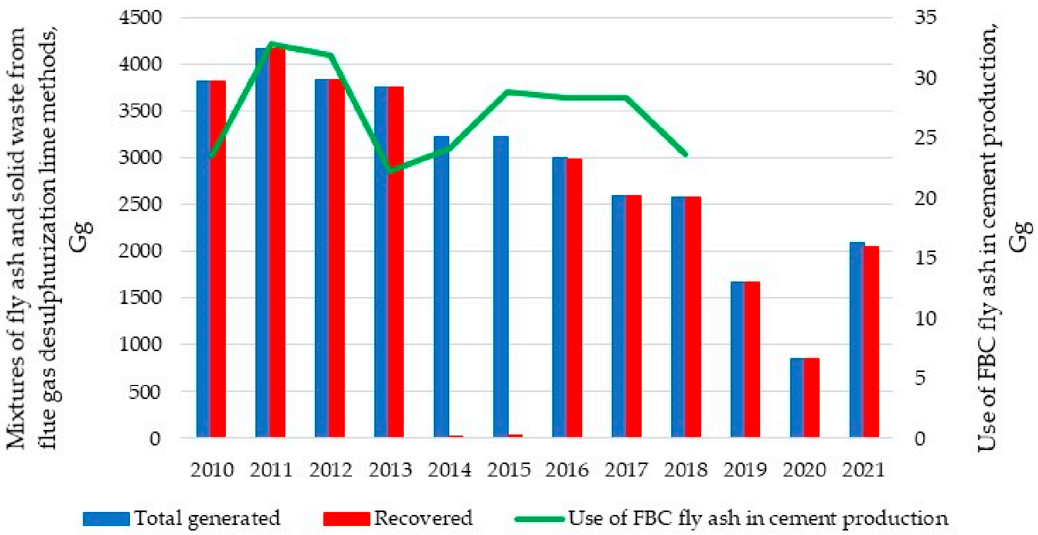

Figure 5. Mixtures of fly ash and solid waste from flue-gas desulphurization lime methods (dry and semi-dry methods of exhaust gas desulphurization, and incineration in fluidized bed) (10 01 82); recovery and use of FBA fly ash as additives in clinker and cement production in 2006–2019 in Poland Gg, based on [10–12].

The most important trend in the management of fluidized ash management in Poland relates to its use in underground mining. Recovered FBC fly ash is also used in cement production as a bonding time regulator, and for the addition of superpozzolan or comprehensive pozzolanic-sulphate [33]. However, the number of fluidized-bed ashes used for cement production in Poland (Figure 5) is smaller than the number of ashes from conventional boilers (Figure 3).

In 2010–2011, the cement industry used an average of 37 Gg/year of fluidized-bed ashes, representing about 1.2% of the waste generated in the 10 01 82 group. In 2019, the corresponding figures were 25.8 Gg and 1.5%, respectively. However, it should be emphasized here that, under classification 10 01 82, fly ashes which include waste products of dry-method desulphurization are also reported, and that all amounts of used waste recovered also represent a reduction in emissions and protection of non-renewable resources.

3. Fly Ashes as Waste, By-Products, and Anthropogenic Raw Materials

As mentioned above, analysis of fly ashes is very difficult because they can be classified as waste or by-products. Therefore, it has been proposed [34] that fly ashes be classified as (1) anthropogenic resources, i.e., mineral substances produced as a result of technological processes, which can and should be raw materials for processing into a building product; and (2) anthropogenic streams, i.e., mineral substances produced on an ongoing basis in technological processes, being raw materials for processing into a construction product. To describe the recovery of fly ashes for use in cement plants, an indicator can be used; specifically, an indicator of anthropogenicity that determines the share of secondary raw material/reduction in the consumption of primary raw material in a product, or in its production, in relation to the total amount of raw materials used. The anthropogenicity index is one of 17 indicators which have been developed to assess the effectiveness of energy companies in the context of the implementation of circular-economy assumptions, and to identify changes that should be carried out as part of the cooperation of entities in the value chain, in order to minimize the amount of waste [35], as part of an ongoing process which includes industrial symbiosis.

Regardless of production volumes, the anthropogenicity index indicates the level of technological advancement and the substitutability of primary raw materials for secondary raw materials. In the case of the cement industry, it should be noted that, in addition to fly ashes and fluidized ashes, other waste materials are also used, including blast furnace slag, REA gypsum, iron bearings, cement kiln dust and by-pass kiln dust [12,14]. Therefore, when analyzing the anthropogenicity index, all wastes used as additives in the production of cement were taken into consideration (Figure 6).

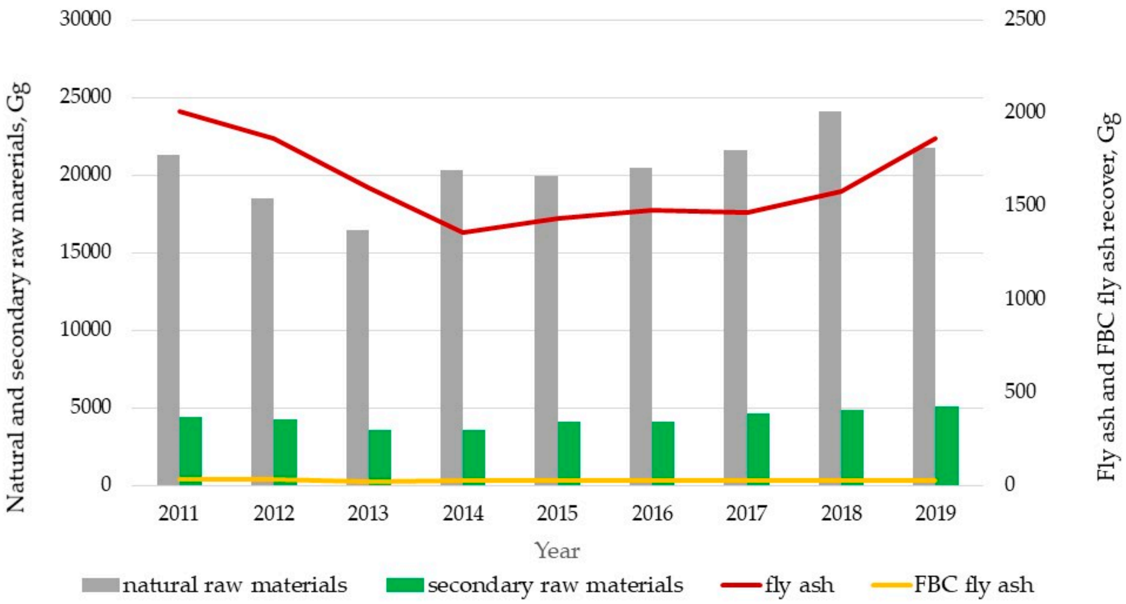

Figure 6. The use of natural and secondary raw materials in the cement industry in Poland, based on [12].

In 2018, the anthropogenicity factor, expressed as the percentage share of waste used for production by the cement industry in Poland, was 16.80% for all secondary raw materials, 5.45% for fly ashes, and 0.08% for fluidized ashes. For 2019, the corresponding figures were 19.0%, 7.0%, and 0.1%, respectively. The anthropogenicity factor calculated for the fly ash used in cement production in Germany in 2019 is 0.5%. The calculations were based on the data from [36]. Despite the relatively low use of fly ashes, the cement industry in Germany is characterized by a high substitution ratio (21.3% in 2019) [9]. The basic waste used is the granulated blast furnace slag (7,020,000 Mg) [9,36].

Taking into account the raw material conditions relating to cement production established by the European Standard EN 197-1:2011, it can be said that the Polish cement industry is characterized by a very high degree of industrial symbiosis, especially with respect to fly ashes and fluidized ashes (Figure 6).

4. Conclusions

Fly ashes from conventional boilers and ashes from fluidized-bed boilers have been subjected to recovery processes in various branches of the economy for many years. In power plants, or combined heat and power plants, fly ash is a waste or a by-product. In other plants, it is a raw material. High levels of recovery of fly ashes from coal combustion and of ashes from fluidized-bed boilers mean that these types of waste can be termed anthropogenic raw materials.

One of the basic directions for using fly ashes and FBC fly ashes involves their use in cement production. This kind of activity is now carried out all over the world, bringing measurable benefits not only to the industry but also to the environment and society as a whole, by reducing the use of natural resources and the emission of greenhouse gases.

For example, in cement plants belonging to the Cemex group in Poland in 2021, fly ashes accounted for 22.96% of the alternative raw materials used, and fluidized ashes for 2.97% [37] This can be seen as an excellent functioning example of industrial symbiosis.

However, some analyses [8,38] have highlighted a potential future problem for cement production using waste obtained from the coal power industry, i.e., the gradual reduction in the production of energy from coal. For the cement industry, the continuing decline in coal power generation brings uncertainty regarding the supply of alternative raw materials in the future, and may necessitate a search for alternative kinds of waste or by-products, such as fly ashes from biomass combustion.

In Poland, due to a large amount of landfilled fly ashes—25.0 million tons (accumulated at the end of 2021) [10,11], an increase in fly ash consumption is expected, although its availability will decrease in the other EU countries [9]. Therefore, Poland is indicated as a country that can be an exporter of fly ashes accumulated in landfills to the neighboring countries, e.g., Germany [9].

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