

Modelling GHG emissions in the mineral products industry in Poland: An uncertainty analysis

Charkovska N. V.¹, Bun R. A.^{1,2}, Nahorski Z.³, Horabik J.³

¹Lviv Polytechnic National University
 12 S. Bandera str., 79013, Lviv, Ukraine
 ²Academy of Business in Dąbrowa Górnicza
 1C Cieplaka str., 41-300, Dąbrowa Górnicza, Poland
 ³Systems Research Institute of the Polish Academy of Sciences
 6 Newelska str., 01-447, Warsaw, Poland

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An improvement of methods for the inventory of greenhouse gas (GHG) emissions is necessary to ensure effective control of commitments to emission reduction. In this article the mathematical models of greenhouse gas emission processes from cement, lime, and glass production at the level of individual plants in Poland have been analysed. Results of the spatial analysis are presented in the form of a geo-spatial database of emissions, and visualised as layers on digital maps. Uncertainty of the inventory results is calculated using the Monte Carlo approach.

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1. Introduction

The main object of international agreements aimed at eliminating the effects of climate change and stopping warming in our planet is greenhouse gas (GHG) emissions. For these agreements, knowledge of estimates for GHG emissions and absorptions at the national level is very important [1]. However, a spatial distribution of GHG emissions within different regions (and even production enterprises) of some countries is needed in view of the requirements of many climate models [2].

Knowledge about the location of the biggest GHG emission sources is useful for the decisionmaking process, concerning emission reductions within an individual country or region. The GHG spatially distributed modelling shows the places where emissions actually occur. The usage of this type of inventory provides an opportunity to improve the inventory process and to reduce its overall uncertainty [2-4].

Uncertainty estimation is an integral part of the multifaceted process of GHG inventory taking. High-quality uncertainty estimates for a GHG inventory are crucial for the implementation of mechanisms under the Kyoto Protocol (such as Emissions Trading, the Clean Development Mechanism, and Joint Implementation), as well as for establishing new treaties of environmental protection [5]. The assessment of the uncertainty of GHG inventory results at the national level as well as at the level of individual emission sources is an extremely important problem due to the fact that incorrect estimates may have a significant impact on the process of trading quotas for greenhouse gas emissions. The results of a GHG inventory have a practical sense only together with estimates of uncertainties

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in the input data (statistical data on the results of economic activity, emission factors, etc.) and the output data (emissions) [6].

At present, a large number of industrial enterprises are involved in the GHG emission reductions programme for 2013–2020. Therefore, the assessment of uncertainties is an important step in the carrying out the GHG inventory. It gives the possibility of verifying compliance with obligations that a certain country, region or particular production plant have accepted. If the emission estimates, taking into account uncertainties, do not exceed the level of emissions set by international or local agreements, at the appropriate time interval, we can confidently confirm real emission reductions.

2. Specifics of emission processes caused by the mineral product industry

Every year the national inventory report (NIR) on GHG emissions is prepared in Poland [7]. The data of these reports are used to certify fulfilment of international obligations. Based on the classification of the Intergovernmental Panel on Climate Change (IPCC), the Industry sector includes GHG emissions from the physical and chemical transformation of materials during the production of its main industrial products.

In accordance with Poland's National Inventory Report to the United Nations Framework on Climate Change, the Industrial sector was responsible for 7% of all GHG emissions in Poland in 2010 [7]. Carbon dioxide emissions from the cement industry are about 5% of the total GHG emissions. Approximately half of these emissions is caused by industrial chemical processes, and the remainder comes from fuel combustion in cement production, which is included in the Energy sector.

According to the Polish NIR for 2012, the CO_2 emissions from cement production amounted to 6,693,000 tons (2.A.1 Cement Production). The share of this category is 1.6% of total GHG emissions from all sectors of human activity and 67.5% in the IPCC Mineral Products subsector of the Industrial Processes sector [7].

The amount of clinker produced in Poland was 11,767,000 tons in 2010 [8,9]. The cement industry is widely developed in 7 of 16 voivodeships.

The cement industry is represented by 11 cement production plants with a full technological cycle, one production plant for cement grinding, and one production plant for alumina cement. A full production cycle means all stages of cement production, in particular the processes of clinker calcination and cement grinding. The largest cement producers are Góraźdźe Cement S.A. (Heidelberg Cement Group), Lafarge Cement S.A. (Lafarge Group), and Grupa Ozarów S.A. (CRH Group). The market shares of these groups in total cement production are 26%, 21%, and 17%, respectively [8].

Carbon dioxide emissions in the cement industry occur during the production of clinker, which is an intermediate component in the cement manufacturing process. During the production of clinker, 95% of the limestone used, which consists mainly of calcium carbonate, CaCO₃, is calcined to produce lime, CaO, and CO₂ as a by-product. The CaO then reacts with silica, aluminium, and iron oxides in the raw materials to make the clinker minerals, which are predominantly hydraulic calcium silicates. During these reactions, CO₂ is not emitted any further [1].

The main challenge in the estimation of CO_2 emissions from cement production is to deal with the varying CaO content in clinker. A good practice is to estimate CO_2 emissions using data for clinker production as well as for the CaO content of the clinker, and to correct for the loss of the so-called cement kiln dust (we denoted this as the coefficient K_{CKD}). This approach assumes that 100% of the CaO comes from a carbonate source (e.g. CaCO₃ in limestone). The cement kiln dust may be recycled in the kiln partially or completely. Any cement kiln dust that is not recycled can be considered lost to the system in terms of CO_2 emissions [8, 10].

Below we present the developed mathematical model of the emission processes from cement production that was used for spatial inventory of GHG emissions.

3. Mathematical models of GHG emissions

In terms of GHG inventory, each cement production plant is considered as a point-type source of emissions. Carbon dioxide emissions from a single point source are calculated as a product of the quantity of clinker produced, the CaO content in clinker, and the cement kiln dust losses according to the formula below:

$$E_{\text{Cement}}^{\text{CO}_2}\left(\zeta_n\right) = F_{\text{stat}_{\text{clinker}}}\left(\zeta_n\right) \cdot K_{\text{clinker}}^{\text{CO}_2}\left(\zeta_n\right) \cdot K_{CKD},$$

$$\zeta_n \in \Xi_{\text{cement}}, \quad n = \overline{1, N_{\text{cement}}},$$
(1)

where:

 $E_{\text{Cement}}^{\text{CO}_2}$ is the amount of annual carbon oxide emissions from the cement plant;

 $F_{\text{stat}_{\text{clinker}}}$ is the activity data on (the quantity of) clinker production for the cement plant ζ_n ; $K_{\text{clinker}}^{\text{CO}_2}$ is the emission factor for clinker for cement plant ζ_n ;

 K_{CKD} is the correction factor for losses of cement kiln dust (it was assumed that $K_{CKD} = 1.02$); Ξ_{cement} is the set of cement production plants;

 N_{cement} is the number of these plants.

The proposed approach allows the GHG inventory at point-type emission sources to be carried out taking into account the specifics of the statistical data and the geographic location of the emission sources. The GHG emissions for one cement production plant are a function of the activity results in production and the proper emission coefficients, although the location of these emissions depends on the geographical coordinates of the production plant.

Geographic references of emission sources are not directly reflected in the mathematical description of the emission processes. However, a developed geoinformation system (GIS) distinguishes the variable n, which describes the geographic coordinates of cement production plants.

The emission factor $K_{\text{clinker}}^{\text{CO}_2}$ is calculated as the ratio of the mass of CO₂ emitted into the atmosphere from a unit mass of clinker. Traditionally, this coefficient is represented in kilograms of CO₂ per ton of clinker. In this study it was accepted that $K_{\text{clinker}}^{\text{CO}_2} = 529 \text{ kg}_{CO2}/\text{t}$.

The amount of produced clinker/cement is known at the national level according to GUS (Główny Urząd Statystyczny – Central Statistical Office of Poland) yearbooks [9]. The statistical information on cement production by each plant is unknown. To carry out spatial GHG inventory from cement production, alternative sources of information were used. For instance, data on cement production capacities are available from the official websites of the main cement producers. The Polish Cement Association reports annually on its website about the situation in the cement industry in a special yearbook (Informator SPC). In the yearbook for 2010 there is a diagram that displays the shares of each cement group in the Polish cement production sector in 2009.

As data on the amount of cement produced by each plant are needed for the compilation of GHG spatial inventory, we used information from diagrams and production capacities from enterprise websites as an effective indicator of the disaggregation of the national data to the level of cement plants. We assumed that the amounts of cement, produced by each plant are distributed proportionally in relation to their nominal capacities.

The quantity of lime/quicklime produced in Poland amounted to 1,798,900 tons in 2010 [9]. There are seven large industrial groups in Poland which mine limestone, and based on this limestone produce different types of lime (quicklime, slaked lime, dry calcium hydroxide powder, milk of lime, lime putty, etc.). Germany, France, Poland, Belgium, Spain and Italy are the largest producers of lime in the EU-27. The production of these countries altogether accounts for about 20% of the world's total lime production [10].

The technological process of the lime production emits carbon dioxide through the thermal decomposition (calcination) of the CaCO₃ in the limestone to produce quicklime CaO, or through the decomposition of dolomite, CaCO₃·MgCO₃, to produce dolomitic quicklime, CaO·MgO. Carbon dioxide emissions are calculated as the product of the quantity of produced lime and the emission factor for the lime [1].

Similarly to cement plants, lime production plants are also represented as point-type emission sources. To compile an inventory for this category of emission sources, we use the following mathematical model:

$$E_{\text{Lime}}^{\text{CO}_2}\left(\zeta_l\right) = F_{\text{stat}_{\text{lime}}}\left(\zeta_l\right) \cdot K_{\text{lime}}^{\text{CO}_2}\left(\zeta_l\right), \quad \zeta_l \in \Xi_{\text{lime}}, \quad l = \overline{1, N_{\text{lime}}}, \quad (2)$$

where:

 $E_{\text{Lime}}^{\text{CO}_2}$ is the amount of carbon oxide emissions from the lime production plant ζ_l ;

 $F_{\text{stat}_{\text{lime}}}$ is the activity data on (quantity of) lime production for the plant ζ_l ;

 $K_{\text{lime}}^{\text{CO}_2}$ is the emission factor for quicklime (including the dolomite quicklime) for the plant ζ_l ; it was assumed that CO₂ emission factor is equal to 785 kg CO₂ per tonne of lime;

 Ξ_{lime} is the set of lime production plants;

 N_{lime} is the number of these plants.

Emission factors may vary for different types of quicklime with high calcium or dolomite content. The above mathematical model can be applied at the level of separate lime production plants, provided that detailed input data are available.

The mathematical model for GHG emission processes from glass production is described in detail in publication [11].

4. Uncertainty estimation

The total uncertainty of GHG emission inventories depends on uncertainties for all the input parameters. These input uncertainties can be combined into an uncertainty for the total emission estimates using the statistical tools specified in the IPCC Guidelines [1, 12]. For such an analysis it is important to have independent uncertainty ranges for emission coefficients, statistical data and other parameters of the inventory process [4].

An important factor in the uncertainty of statistical data is the specificity of the functioning of statistical offices and agencies with all the features of presenting and reporting such information. The uncertainty of statistical data in industrial activity (activity data in terms of a GHG emission inventory) is quite high.

The geographical references of cement, lime, and glass production plants as large-scale point-type emission sources are essentially new independent parameters of emission processes. The usage of geospatial databases extends the knowledge of emission processes and creates new opportunities for the research and reduction of uncertainties that appear during a GHG inventory.

The main factors that impact uncertainty during a GHG inventory are algorithms of disaggregation of industrial activity data (e.g. the level of large industrial enterprises, taking into account production capacity or production volumes, and thus how they cause increasing uncertainty in GHG inventory results), and the uncertainty of used emission factors (the application of specific emission factors for point-type emission sources reduces the uncertainty of GHG inventory results in comparison to the usage of average national or global emission factors) [6].

In this investigation, the assessment of uncertainty of GHG emissions has been carried out using the Monte Carlo approach. As a result, the output value of GHG emission, with its 95% uncertainty range, was obtained for each production plant. The advantage of the Monte Carlo approach is the opportunity to model random or stochastic variables with different distributions and probability density functions, even for emission factors with uncertainty ranges greater than 30%.

For the estimation of uncertainty of GHG emissions from cement, lime and glass production, appropriate effective software has been developed. The main feature of this software is the ability to conduct GHG inventory and uncertainty analysis at different levels (production plants, voivodeships, and the country as a whole). The algorithm for calculating the Monte Carlo method consists of 5 steps:

- 1) setting the probability distribution functions of each parameter of the mathematical model separately for each cement, lime or glass production plant;
- 2) generating pseudo-random data samples of statistical data and emission factor according to the density of the probability distribution;
- 3) using modelled random values of input parameters, estimation of annual emissions (one random emission value is calculated for each emission source) on the basis of mathematical model (1);
- 4) calculation of the total GHG emissions from all sources;
- 5) estimation of the expected value and the 95% confidence interval (lower and upper bounds of the uncertainty range).

The main features of the developed software are the creation of an input file and the setting of the probability distribution for each parameter by the researcher.

The software is implemented using Delphi 7.0. The main form of software, implemented for this purpose, consists of three tabs:

- 1) "Generation" (provides the ability to input the number of realizations, to choose a greenhouse gas for which the modelling will be conducted, to determine the probability distributions for each parameter, and the visualisation of the generated values in a graph);
- 2) "Variational series" (on the basis of values generated in the first tab, a variational series is prepared and displayed in the appropriate fields: the width of interval, the number of partitions, the expectation value, and the lower and upper bounds of the uncertainty range);
- 3) "Histogram" (using the variational series, a histogram is created).

Polish statistical yearbooks [9] do not contain any information on the uncertainties of statistical data on industrial activity at the national level or specific regions. This kind of information at the level of industrial production plants is not available, either. However, according to Polish experts in statistics, the uncertainty ranges of statistical data for different categories of emission sources at the national level are within 2-5% [7]. Analysis of the uncertainty of statistical data on industrial activity in Poland is conducted during preparation of the Polish NIRs.

In the industrial sector there are a few emission source categories and uncertainties for emission factors that are relatively low (an average of 5-10%, in extreme cases, 20-100%). The 2010 NIR reported that the most accurate estimates of emission factors for carbon dioxide have been obtained for the category "Cement production" (relative uncertainty is equal to 15%). It is accepted in Polish NIRs that the emission factors of main GHGs are normally distributed.

5. Results

In our calculations we mainly used "national" uncertainty ranges for statistical data and emission coefficients. In Tabl. 1 data are presented on the uncertainty ranges of statistical data on the activity and respective specific emission factors of carbon dioxide for cement production plants, which are the main sources of emissions in the IPCC Industrial Processes sector. Using GIS, a geoinformation technology has been developed in which model (1) mentioned above is used to estimate the emissions from the production of cement/clinker.

The total carbon dioxide emissions from cement production in 2010 amounted to 6,254,600 tons. The three largest emission sources are the Górażdże plant (1,253,400 tons); the Małogoszcz plant and the Kujawy plant (644,400 tons each) (see Fig. 1). The largest emissions of carbon dioxide are concentrated in Opolskie (1,441,000 tons) and Swiętokrzyskie (1,856,000 tons) voivodeships, and the smallest emissions are in the Lesser Poland (Małopolskie) voivodeship (70,000 tons).

The low emissions in this province are due to the fact that there are only 2 plants operating with small production capacities there. We compared the results of our modelling of CO_2 emissions from cement production with the official data published by GUS, and calculated that our results were 6.5% lower.

№	Name of plants	Clinker production, Gg/year	Activity data un- certainty, %	CO ₂ emission factor, MgCO ₂ /Mg	CO_2 emission factor un- certainty, %	Results of modeling (mathemat- ical model), Gg	Results of modeling (Monte Carlo approach), Gg	CO ₂ emissions uncertainty ranges, %
1	Cementownia Górażdże	2400	2	0.512	15	1253.3	1242.3	± 15.07
2	Cementownia Małogoszcz	1215	2	0.52	15	644.4	632	± 15.09
3	Cementownia Kujawy	1215	2	0.52	15	644.4	632	± 15.09
4	Grupa Ożarów	1144.4	2	0.529	15	617.5	606	± 15.18
5	Cementownia Rejowiec	1065.6	2	0.529	15	574.9	564	± 15.18
6	Cementownia Chełm	1137.5	2	0.529	15	613.7	564	± 15.18
7	Cementownia Rudniki	682.5	2	0.529	15	368.2	361	± 15.18
8	Dyckerhoff Pol- ska Sp. z o.o.	1050	2	0.529	15	566.5	556	± 15.18
9	Cementownia Warta	1320	2	0.529	15	712.2	698	± 15.18
10	Cementownia Odra	350	2	0.529	15	188.8	185	± 15.18
11	Górka Cement	50	2	0.529	15	26.9	26	± 15.18
12	Cementownia Nowa Huta	80	2	0.529	15	43.1	42	± 15.18
	Total					6254.5	6132	± 5.13

Table 1. Assessment of the uncertainty of input data, and results of the calculations of GHG emissions in Polish cement production plants for 2010.

In Tabl. 2 we present the input data (statistical information and emission factors) for uncertainty estimation in the category "Lime production", and the calculated results (the emission values and uncertainty ranges). The results of the GHG spatial inventory from lime production are presented in Fig. 2 and Fig. 3. In this category, the companies which are leaders in terms of emissions are Bukowa, Czatkowice, and Labtar. The largest CO_2 emissions are concentrated in the Swiętokrzyskie (403,400 tons) voivodeship, followed by in the Opolskie (336,200 tons), Lesser Poland (Małopolskie) (201,700 tons), Lower Silesian (Dolnośląskie) (168,100 tons), Kuyavian-Pomeranian (Kujawsko-Pomorskie) and Podlaskie (100,800 tons each) voivodeships, and with the smallest in the West Pomeranian (Zachodniopomorskie) and Lódzkie (50,400 tons each) voivodeships.

GHG emissions from the following Polish glass production plants were analysed: Owens-Illinois Polska S.A. (estimated annual glass production in 2010 – 288.29 Gg); Ardagh Glass Gostyń S.A. (183.59 Gg); Ardagh Glass Ujście S.A. (124.78 Gg); Huta Szkła Warta S.A. (91.80 Gg); Stolzle Częstochowa S.A. (91.80 Gg); Huta Szkła "Jedlice" S.A. (53.07 Gg); Ardagh Glass Wyszków S.A. (53.07 Gg); Huta Szkła Wymiarki S.A. (38.73 Gg); Huta Szkła Sława S.P. (53.07 Gg); Vitrosilicon S.A. (53.07 Gg); Huta Szkła "Czechy" S.A. (37.29 Gg); Saint-Gobain Glass Polska Sp. z o.o. (272.10 Gg); Pilkington Polska Sp. z o.o. (272.10 Gg); Guardian Industries Poland Sp. z o.o. (272.10 Gg); Euroglas Polska Sp. z.o. (272.10 Gg).

We used the following input parameters for the emission calculations and uncertainty estimation: activity data uncertainty -5% (normal distribution; 95% confidence interval); carbon dioxide emission factor $-0.21 \text{ Mg}_{CO2}/\text{Mg}$; CO₂ emission factor uncertainty -10 % (normal distribution; 95% confidence interval); cullet ratio in the glass -45%; uncertainty of the cullet ratio in the glass -15% (normal distribution; 95% confidence interval).

In Fig. 4 the emissions from glass production are depicted for the following voivodeships: Kuyavian-Pomeranian (Kujawsko-Pomorskie) (1 plant), Lublin (Lubelskie) (1), Opole (Opolskie) (1), Silesian (Śląskie) (4), Swiętokrzyskie (2), Lesser Poland (Małopolskie) (1), Subcarpathian (Podkarpackie) (4),



Fig. 1. Thematic map of CO_2 emission from cement production in Poland (thousands of tons, 2010).



Fig. 2. Thematic map of CO_2 emissions from lime production at the level of individual plants (thousands of tons, 2010).

Masovian (Mazowieckie) (4), Greater Poland (Wielkopolskie) (8), Lower Silesian (Dolnośląskie) (3), and Lubusz (Lubuskie) (1). The largest emissions of carbon dioxide are reported in the Greater Poland (87,900 tons) and Silesian (97,200 tons) voivodeships, while the smallest are in the Lesser Poland and Kuyavian-Pomeranian (900 tons each) voivodeships.

№	Name of plants	Lime pro- duction, Gg/year	Activity data un- certainty, %	${ m CO_2} \ { m emission} \ { m factor}, \ { m Mg_{CO_2}/Mg}$	CO ₂ emission factor un- certainty, %	Results of modeling (mathem. model), Gg	Results of modeling (Monte Carlo approach), Gg	CO_2 emissions uncer- tainty, %
1	KW "Czatkowice" Sp. z o.o.	257.0	10	0.767	10	197.2	196.5	± 14.1
2	Labtar Sp. z o.o. Tarnów Opolski	257.0	10	0.767	10	197.0	197.9	± 14.1
3	Nordkalk Sp. z o.o. Miedzianka	64.2	10	0.767	10	49.3	50.1	±14.1
4	Nordkalk Sp. z o.o. Wolica	64.2	10	0.767	10	49.6	49.2	± 14.1
5	Nordkalk Sp. z o.o. Sławno	64.2	10	0.767	10	50.2	49.5	± 14.1
6	Nordkalk Sp. z o.o. Szczecin	64.2	10	0.767	10	49.7	50.8	± 14.1
7	ZW Lhoist S.A. Tarnów Opolski	85.7	10	0.767	10	65.7	66.4	± 14.1
8	ZW Lhoist S.A. Górażdże	85.7	10	0.767	10	65.1	66.6	±14.1
9	ZW Lhoist S.A. Wojcieszów	85.7	10	0.767	10	65.5	65.9	± 14.1
10	Lhoist Bukowa Sp. z o.o. Bukowa	257.0	10	0.767	10	197.7	197.0	±14.1
11	Omya Sp. z o.o. Romanowo	128.5	10	0.767	10	98.5	99.4	±14.1
12	Omya Sp. z o.o. Mielnik	128.5	10	0.767	10	98.9	100.1	±14.1
13	ZPW Trzuskawica S.A. Sitkowka	128.5	10	0.767	10	100.2	99.5	±14.1
14	ZPW Trzuskawica S.A. Kujawy	128.5	10	0.767	10	98.6	99.3	± 14.1
	Total					1383.2	1379.97	± 4.3

Table 2. Assessment of the uncertainty of input data, and results of the calculations of GHG emissions in Polish lime production plants for 2010.

The spatial analysis of CO_2 emissions from large point sources has been done for all the voivodeships in Poland, except for the Pomeranian (Pomorskie) and Warmian-Masurian (Warminsko-Mazurskie) voivodeships, as there is no developed mineral products industry there. The results show that the territorial distribution of emission sources is extremely uneven. The largest emissions are observed in the Opolskie, Swiętokrzyskie and Lubelskie voivodeships.

6. Conclusions

The spatial analysis of the uncertainties of GHG emission inventories in the "Mineral Products" subsector of the "Industrial Processes" sector, namely the production of cement, lime and glass, is appropriate since participation in and implementation of the Kyoto protocol or any other commitments necessarily include not only reporting the amounts of greenhouse gas emissions, but also providing estimation of their uncertainty ranges.

The developed approach and software give the possibility of analysing emission uncertainties at the level of separate plants, voivodeships or for the country as a whole. The obtained results of the mathematical modelling and spatial analysis of GHG emission processes essentially demonstrate low uncertainties of emissions during the production of cement and lime by respective companies. This has a positive impact on the uncertainty of total regional or national GHG emission inventories for all categories of economic activity, thus enabling authorities to take into account this factor when verifying the performance of international agreements on the reduction of GHG emissions.



Fig. 3. Thematic map of CO_2 emissions from lime production at the level of individual voivodeships (thousands of tons, 2010).



Fig. 4. The structure of CO_2 emissions from glass production at the level of individual plants (thousands of tons, 2010).

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Моделювання процесів емісії парникових газів від виробництва основних мінеральних речовин у Польщі: Аналіз невизначеності

Чарковська Н. В.¹, Бунь Р. А.^{1,2}, Нахорскі З.³, Хорабік І.³

¹ Національний університет «Львівська політехніка» вул. С. Бандери, 12, 79013, Львів, Україна ² Академія бізнесу в Домброві-Гурничій вул. Чепляка, 1С, 41-300, Домброва-Гурнича, Польща ³ Інститут системного аналізу Польської академії наук вул. Невельська, 6, 01-447, Варшава, Польща

Вдосконалення методів інвентаризації емісій парникових газів є необхідним для забезпечення ефективного контролю зобов'язань щодо скорочення емісій. У цій статті подано математичні моделі процесів емісії парникових газів від виробництва цементу, вапна та скла на рівні окремих підприємств Польщі. Результати просторового аналізу наведено у вигляді геопросторової бази даних емісій і візуалізовано у вигляді шарів цифрових карт. Невизначеність результатів інвентаризації оцінено з використанням методу Монте-Карло.

Ключові слова: математичне моделювання, емісія парникових газів, промисловий сектор, невизначеність

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