

## Article

# Sources and Markets of Limestone Flour in Poland

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**Abstract:** Limestone flour is used in a variety of industrial sectors such as power and heat generation, glass-making, paper-making, the construction industry and construction materials manufacturing, the production of agriculture, plastics and rubber goods, as well as coal mining, and environmental protection. This paper aims to characterize and interpret the limestone flour supply and demand trends in the main applications in Poland in the last decade. In order to track the changes of domestic consumption for this commodity, its major users have been surveyed along with analyses of data from the Statistics Poland (GUS). The results showed that during 2009–2018 the coal-fired power plants became the biggest customer of limestone flour utilized as a sorbent in the wet flue gas desulfurization method (FGD). This method has been implemented in the vast majority of Polish power plants. It is estimated that in the last decade the total annual production capacity of limestone flour at the milling plants in Poland increased by 1.5 million tons, to around 6.0 million tons. In the forthcoming years, this is expected to continue to increase despite EU restrictions on fossil fuel use (especially hard coal and lignite). Other promising areas of future growth are: for limestone flour of medium quality—the construction materials manufacturing and the construction industry itself, for limestone flour of the highest quality—the glass industry, while for fine-grained limestone flour of high-quality—the plastics and rubber industries. In terms of resource security, it is worth noting that Poland is a country rich in limestone deposits. However, only some of them, that is, limestone rock varieties of the Jurassic and Devonian ages from the Kielce vicinity in central Poland are suitable for the production of high-quality limestone flour for the needs of the mentioned industries. The paper analyses the potential of the limestone flour production from the available limestone rock deposits with respect to the current and future needs of the domestic market in Poland.

**Keywords:** limestone deposits; limestone flour; limestone sorbents for the power industry; limestone flour for the glass industry; limestone flour for paper filling and coating

## 1. Introduction

Limestone is a sedimentary rock composed mainly of calcium carbonate ( $\text{CaCO}_3$ ), usually in the form of calcite or aragonite. It may contain considerable amounts of magnesium carbonate (dolomite  $\text{CaMg}[\text{CO}_3]_2$ ). Minor constituents also commonly present include clay, iron carbonate, feldspar, pyrite, silica, and quartz. A peculiar variety of limestone rock, owing both to its genesis as well as properties and uses, is chalk [1,2].

Limestone is a rock with an enormous diversity of uses. Most limestone is made into crushed stone and used as a construction material for road base and railroad ballast as well as an aggregate in concrete [3]. Some additional but also important uses of limestone include dimension stone for use in construction and in architecture, roofing granules applied as a weather and heat-resistant coating on asphalt-impregnated roofing, flux stone in smelting and other metal refining processes, as well as the

production of Portland cement. Calcium carbonate is one of the most cost-effective acid-neutralizing agents. When crushed to sand-size or smaller particles, limestone becomes an effective material for treating acidic soils on farms throughout the world. It is also utilized as a dietary supplement added to the feed of farmed animals. Pulverized limestone, called mine safety dust, reduces the explosion hazard in underground coal mines [1–3].

More sophisticated are the products obtained by grinding to the required granulation of limestone, called limestone flour or powdered limestone. They are primarily applied in: the glass-making industry as a stabilizer of molten glass parameters and calcium oxide supplier, the paper industry—as a filler and coating pigment, in the production of stone-mastic asphalt and building materials—as a filler, in the power industry—as a sorbent to desulphurization of flue gases, in the plastic industry and rubber products as filler or extender, and in the fodder industry—as a component of fodders and premixes [1–6].

Despite quite a large resource base of limestone rocks in Poland, only some deposits can be utilized in the lime industry, in particular for the production of limestone flour. The aim of this paper is to address the deposits that can be suitable for the limestone flour manufacturing as well as to assess the trends on the limestone flour markets in Poland over 2009–2018. It was found that raw material of the best quality can be found only in deposits mainly of Jurassic and Devonian origin located in the central part of the country, in the Kielce region (Figure 1).



**Figure 1.** Major areas of occurrence of limestone rocks in Poland (in the upper right corner—the location of Poland on the globe). The age of rocks: 1—Cambrian, 2—Devonian, 3—Triassic, 4—Jurassic, 5—Cretaceous, 6—Tertiary (acc. to [1], modified).

Over the recent decades, the topic of emissions reduction and control has remained an important area of research due to the enforcement of the European Union’s Directive 2001/80/EC [7] and Directive 2010/75/EU [8], as well as various EU members’ government policies in an attempt to minimize the impact on the environment (e.g., in Poland: the Polish Energy Policy 2030 [9] and the Project of

the Polish Energy Policy 2050 [10]). One area in which a great deal of research has been conducted to address this policy was  $\text{NO}_x/\text{SO}_2$  suppression. Following increasingly stringent environmental regulations, since the 1990s the Flue Gas Desulfurization (FGD) technologies have been applied in the majority of Polish coal-fired power plants to abate sulfur emissions. As a consequence,  $\text{SO}_2$  capture in the energy sector in Poland reached over 80%, while the annual consumption of various limestone sorbents in the power industry exceeded 3 million tons. Nowadays, the wet limestone FGD system (utilizing fine-grained limestone flours) is the method most widely used worldwide because of its high desulfurization performance and low operating cost [11,12]. Additionally, the key by-product obtained in the course of this process, that is, FGD gypsum (hydrated calcium sulfate) is profitable for several industrial applications, including cement clinker and Portland cement as well as gypsum binders and plasterboards manufacturing. This also made a significant contribution to the growth in demand for limestone flour in Poland, which was coupled with the large-scale capacity expansion and modernization of the domestic limestone milling plants [11].

## 2. Materials and Methods

For the analysis of the resource base of limestone deposits in Poland and its use, the most important sources of information which were examined including annual publications referring to the mineral resource base in Poland [13–15], reviews of this resources base [1,16], the previous analyses of the limestone raw materials market in Poland [17], as well as the official data published by the Statistics Poland (GUS) [18]. Moreover, the main limestone flour producers were identified and surveyed to estimate their production capacities.

To obtain reliable information on the domestic consumption of limestone flour by the main users in Poland, that is, the power and glass industries, the authors surveyed appropriate establishments, mailing them different survey forms. In the power industry, there were 17 companies surveyed, of which 14 responded. In the glass industry of the 42 largest domestic operations representing all the glass-making branches (container, float, household, technical, and glass fiber) that were canvassed, data were received for 30 glass plants. The demand of the remaining companies was assessed basing on the production data from the Statistics Poland (GUS) and the approximate content of limestone flour in various glassware types. Consumption figures for the non-respondents in the power industry were deduced from their actual energy production and other information, that is, the parameters of the FGD installations operating in these plants as well as prior available data on their sorbent consumption. The information on the demand for limestone flours of other customers was sourced from their websites, press releases, official data published by the Statistics Poland (GUS), industry reports, specialized studies of the branch associations (e.g., the lime and cement industries), and BAT reference documents as well [18–22].

## 3. Results

### 3.1. The Resource Base of Limestone Deposits in Poland and Its Use

Poland has numerous limestone deposits, aside from the highest purity varieties of marbles for sculpturing and architecture. These rocks are documented as suitable for various purposes, mostly for the lime and cement industries, the production of crushed-stone aggregates, and dimension stone. However, practically speaking, this division is of conventional nature, as individual deposits may be extracted by combined cement and lime processing plants, which use cleaner parts of the deposits for the production of limestone-based assortments, while the remaining ones for cement or crushed-stone aggregates [17].

When categorizing by the age of limestone formations, Jurassic limestone is of the greatest importance (constituting nearly 60% of resources), then limestone and related rock of the Cretaceous age (over 21%), Devonian limestone (approx. 8%), Triassic limestone (approx. 8%), Tertiary limestone

(approx. 3%), and marginally—limestone from the Carboniferous, Cambrian and Precambrian periods (Figure 1) [13,16].

Limestone deposits for the lime industry are mainly located in the Kielce region (Świętokrzyskie Province, 60% of the total resources) as well as in the Opole, Łódź, and Katowice vicinity (Figure 1). There were 120 deposits of the total resource of 5549 million tons at the end of 2018 [13]. Limestone and related rock deposits for the cement industry are spread throughout the country, and occur, for example, in the Lublin (26%, mostly marl and chalk), Kielce (17%), Łódź (15%), Warszawa (12%), and other regions. At the end of 2018, the total resource of 69 deposits of these rocks amounted to 12,724 million tons [13]. Limestone deposits for the building and road construction industries are found first of all in the Kielce region (approx. 90% of the total resources) and on the Kraków-Częstochowa Upland, while single deposits occur in the Carpathians (SE Poland), the Sudetes (SW Poland), and on the Lublin Upland (E Poland). At the end of 2018, the total resources of 148 limestone and related rock deposits for the building and road construction industries amounted to 2208 million tons [14].

In 2018, limestone was extracted in 77 quarries including 19 limestone and marl mines for the cement industry, 22 limestone mines for the lime industry (whereby GóraŹdże and Bukowa deposits were exploited for the needs of the cement and lime industries at the same time, Table 1), and also 36 mines for the building and road construction industries [13,14].

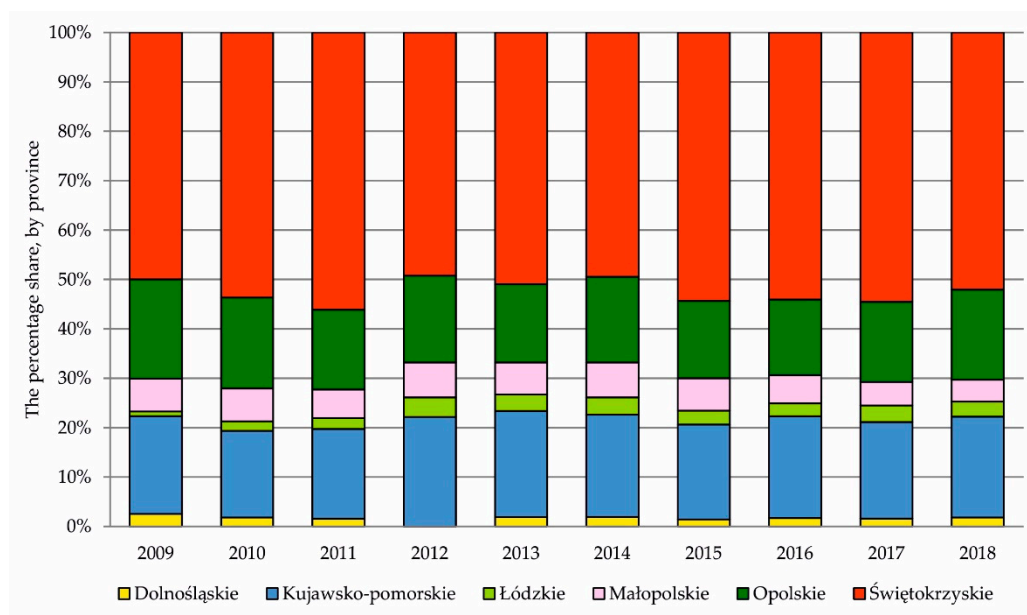
**Table 1.** Mining output of limestone rock in the most important limestone mines in 2018 [13,14,17].

Mine	Province	Group of Deposits <sup>1</sup>	Applications <sup>2</sup>	Mining Output (kt)
Barcin-Piechcin-Pakość	Kujawsko-pomorskie	C	c,a,r,f,g,l	7855
Ostrówka	Świętokrzyskie	L	a,r,g	5878
GóraŹdże	Opolskie	C, L	c,r,n,l	535
Trzuskawica	Świętokrzyskie	L	a,r,g,f,l	4002
Morawica III-1	Świętokrzyskie	A	a,r,f,d	3237
Bukowa	Świętokrzyskie	C, L	r,g,f,l	2585
JaŹwica	Świętokrzyskie	A	a,r,f	2081
Czatkowice	Małopolskie	L	r,g,a	1673
Celiny I	Świętokrzyskie	A	a,r	1308
Szymiszów	Opolskie	A	a,r,f	1018
Izbicko II	Opolskie	L	r,f,l	843
Wierzbiica	Świętokrzyskie	L	a,r,g	738
Połom	Dolnośląskie	A, L	a,r,g,f,l	703
Tarnów Opolski	Opolskie	L	r,g,f,l	645

<sup>1</sup> Group of deposits: A—limestone for the aggregates production, C—limestone for the cement industry, L—limestone for the lime industry. <sup>2</sup> Applications: a—crushed aggregates, c—cement, d—dimension stone, f—limestone fertilizers, g—limestone flour (ground limestone), l—lime, r—limestone rock for sale.

According to the Statistics Poland (GUS) [18], the total mining production of limestone in Poland increased from less than 30 million tons in 2009 to 36.3 million tons in 2018. Last year, approximately 16.4 million tons were represented by limestone for sale used as a flux in the metallurgy of iron and non-ferrous metals as well as in the chemical and sugar industries. According to the official data (GUS), in recent years, the production of limestone flour in Poland has ranged from 1.0 to 1.4 million tons per year (tpy). Yet, the actual volume of this production is much higher. Currently, it is estimated at 5–6 million tpy, whereby in the last decade it increased by at least 20%, mainly due to the development of demand for limestone sorbents in the power industry [11,17].

In terms of the volume of limestone rock extraction, the most important are the deposits located in the Kielce vicinity (50–60%), while deposits in Łódź, Kraków, and Opole regions are of much lesser significance (Table 1, Figure 2).



**Figure 2.** Limestone rock mining output in Poland in 2009–2018 (calculations based on [15]).

Importation plays a marginal role as a source of limestone commodities in Poland. Small amounts (usually several thousand tons per year), predominantly of limestone flour, were primarily imported from the Czech Republic. At the same time, foreign sales of these products were systematically increasing (to 1.0–1.3 million tpy in the last two years) and mainly went to Germany [18].

In the Kielce region, limestone types suitable for the production of limestone flour are the following (Figure 1):

- Devonian limestone from the neighborhood of Kielce, in which  $\text{Fe}_2\text{O}_3$  content is usually between 0.1–0.3% (e.g., in Trzuskawica and Ostrówka deposits);
- Some varieties of Upper Jurassic limestone, characterized by a very high content of  $\text{CaCO}_3$  (approaching 99%) and low content of  $\text{Fe}_2\text{O}_3$  (below 0.3%, sometimes <0.05%), even though the content of coloring oxides shows considerable variability (e.g., in Bukowa, Chęciny-Wolica, Wierzbica, and Anna two deposits) [1,16].

In the Łódź region, the usability for the production of limestone flour (mostly sorbents for flue gas desulfurization, fillers for mineral-asphalt masses, and construction chemicals as well as fodder chalk) is demonstrated by the Jurassic limestone of the Raciszyn and Raciszyn II deposits. The mineral raw material contains up to 98%  $\text{CaCO}_3$  (usually 91–93%), and from 0.11 to 0.64%  $\text{Fe}_2\text{O}_3$ . Locally, higher concentrations of  $\text{SiO}_2$  and clays are found.

In the vicinity of Kraków (Małopolskie Province), limestone flour and sand are obtained from cleaner parts of Carboniferous microcrystalline limestone of the Czatkowice deposit [16]. They are used mainly as sorbents for the flue gas desulfurization in the power industry, and also in the construction, agriculture, or mining (as safety dust) industries. In the predominant part of this deposit, the content of  $\text{CaCO}_3$  exceeds 95%, reaching at times 97–99%. The content of  $\text{Fe}_2\text{O}_3$  is variable, however, sometimes it is not higher than 0.2%. It is possible to selectively extract the limestone rock with lower  $\text{Fe}_2\text{O}_3$  content for the production of high-purity limestone flour, for example, suitable for the building chemistry production [1,16,17].

The majority of domestic milling plants producing limestone flour for various purposes and other limestone-based products are located in close proximity to limestone deposits. It helps to minimize the costs of limestone transportation from a mine to a processing plant. The concentration of the domestic limestone rock resource base in the Kielce region is the reason why the majority of milling plants

are located in the same area or in neighboring provinces, that is, Łódzkie (Łódź) and Małopolskie (Kraków) [17]).

The main producers of limestone flour in Poland are: Lhoist with plants in Bukowa, Tarnów Opolski, Góraźdże and Wojcieszów; Trzuskawica S.A. with the Sitkówka plant; Czatkowice Limestone Mine; Nordkalk with plants in Miedzianka, Chęciny-Wolica, Sławno, and Szczecin as well as milling plants operating at the power plants: Bełchatów, Turów, and Połaniec. The smaller suppliers are Labtar in Tarnów Opolski, Omya in Mielnik, EGM in Wierzbica, WKG in Raciszyn, ZPSChM Piotrowice II, and Dewon in Jaworznia (Table 2).

**Table 2.** The main limestone milling plants in Poland (surveyed producers' data, authors' estimation).

Province	Milling Plant/Company	Estimated Total Production Capacity (ktpy)
Dolnośląskie	Wojcieszów/Lhoist;	<300
Kujawsko-pomorskie	Turów/PGE GiEK	400
	MOWAP Wapienno/Lafarge Cement	
Łódzkie	Bełchatów/PGE GiEK;	2000
	Raciszyn/WKG;	
	Sławno/Nordkalk	
Małopolskie	Czatkowice/KW Czatkowice;	830
	Płaza/Kans-Pol	
Opolskie	Tarnów Opolski/Lhoist;	550
	Tarnów Opolski/Labtar	
Podlaskie	Mielnik/Omya	<10 <sup>1</sup>
	Bukowa/Lhoist;	
Świętokrzyskie	Sitkówka/ZPW Trzuskawica (CRH);	1700
	Wolica/Nordkalk; Wierzbica/EGM;	
	Piotrowice-Zawichost/Piotrowice II;	
	Połaniec/Enea; Jaworznia/Dewon	
Zachodniopomorskie	Szczecin/Nordkalk	200
Total		ca. 5990

<sup>1</sup> natural chalk that does not require milling.

The total domestic production capacity of limestone flour for various applications is currently estimated at around 6.0 million tons annually. In the last decade, it increased by approximately 1.5 million tpy owing to the expansion of some existing milling plants (Bełchatów, Sławno, Wolica) and setting up several new ones (Turów, Raciszyn, Wierzbica, Połaniec) [11]. In the following years, this potential may be increased even more, first of all, due to the extension of the milling plant at the Bełchatów Power Plant (by approx. 0.4 million tpy). Further development of this commodity production capacity in the nearest future is also possible in the EGM plants in Wierzbica and Trzuskawica S.A. in Sitkówka.

### 3.2. The Main Markets of Limestone Flour in Poland

#### 3.2.1. Limestone Sorbents for Flue Gas Desulfurization in the Power Sector

Limestone sorbents (limestone flour and sand and, in some cases, also burnt or hydrated lime) are the most numerous group of reagents utilized in the flue gas desulfurization plants. In Poland, these sorbents are applied, first of all, in the wet limestone method and in fluidized bed boilers as well as, to a smaller extent, in semi-dry and dry methods (Table 3). However, it is estimated that nearly 80%–85% of the global FGD units set up in power plants use wet limestone scrubbing compared to other technologies [12]. In the Polish energy sector, limestone sorbents are used in nearly all existing flue gas desulfurization plants. It results from their broad availability, low price, and—in the case of the wet limestone method—easy utilization of synthetic gypsum arising in the process [11,23]. Depending on the method, the most commonly utilized sorbents are: ground burnt lime and limestone—in the dry processes, hydrated lime and ground burnt lime—in the semi-dry processes, and ground burnt lime, ground limestone, and chalk—in the wet desulfurization technologies [24]. It should however be



noticed that both burnt lime and hydrated lime, when compared to limestone flours, are consumed in negligible quantities (they are utilized only in a few minor power plants).

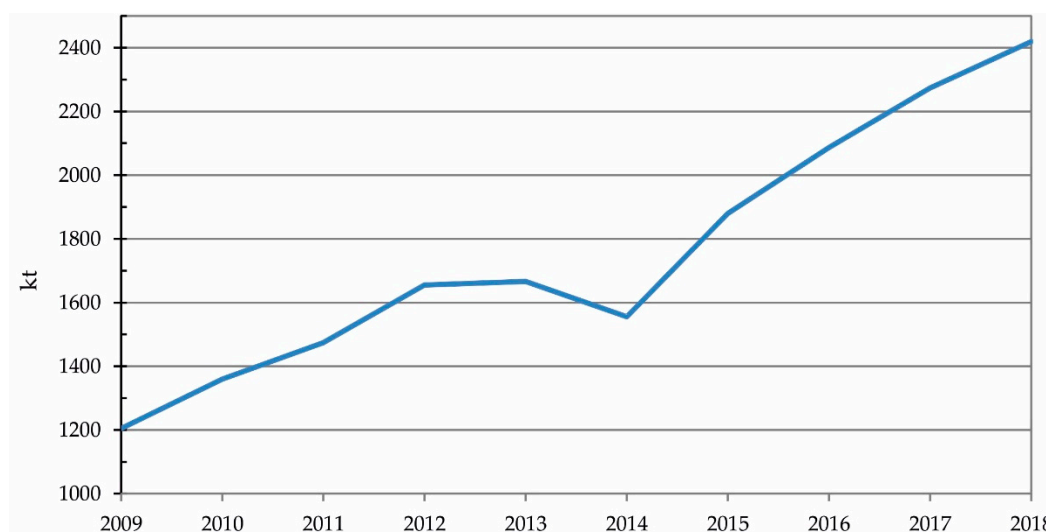
**Table 3.** Flue gas desulfurization installations in the Polish power plants and central heating plants, with estimated demand for limestone sorbent ([25], surveyed consumer data, authors' estimation).

Power Plant (PP)/ Central Heating Plant (CHP)	Method of Flue Gas Desulfurization	Maximum Demand for Limestone Flour (ktpy)	Real demand for Limestone Flour in 2018 (ktpy)
PP Bełchatów <sup>1</sup>	Wet limestone	1600	1486
PP Opole <sup>1</sup>	Wet limestone	450	70
PP Kozienice <sup>1</sup>	Wet limestone	400	185
PP Pątnów <sup>1</sup>	Wet limestone	240	125
PP Połaniec <sup>1</sup>	Wet limestone	160	130
PP Jaworzno III <sup>1</sup>	Wet limestone	135	60
PP Rybnik <sup>1,3</sup>	Wet limestone, Semidry and dry	100	70
Other PPs (Łaziska, Dolna Odra, Ostrołęka, Konin, Turów)	Wet limestone	260	138
CHPs (Warszawa-Siekierki, Kraków, Wrocław, Gdańsk, Gdynia)	Wet limestone	145	80
PP Turów <sup>1,2</sup>	Fluidized bed	ca. 500	ca. 400
PP Łagisza and Siersza <sup>2,3</sup>	Fluidized bed and semidry		
CHP Bielsko-Biała and Chorzów <sup>2,3</sup>	Fluidized bed and dry	ca. 1000	ca. 600
PP Jaworzno II <sup>2</sup>	Fluidized bed		
CHP Żerań, Katowice, and Starogard Gdański <sup>2</sup>	Fluidized bed		
PP Skawina <sup>3</sup>	Semidry	–	–
CHP Łódź 4, Poznań Karolin, Zabrze, Głogów, Lublin- Megatem <sup>3</sup>	Semidry	–	–
CHP Miechowice, Lublin Wrotków, Zgierz <sup>3</sup>	Dry	–	–
CHP Legnica <sup>3</sup>	Absorption	–	–
TOTAL		4990	3344

<sup>1</sup>—fine limestone sorbent <100 µm is used; <sup>2</sup>—coarser limestone sorbent 0.1–1.2 mm is used; <sup>3</sup>—burnt lime CaO or hydrated lime Ca(OH)<sub>2</sub> is used.

Utilization of fine-grained limestone flour (<100 µm) as a sorbent to abate sulfur emissions in coal-fired power plants began in Poland in 1994, when the first wet limestone flue gas desulfurization plant (FGD) was commissioned in the Bełchatów power plant [26,27]. In the subsequent years, FGDs were built in another 17 power and combined power and heating plants (Table 3).

In the period of implementation of desulfurization technology in Polish conventional power plants, that is, in the years 1995–2000, consumption of limestone flour demonstrated a strong growth trend. It was disrupted in the years 2001–2006 as a result of reduced electrical energy production [17]. In order to meet new standards of gas emissions imposed by the European Parliament's Directives number 2001/80/EC and 2010/75/EU [7,8] and obligations contained in the Accession Treaty of Poland to the European Union in 2004 [28], numerous desulfurization installations were established in other domestic plants in the following years. Consequently, the consumption of sorbents with particle size <100 µm for the wet limestone FGDs processes doubled, reaching 2.3–2.4 million tons per year (Figure 3). Currently, 12 power plants and 5 combined power and heating plants in Poland utilize limestone flour <100 µm in the wet limestone FGDs (Table 3). Moreover, approximately 1.0 million tpy of limestone flours of particle size 0.1–1.2 mm (the so-called limestone sands) are utilized as sorbents in fluidized bed boilers of four power plants and five combined power and heating plants [11]. In 2018, the total consumption of milled limestone as an SO<sub>2</sub> sorbent in the Polish energy sector reached 3.3–3.4 million tons (Table 3).



**Figure 3.** Estimated consumption of limestone sorbents for flue gas desulfurization by the wet limestone method in Poland (surveyed consumer data).

The limestone sorbents for FGD processes in the coal-based power plants in Poland are sourced from deposits of high-quality limestone rock of various origins. At present, the varieties of the Jurassic limestone (from Kielce, Łódź and Bydgoszcz regions), as well as the Devonian (Kielce region), Carboniferous (Katowice and Kraków regions), and Triassic ones (Opole region) are of primary importance [11].

The major domestic suppliers of carbonate sorbents are at least seven production companies, that is, Lhoist Polska Sp. z o.o., KW Czatkowice Sp. z o.o., Trzuskawica S.A., Nordkalk Sp. z o.o., Labtar Sp. z o.o., EGM Sp. z o.o., and WKG Sp. z o.o. [11]. The major supplier is the Lhoist Polska, which produces fine-grained sorbents in its two branches: Lhoist Bukowa near Kielce on the basis of Jurassic limestone from the Bukowa deposit, and Lhoist Opolwap in Tarnów Opolski on the basis of Triassic limestone from the Tarnów Opolski deposit. The chemical purity of limestone is one of the primary parameters determining the efficiency of the FGD process. This usually means the minimum content of 94%  $\text{CaCO}_3$ ,  $\text{Fe}_2\text{O}_3$  below 0.4%, and  $\text{MgO}$  also usually below 1%, with variable content of  $\text{SiO}_2$ . Sorbents obtained from the Bukowa deposit are characterized by the higher content of  $\text{CaCO}_3$  (97–98%) when compared to the mineral raw material from the Tarnów Opolski deposit (94.7–96.5%  $\text{CaCO}_3$ ). Limestone rock from the Tarnów Opolski deposit has been also utilized by Labtar Sp. z o.o. for the small-scale production of fine-grained carbonate sorbents.

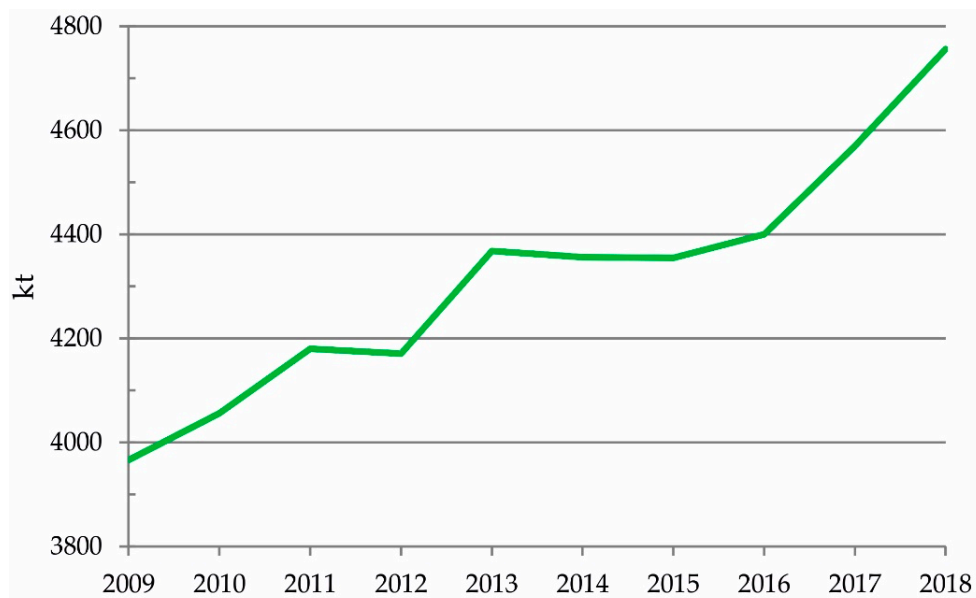
High-quality limestone sorbents are offered by the Czatkowice Limestone Mine in Krzeszowice near Kraków. They are obtained from the Early Carboniferous Czatkowice limestone deposit. These products are characterized by a high content of  $\text{CaCO}_3$  (>96%) and excellent indices: reactivity (RI)—below 2.5 and sorption (CI)—above 120.

Significant quantities of sorbents are obtained from the Jurassic limestone of the Chęciny-Wolica deposit in the Wolica plant near Kielce, operated by Scandinavian Nordkalk Sp. z o.o. Another large supplier of limestone sorbent for the power industry in the region is WKG Sp. z o.o. of Raciszyn extracting two Jurassic limestone deposits. The company offers good-quality sorbents with high  $\text{CaCO}_3$  content (>96%) and excellent indices: reactivity (RI)—below 2.5 and sorption (CI)—above 120. Smaller amounts of sorbents are produced by Trzuskawica S.A. (branch of the Irish CRH) in its Sitkówka plant. They are obtained from the Trzuskawica deposit of Devonian limestone, characterized by high purity and excellent reactivity (RI)—2.32 mol/mol and sorption (CI)—135 as well as by relatively high hardness. Fine-grained limestone sorbents of high purity, excellent reactivity (RI), and sorption (CI) are also produced by EGM Sp. z o.o., operating the Wierzbica deposit near Chęciny [11,19].



### 3.2.2. Limestone Flour for the Production of Dry Mortars

The production of building mortars in the form of dry mixes with chemical additives in Poland dates back to the 1990s. In the following decades, their supplies have grown rapidly (Figure 4).



**Figure 4.** Production of dry mortars in Poland in 2009–2018 [18].

Initially, the most popular among the construction materials were the tile adhesives. They conquered the domestic construction market thanks to such properties as easy mixing with water, long-lasting working properties, easy application, and very good adhesion to various surfaces, as well as reduced slip and mortar resilience. The gradual increase in the use of chemical additives led to the dynamic development of the construction mortars sector, giving rise to the establishment of many companies producing dry construction mixes, including both international corporations (e.g., Baumit, Knauf, Kreisel, Henkel, Mapei, Saint Gobain) and domestic companies (e.g., Atlas, Piotrowice, Cedat, Bolix, and many others). At the same time, in the face of high supplies of gypsum (being by-product of FGD, in particular) the production of gypsum binders has also developed [23,26,27]. The share and role of products based on gypsum binders have significantly grown, considering similar technological progress and development of the product range as in the case of products based on cement binders.

Taking into account that chemical additives used in the mortars (whether cement- or gypsum-based) became more and more expensive, alternative fillers such as limestone or dolomite flours have been utilized in increasing quantities [29,30]. The use of limestone flour offers not only the reduction of the unit cost of a product, but also—however, to a lesser extent—ensures its improved properties (for example grain size distribution, extended bonding time, etc.).

The general economic situation on the market of dry mortars and, consequently, the demand for limestone flour, depends on the trends in the single-family housing, multifamily residential building as well as public/commercial construction sectors, which are the main users of these products. The renovation and modernization activities in residential housing also encourage demand growth. Taking into account the estimated data on dry cement and gypsum mortar production [18] as well as typical limestone flour percentage in these products [19], it was estimated that the recent consumption of these flours in the production of dry mortars in Poland might reach even 1.0 million tons per year.

Virtually all the producers of limestone flour supply the dry mortars plants, among which the most important are the following: Lhoist, Nordkalk, KW Czatkowice, EGM in Wierzbica, WKG in Raciszyn, and ZPSChIM Piotrowice (Table 2).

### 3.2.3. Limestone Flour for the Glass Industry

Limestone flour of the highest purity has long been used by the domestic glass industry. In the raw material batches for the production of glass, it plays the role of a stabilizer, introducing the calcium oxide CaO, which prevents the glass from dissolving in water. Its presence provides increased gloss and durability of a product and makes the glass more resilient to weather conditions [31]. The calcium oxide may also be introduced into the batch with another raw material: dolomite  $\text{CaMg}[\text{CO}_3]_2$ , provided that the presence of magnesium oxide MgO contained in dolomite is advisable in a given raw material batch [32,33]. In the majority of glasses, the presence of magnesium oxide is beneficial. The exceptions are special kinds of glass, for example, optical, and fiberglass, where the admixture of MgO is unacceptable [21].

Since the beginning of the 1990s, the production of glass in Poland has been continuously growing, and in 2018 it exceeded 3 million tons (Figure 5). This mostly resulted from the increase in container and flat glass production—the two most dynamically developing branches of the domestic glass industry. In the structure of glass production in Poland, as in the rest of Europe, glass containers are prevalent, accounting for 58% of the total glassware sales in 2018, while flat glass (mainly for building and automotive applications) represented 34% [33,34]. Household glass (tableware, craft, and crystal glass), technical glass (laboratory, lighting, and electro-technical), and a wide range of products based on glass fiber were of lower importance.

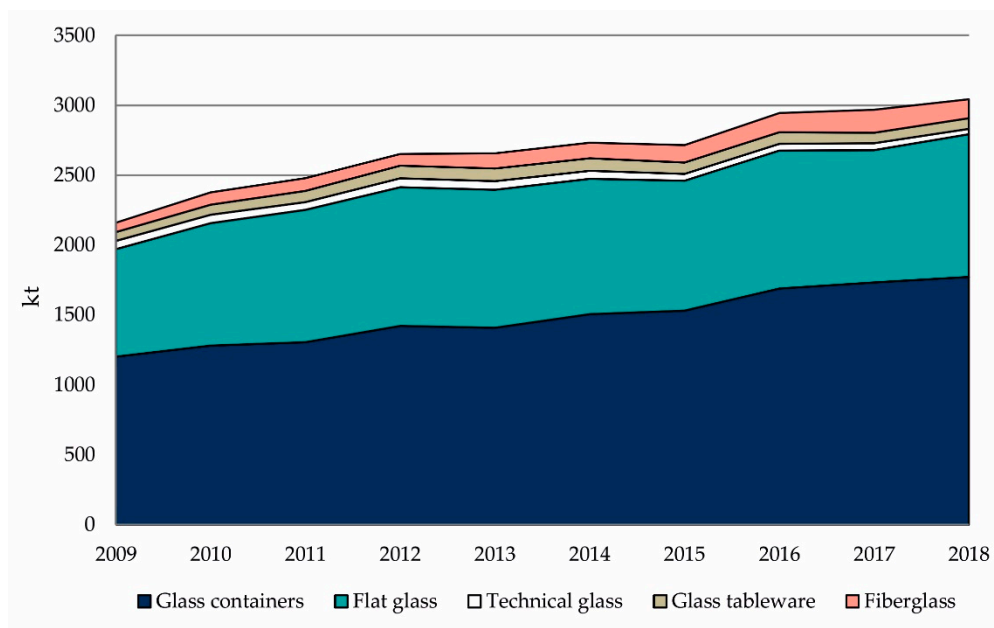


Figure 5. Production of glass in Poland in 2009–2018 [18].

The development of glass production has forced increased demand for limestone flours, and—consequently—their increased domestic sales. Typical container glass contains 9–12% of CaO [21,35] with a small admixture of MgO (0.2–3.5%). In the case of flat glass of the float type, the contents of CaO in the raw material batch may reach 8.5–11.2%, while in household and crystal glass it is much lower (maximum 5%). Based on the mentioned percentages, the volume of domestic consumption of limestone flour in the production of a given type of glass has been evaluated. As a result, the current demand for limestone flours in the container glass manufacturing (taking the present level of glass cullet recycling into account) may be assumed at 180–220 kt per year. This accounts for 80% of these flours' consumption in the glass industry. In the case of float glass, the annual consumption of limestone flour may reach approximately 40 kt (ca. 15% of the total consumption), while the production of household and other types of glass requires ca. 10 ktpy (approx. 5%). The total

current limestone flour consumption in the glass industry in Poland may be estimated at 240–270 kt per year.

In recent years, limestone flours for glass production have been sourced only from the deposits located in the Kielce region, including the Devonian limestone from the Trzuskawica (Trzuskawica S.A.) and the Ostrówka (Nordkalk) deposits as well as Upper Jurassic limestone from the Bukowa (Lhoist), the Chęciny-Wolica (Nordkalk), and the Wierzbica (EGM) deposits. The total production of limestone flour for glassmaking in these plants is estimated at 200–220 ktpy. The domestic supplies have been supplemented by a small importation (maximum 40–50 ktpy), coming mostly from the Omya Group plants in the Czech Republic [17]. In the nearest future, with favorable forecasts for the glass industry development, further moderate growth in the demand for limestone flour may be expected [34].

### 3.2.4. Limestone Flour for the Paper Industry

In the paper industry, limestone flour, similar to other raw materials, added to the pulp (such as e.g., kaolin) is utilized as the filler as well as the coating material. The use of fillers has enabled the lowering of paper production costs by replacing the part of cellulose (wood) fibers with cheaper raw materials [22].

For centuries, the only method of printing paper sizing was the acid method, and a raw material that dominated in paper filling and coating was kaolin [22,36]. Both economic and ecological reasons (high price of kaolin and progressing self-destruction of acid paper, respectively) contributed to the development and implementation of new methods of paper sizing in a neutral or alkaline environment [22,37]. As a result, calcium carbonate, which is markedly cheaper than other fillers, was gradually becoming the preferred filler for the paper industry, displacing the previous favorite, kaolin. Calcium carbonate comes in two varieties. In its natural form, ground calcium carbonate (GCC) is mined, while precipitated calcium carbonate (PCC) is its synthesized form. PCC is made by purifying the raw material from the mine via dissolution and precipitation. This process allows the adjustment of the properties of PCC according to the customer's needs.

The first company in Poland to implement the alkaline/acid-free method of paper sizing was International Paper Kwidzyn S.A. (in 1995), which built a precipitated calcium carbonate (PCC) plant next to the paper mill. Other companies that changed the technology were: Frantschach Świecie—at present Mondi Świecie S.A. and former Intercell—now Stora Enso Poland S.A. in Ostrołęka, which are currently the largest domestic manufacturers of paper for the production of corrugated board, as well as Konstans Sp. z o.o. in Konstancin Jeziorna. The ground calcium carbonate (GCC) has been utilized by Arctic Paper Kostrzyń S.A. in Kostrzyń upon the Odra River—the largest offset printing paper manufacturer in Poland and the second largest (after International Paper Kwidzyn) producer of graphic paper. The supplier of GCC for this company is presumably the Swedish branch of the Nordkalk Company [38–40].

The constant trend observed over the last decade on the Polish paper market has been a growing demand for paper and coated board for the production of packaging (cartons, boxes, cases) coupled with increased consumption of coating pigments. As a result, some paper products may contain even more than 50% of pigments (both kaolin and GCC + PCC or their mixes). According to the Statistics Poland (GUS) [18], in 2009–2018 the production of paper and board in Poland grew by nearly 150% (to 4.9 million tpy, Figure 6), while the production of paper or board cartons and boxes grew by 175% (to 2.7 million tpy.) At the same time, the production of corrugated board doubled to 2.4 million tpy. Another characteristic feature of the paper market in Poland, as in other countries in the world, is the declining demand for newsprint paper (since 2009 its production decreased by 60%, to ca. 70 kt in 2018) as well as flat demand for other graphic grades (which stood at ca. 700 ktpy). This is a clear indication of the reorientation of the paper industry towards the production of special kinds of paper and carton packaging resulting from, among other reasons, continuing digitalization [38–40].

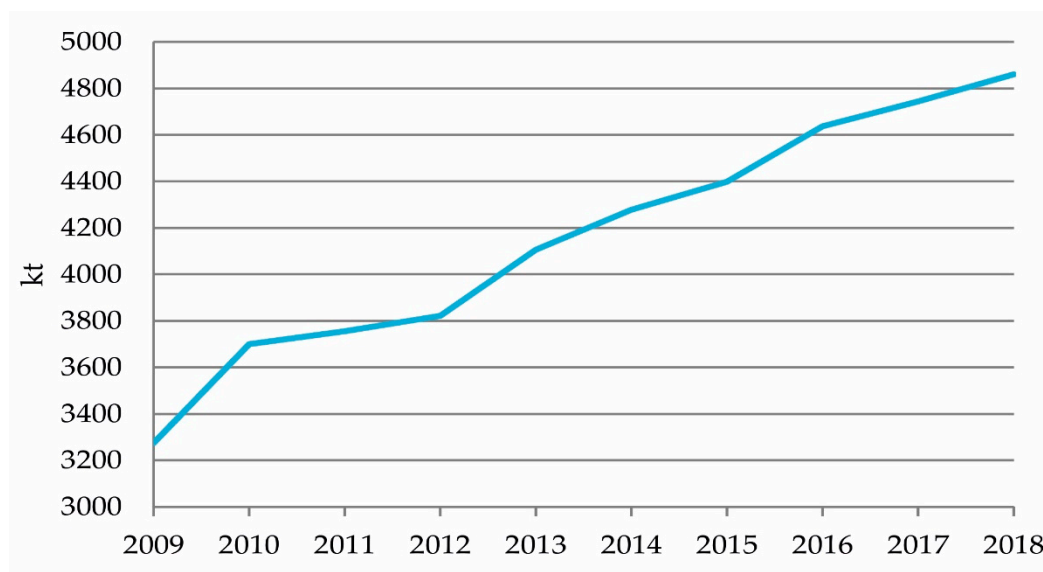


Figure 6. Paper and board production in Poland in 2009–2018 [18].

In order to estimate the demand for fillers and coating pigments, the authors used an exemplary comprehensive record of raw materials used in the production of high-quality wood-free papers for printing and writing in one of the largest European paper plants [22]. According to the most recent available data, this plant's production reached approximately 1228 million tons in 2012. Per 1 ton of the paper manufactured, 385 kg of fillers ( $\text{CaCO}_3$ ) and coating pigments ( $\text{CaCO}_3$  and kaolin) were utilized. This means the use of 472,780 tons of  $\text{CaCO}_3$  and kaolin, which represents 38.5% of the weight of the paper produced. Adopting the same proportion, the annual consumption of fillers and coating pigments for this kind of paper in Poland may be estimated at approximately 270–280 kt.

Possibilities for the future consumption development of both ground and precipitated calcium carbonate (GCC and PCC) in the paper industry in Poland stem from, first of all, the production of coated boards for packaging (cartons, boxes, and cases) as well as special paper manufacturing [41]. This development may be limited, however, by the common striving for the increase of the share of recycled paper (secondary fibers) in the pulp as it is an indispensable source of cheap raw material, and the technology of its processing does not require the utilization of fillers [42,43].

### 3.2.5. Other Users

In addition to the above applications, domestically produced limestone flours are utilized in [17]:

- Agriculture—as the so-called fodder chalk in the animal feed industry and as well as fertilizer (fertilizer chalk, often granulated) to liming of acid soils;
- Road construction—as a filler in the mineral-asphalt masses for road bases and bonding layers;
- Production of concrete and precast units—as the so-called filler aggregate (replacing cement), also in self-consolidating concrete mixes [44–47];
- Production of roof coverings (tar paper and bituminous shingles)—as a filler;
- Coal mining—for the prevention of coal dust explosions in underground coal mines;
- Environmental protection—for water treatment, waste water neutralization, and sludge hygienization;
- In the plastics and rubber industries—as a filler [48,49].

The estimation of limestone flour consumption in the mentioned economy branches in Poland is difficult due to the large number of users and the dispersion of suppliers throughout the country. Presumably, the greatest quantities of limestone, even up to 1 million tons per year, are consumed in agriculture as fertilizer and animal feed as well as in road construction. Other consumers are of lower importance; their annual demand may be estimated at 30–150 kt per year.

#### 4. Discussion and Conclusions

There is a rich resource base of limestone rock in Poland. In the last decade, the domestic mining production of limestone varied from 30 to 40 million tpy, demonstrating a growing trend. Limestone rock from individual deposits has numerous and different applications. This mineral raw material is commonly used in the production of cement and lime as well as crushed-stone aggregates, construction products, and fertilizers. In the mid-1990s a new important direction of this rock utilization, however in the ground form, emerged in Poland. Limestone flour, which is the subject of this study, over the last 20 years has become ever more important, mainly due to its growing consumption in the power industry in the wet limestone FGD systems. The installation of FGD was forced by increasingly stringent environmental regulations regarding the emission of harmful gases from the power industry [20,50]. This played a crucial role in the demand growth for the domestic limestone industry products in Poland, and therefore, the condition of this industry itself. The development of limestone flour utilization played a decisive role in the transformation of the Polish power industry towards reducing its adverse impact on the environment. Currently, the consumption of fine-grained limestone flour as an SO<sub>2</sub> sorbent in the power industry may be estimated at 2.3–2.4 million tpy, while the total demand for ground limestone in the energy sector in Poland may approach 3.3–3.4 million tpy.

According to the Energy Policy of Poland until 2030 [9], in the coming years, the domestic demand for limestone sorbents in the power industry should remain on the current level due to recent or planned modernization of individual power plants. Moreover, in the nearest future, new power units equipped with wet limestone flue gas desulfurization installations are to be commissioned in the Jaworzno III and Turów power plants [10]. As a result, beginning from 2021, the total demand for limestone flours in the domestic power industry may increase by another 200–300 ktpy, to approximately 3.6–3.7 million tpy, of which approx. 2.6–2.7 million tons will be utilized in the FGD process. By 2050, the use of hard coal and lignite in electricity generation in Poland may be reduced from almost 80% in 2018 to approx. 30%. Therefore, after 2030 the old run-down power units will be gradually withdrawn from use. They will be replaced mainly by gas turbines and renewable energy sources, such as off-shore wind turbines on the Baltic Sea, possibly also by nuclear energy. The power units that were built in 2017–2020 and some of the older but modernized ones will remain in operation. As a result of the transition to renewable energy, by 2050, the total demand for limestone sorbents is expected to decrease to a maximum of 1.7 million tons per year, of which approx. 1.2 million tons will be utilized in the wet limestone method of the flue gas desulfurization.

Other recipients of limestone flours, that is, manufacturers of dry mortars, glass, paper, and other goods have a lower share in the structure of these commodities' use in Poland. The total recent consumption of limestone flours in the mentioned industries may be estimated at 2.5–2.6 million tpy, while the future growth opportunities will depend mainly on the situation in the construction sector and on the general economic situation of the country.

It is worth mentioning that the key by-product of the wet limestone FGD method, that is, synthetic gypsum has been used in increasing quantities in the production of cement clinker and Portland cement (recently almost 70% of the total gypsum utilized by the cement industry) as well as gypsum binders and plasterboards manufacturing. The recent annual consumption of 2.4 million tons of limestone flour has resulted in the generation of around 3.3 million tons of synthetic gypsum, almost entirely consumed in the mentioned applications.

Among the deposits extracted for the production of various limestone-based commodities, including limestone flour, those located in central Poland, especially in the Kielce region (50–60% of domestic output), are of key importance [11]. The principal feature determining the volume of output is the mineral quality (high content of CaCO<sub>3</sub> and low content of coloring compounds, mainly Fe<sub>2</sub>O<sub>3</sub>). Types of limestone from the Kielce region, suitable for the production of limestone flour, are first of all Devonian limestone from deposits located in the close vicinity of Kielce (e.g., Trzuskawica and Ostrówka deposits) as well as some types of Upper Jurassic limestone of high chemical purity (e.g.,



Bukowa, Chęciny-Wolica, Wierzbica, Anna two deposits). Usability for the production of limestone flours, utilized mostly as sorbents in the FGD systems and as fillers for stone-mastic asphalt and dry mortars, is also demonstrated by Jurassic limestone of Raciszyn and Raciszyn II deposits in the Łódź region as well as by Carboniferous limestone from the Czatkowice deposit in the Kraków vicinity.

The total production capacity of domestic milling plants producing limestone flour for various applications is currently estimated at 6.0 million tons per year. It is expected to increase even more, mainly due to the expansion of the mills at the Bełchatów power plant and possibly also those in EGM in Wierzbica and Trzuskawica S.A. in Sitkówka plants. It is envisaged that available domestic sources of limestone flour of appropriate quality make it possible to meet the growing demand of these new installations.

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