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1. Introduction

Energy costs in the cement industry constitute approximately 30-40 % of the production costs (without capital expenditures). As a result of this, energy management systems have been introduced, as well as various other actions allowing the reduction of both energy consumption and the emissions of pollutants into the air [1, 2]. For a long time using alternative fuels constitutes one of the opportunities for considerably saving primary energy sources in the process of cement clinker production [3-9]. The use of alternative fuels, such as agricultural or wood biomass, biodegradable municipal solid waste, animal waste or paper waste, instead of conventional fossil fuels has the additional advantage of reducing the emission of CO_2 [8, 10]. This is so because the carbon released as a result of the combustion of these alternative fuels is neutral in the global CO_2 balance. Even if wastes from non-renewable sources are used as alternative fuels, the energy recovery from the waste in cement kilns is, as a rule, more efficient than in the case of conventional waste incinerating plants with energy generation systems. To a larger extent, it is also possible here to make use of waste heat – even for the drying of hydrated waste (e.g. sludge) [10-13]. Owing to this, cement kilns constitute a very interesting alternative for the utilisation of various flammable waste types (also including hazardous wastes), particularly since they guarantee a very high degree of destruction for toxic compounds [4-5, 14-16], and unlike in waste incinerating plants no solid residue occurs here, since heavy metals and other solid components become almost entirely incorporated in clinker, which usually does not lose its quality if certain limits regarding the waste fuels being used are kept [17-21].

However, besides the obvious economic benefits, the use of alternative fuels (including waste derived fuels) in cement kilns, in place of conventional fuels, may also involve many problems and hazards. E.g. loading wastes with too high mineral substance (ash) content into the kiln zone, in which the raw material combination is already granulated, may cause clinker inhomogeneity, and thus lower its quality [22]. An excessive content of some heavy metals in alternative fuels may slightly disturb the progress of the clinker formation process and significantly raise their concentration in cement and cement dust, thus increasing the risk of the washing out of these metals in environmental conditions [23-24]. Bringing in too high amounts of chlorine with alternative fuels, and in particular with some wastes, may result in the concentration of chlorides in the external and internal circulation of the rotary kiln, which is particularly unfavourable due to the possibility of a build up of chlorides on the cooler parts of the kiln lining and exchangers heating up raw material, and the corrosion of these elements [25-26]. Due to the combustion of fuels characterised by different calorific values, the following may also change considerably: demand for primary fuel and combustion air, and the amount of generated flue gas [27].

However, the most often analysed problem related to the combustion of waste and fuels obtained from waste in cement kilns is the emission of pollutants into the air, which also involves the necessity to meet additional requirements connected with the waste co-combustion process (including the monitoring of waste gases and the need to comply with stringent emission standards concerning many substances [28]). Often emphasised in previously published works is the fact that the emission of air pollutants taking place during the partial substitution of conventional fuels with alternative ones does not usually differ considerably from the emission occurring while using conventional fuels only [29-30]. Completed analyses of measurement results obtained at different cement plants working around the world prove that no relation is observed e.g. between the emission of polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) and the type and volume of wastes combusted in cement kilns [4, 16, 31, 32]. The following parameters are more important here: the clinker production technology employed, waste feed method, the flue gas cooling rate.

In the case of some substances (SO_2, HCl or heavy metals), a certain change (reduction or increase) in the emission volume was sometimes observed, when the composition of alternative fuels combusted in large volumes considerably differed in the content of sulphur, chlorine or heavy metals, compared to conventional fuels used in a given cement kiln. E.g., in the case of the combustion of PASi type alternative fuel at Małogoszcz Cement Plant (Lafarge Cement Polska S.A. Group), an increase in HCl and CO emissions was observed, while emissions of other substances (SO_2, NO_x, dust and heavy metals) remained almost constant compared to using conventional fuel (coal dust) only [33]. On the other hand, in German and Italian cement plants, a certain increase in the emissions of heavy metals was observed in the case of 50 % substitution of conventional fuel with refuse derived fuel (RDF), in particular with reference to the combustion of petroleum coke only [34], even though emissions of individual substances were always within the limits of the emission standards determined for the waste co-combustion process [28].

It was found that emission standards were also satisfied for all substances (including heavy metals, PCDD/Fs, HCl and HF), in the case when waste tyres and sludge was burnt in cement kilns [35-36]. Research carried out by Cones et al. [35] proves that an increase in the amount of burnt tyres causes higher emissions of some heavy metals (V, Co, Ni, Pb), which in turn does not occur in the case of the growing amount of combusted sludge. On the other hand, the combustion of larger sludge volumes increases NO_x emissions and reduces PCDD/Fs emissions. Slightly higher emissions of PCDD/Fs and volatile organic compounds (VOCs) occured only when waste tyres and petroleum coke (without sludge) were used as fuel. Moreover, these studies allowed the significant variability of the mean SO_2 and HCl concentrations in flue gas for individual measuring series to be obtained, not correlating with the amount of combusted tyres and sludge, also including the volume of sulphur and chlorine brought in with these wastes [35].

The purpose of this work is to try to answer whether and to what extent the use of alternative fuels in a cement kiln affects the level of emissions of selected substances into the air. The analyses were carried out on the basis of data obtained from the system of continuous emission monitoring in one of the Polish cement plants routinely using

alternative fuels, carrying out the processes involving disposal and the recovery of hazardous and other waste types.

2. Examined object characteristics

2.1. Cement kiln

The scope of the research covered a cement kiln operated using dry method with an output reaching 8500 t/day, working with a system of two four-stage cyclone exchangers (low-pressure) and calciner (in form of a vertical two-zone tank).

In a four-stage configuration of cyclones raw material flour is preheated to the temperature of approximately 800-900°C. The cyclones, which constitute individual preheating stages, guarantee a close contact between gas and solid material, and separation between hot gas and charge material. The temperature of waste gasses leaving the preheating system is approximately 300-360°C. These gases are used as a drying medium in raw material mills.

The calciner constitutes an additional combustion chamber fired with coal dust, installed before the rotary kiln and after the cyclone exchanger. The combustion process proceeds here in fuel and burned material mixture, and the combustion temperature reaches ca. 1000-1100°C (maximum 1200°C). Flue gases from the kiln and fuels are delivered to a lower reduction zone. This creates incomplete combustion conditions. 20% of the raw material flour gets into this zone, and the rest of it goes to the upper zone, which is simultaneously supplied with hot air from the clinker cooling bed. This allows the calcination degree of 90-95% for raw material fed into the rotary kiln to be achieved.

The end drying of the charge material and the final calcination, and then the sintering of the charge material to obtain clinker takes place in a rotary kiln sized 5.75×99 m (equipped with a multi-ring coal burner) at temperatures of ca. 900-1450°C (flue gas temperature reaching 1800°C). Clinker cooling takes place in a grate cooling bed provided with three air outlets. Heated air is used as second air for the combustion process in a cement kiln.

This installation is fitted for the combustion of grinded alternative fuels with a high caloric content both in a rotary kiln for clinker firing and in the calciner itself. Alternative fuels are fed into the rotary kiln by the main burner (after mixing with coal), and they are proportioned into the calciner through a hole located under the axis of the burners. It is also possible to supply liquid waste through the main burner in a cement kiln. In both combustion chambers there are suitable thermal conditions for carrying out the waste co-combustion process, and the strong alkaline atmosphere within causes the neutralisation and bonding of acid gases (SO_2, HCl, or HF). However, due to very high temperatures and long flue gas retention time, the cement kiln is more suitable for the combustion of hazardous wastes containing organic compounds, which are hard to decompose.

2.2. Fuels being used

Table 2.2.1 shows the monthly variability of clinker production volume and the worktime of the cement kiln installation and the volume of combusted conventional and alternative fuels during the analysed period (15 months). During that period hard coal was mainly used as the primary fuel (fine coal). Its mean weight in weight contribution compared to all combusted conventional and alternative fuels was almost 85 %. Among conventional fuels, petroleum coke and fly-ash were also occasionally combusted (total share - approximately 1 % by weight). Weight in weight contribution of alternative fuels in individual months ranged from ca. 5 to 25 % (Fig. 2.2.1), on average reaching 14.2 %. On the other hand the energy substitution degree resulting from the use of alternative fuels ranged from ca. 4.5 to 20 %, on average reaching 11.4 %.

Table 2.2.1. Output rate and operating time of the cement kiln and the amount of fuels combusted in the particular months.

No.	Clinker production, t/month	Kiln operating time, hours/month	The amount of combusted fuels, t/month		
			conventional	alternative	total
1	202002.7	578.1	23554.5	3974.9	27529.4
2	216862.4	668.7	25057.1	4745.0	29802.1
3	254573.3	736.3	28845.6	6088.7	34934.3
4	249640.2	707.2	27672.9	6983.3	34656.2
5	245299.9	696.8	26188.9	8666.7	34855.6
6	183122.3	522.1	21733.4	5450.6	27184.0
7	204446.5	598.4	28983.2	4836.7	33819.9
8	45926.8	135.3	5713.4	799.6	6513.0
9	116245.1	361.0	14195.5	2787.2	16982.7
10	218404.4	637.7	24369.0	6717.1	31086.1
11	248235.6	702.8	31506.2	2857.9	34364.1
12	234724.0	671.1	30572.4	2170.3	32742.7
13	233313.5	664.4	30800.8	2646.5	33447.3
14	228282.3	661.9	30378.4	1576.9	31955.3
15	256475.7	727.8	32864.5	2890.0	35754.5

Wastes from mechanical waste processing (e.g. processing by hands, sorting, crushing, granulation) were mainly used as alternative fuels. They are coded as follows:

19 12 10 – combustible wastes (alternative fuel), on average constituting approximately 9.8 % of the weight of all combusted fuels (ca. 69.2 % by weight of alternative fuels);

19 12 11 – other wastes (including mixed substances and objects) from mechanical waste processing, containing hazardous substances, on average constituting approximately 2.8 % of the weight of all combusted fuels (ca. 20.0 % of the weight of alternative fuels).

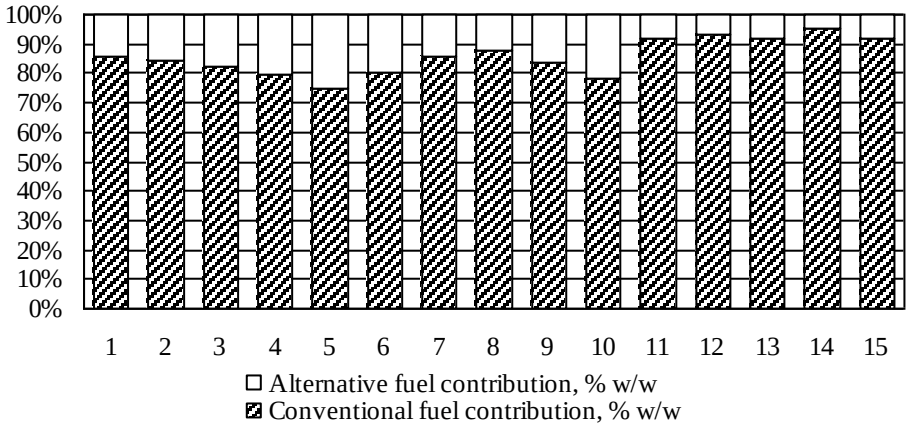

Fig. 2.2.1. Monthly variability of conventional and alternative fuel contribution in the considered period.

Other alternative fuels used during the analysed period were:

- rubber dust (ca. 0.8/5.5 % of the weight of total/alternative combusted fuels),

- sawdust, shavings, cuttings, etc. - wastes from wood processing, belonging to biomass (ca. 0.6/4.3 % of the weight of combusted total/alternative fuels),

- plastics and rubber, plastic packaging, solid combustible wastes containing hazardous substances from physicochemical waste processing, used oils, stabilised liquid municipal wastes and tobacco wastes, with the total weight in weight concentration of approximately 0.2 % compared to all the combusted wastes and ca. 1 % compared to alternative fuels only.

Many parameters should be examined for the alternative fuels delivered for combustion, among other things including chlorine and sulphur content. Figures 2.2.2 and 2.2.3 show the monthly variability of the total chlorine and sulphur load brought in with conventional and alternative fuels during the period in question. The figures demonstrate that chlorine is delivered into this process with alternative fuels to a much larger extent than with conventional fuels (despite the relatively low share of combusted alternative fuels). It is the other way round in the case of sulphur – the role of alternative fuels is decidedly smaller than the role of conventional fuels. It should be added that raw material flour is also an important source of chlorine and sulphur in this process.

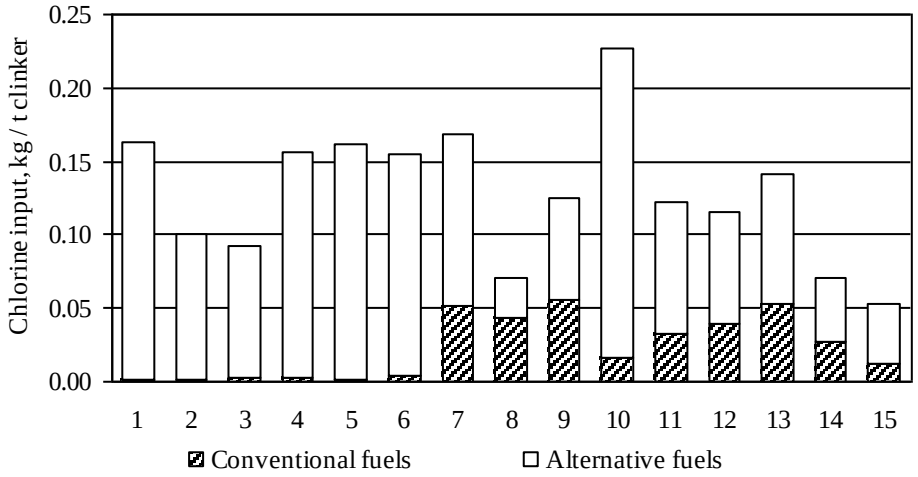

Fig. 2.2.2. Monthly variability of the total chlorine load brought in the process with conventional and alternative fuels.

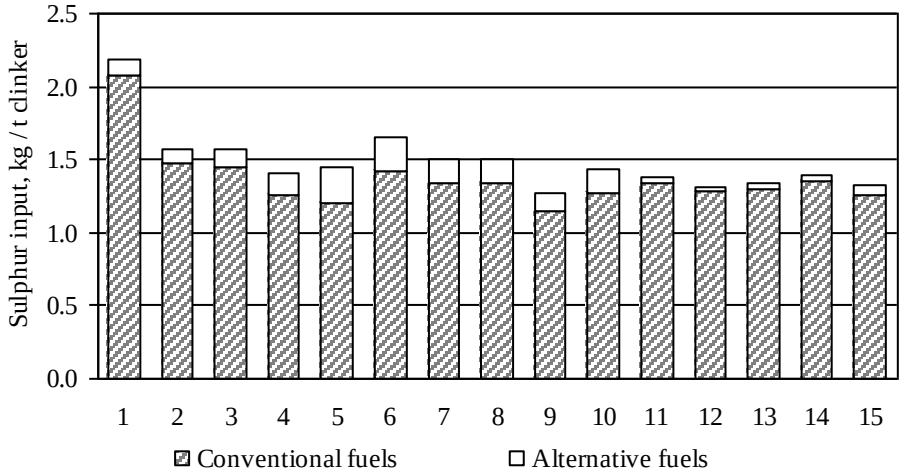

Fig. 2.2.3. Monthly variability of the total sulphur load brought in the process with conventional and alternative fuels.

2.3. Analysed substances

The analysis covered the monthly emissions into the air of the following substances: CO_2, CO, NO_x (NO and NO_2 as NO_2), SO_2, HCl, TOC and total dust. These data originated from the system of continuous emission monitoring, based on a Codel G-CEM 4000 type multi-gas analyser (measurements of CO_2, CO, NO_x, SO_2 and HCl), a Thermo-FID type total organic carbon analyser (using the continuous flame ionisation detection method), a Codel D-CEM 2000 type optical dust meter, and a Codel V-CEM 5000 type flue gas flowmeter. Moreover, the results of the chlorine and sulphur content determinations in the combusted fuels were used in the analyses.

3. Results and discussion

Table 3.1 shows Pearson's linear correlation coefficients determined for individual monthly emissions and selected parameters referring to the same period: clinker production volume, cement kiln operating time, amounts of combusted conventional fuels (CF), alternative fuels (AF) and total fuels (TF), and chlorine and sulphur mass streams brought into the process with individual fuels. Table 3.2 contains the analogical coefficients for the selected quantities referring to 1 metric ton of manufactured clinker.

Table 3.1. Pearson's linear correlation coefficients between air emissions and selected parameters with reference to the month period

Parameter	CO_2	CO	NO_2	SO_2	HCl	Dust	TOC
Clinker production	0,959	0,802	0,872	-0,156	0,393	0,626	0,445
Kiln operating time	0,965	0,826	0,893	-0,177	0,410	0,618	0,457
Amount of CF	0,939	0,747	0,842	-0,085	0,451	0,580	0,406
Amount of AF	0,422	0,597	0,400	0,002	0,332	0,551	0,185
Amount of TF	0,970	0,845	0,875	-0,076	0,502	0,680	0,420
Weight ratio AF/TF	-0,033	0,210	0,007	0,078	0,135	0,258	-0,013
Cl input with CF	0,129	0,067	0,174	0,080	0,338	-0,179	-0,113
Cl input with AF	0,509	0,579	0,493	0,213	0,499	0,604	0,431
Cl input with TF	0,579	0,631	0,578	0,252	0,642	0,571	0,413
S input with CF	0,859	0,704	0,693	0,145	0,474	0,673	0,776
S input with AF	0,283	0,475	0,204	0,168	0,325	0,588	0,114
S input with TF	0,867	0,753	0,695	0,168	0,508	0,744	0,758

The data presented indicates a strong linear dependency between the total monthly CO_2, CO and NO_2 emissions, and the clinker production volume and cement kiln worktime in a given month. A considerably smaller impact of these parameters occurred as regards the monthly dust emissions, and in particular TOC and HCl emissions, whereas a dependency of this type was not observed in the case of SO_2 emissions. Among other things, the monthly emissions of CO_2, CO and NO_2 also seem to depend on the volume of combusted conventional fuels and total fuels, but this is an indirect effect of the production volume and

cement kiln operating time impact. This is confirmed in Table 3.2, which contains the correlated monthly data referring to 1 metric ton of manufactured clinker, where in the case of most substances, definitely lower values of Pearson's linear correlation coefficients have already been obtained.

Table 3.2. Pearson's linear correlation coefficients between emission factors and selected parameters with reference to 1 metric ton of clinker

Parameter	CO_2	CO	NO_2	SO_2	HCl	Dust	TOC
Amount of CF	0,202	0,036	0,019	0,183	0,207	-0,002	-0,077
Amount of AF	0,133	0,419	0,196	0,116	0,131	0,363	-0,003
Amount of TF	0,440	0,560	0,265	0,394	0,444	0,440	-0,110
Cl input with CF	0,478	0,260	0,265	0,194	0,527	-0,123	-0,108
Cl input with AF	-0,168	0,094	0,139	0,080	-0,182	0,250	0,204
Cl input with TF	0,029	0,227	0,281	0,181	0,036	0,228	0,183
S input with CF	-0,202	0,198	-0,255	-0,011	-0,206	0,309	0,494
S input with AF	-0,016	-0,038	-0,312	0,445	0,044	0,375	0,851
S input with TF	0,234	0,434	0,009	0,377	0,303	0,611	0,023

Within the range of its variability under consideration (from 5 to 25 % of weight in weight contribution compared to the total volume of combusted fuels), the amount of combusted alternative fuels has no significant impact on the level of the emissions of examined substances on a monthly scale, in particular referring to 1 metric ton of manufactured clinker (Table 3.2). In particular, this applies to HCl emission, as in theory alternative fuels may constitute a substantial source of this substance. However, an increase in the volume of combusted alternative fuels and the amount of chlorine brought in with these fuels does not result in a higher HCl emission factor, referring to 1 metric ton of clinker (Fig. 3.1-3.2).

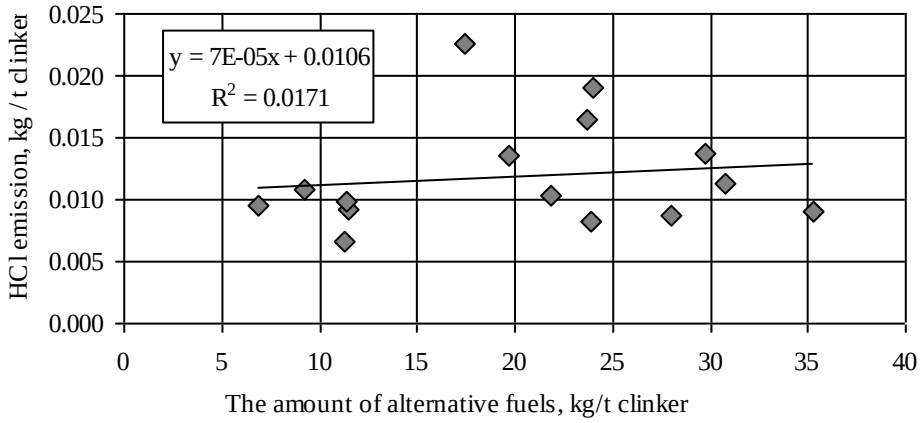

Fig. 3.1. HCl emission in comparison to the amount of combusted alternative fuels.

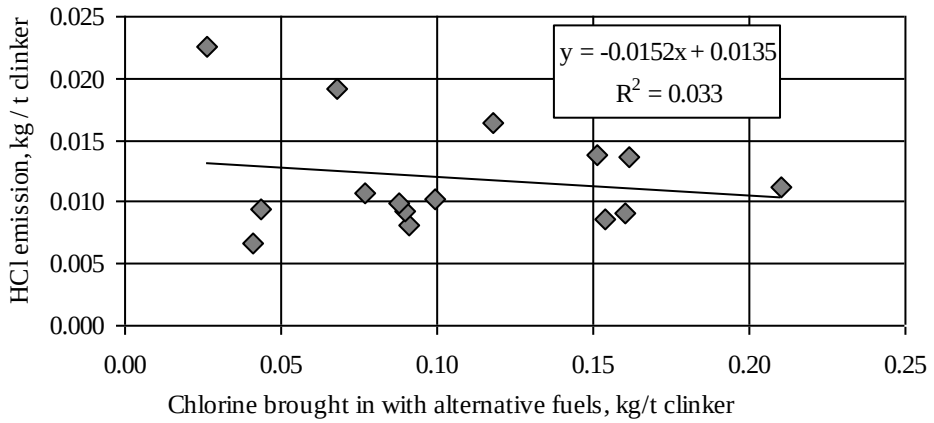

Fig. 3.2. HCl emission in comparison to the total chlorine load brought in the process with alternative fuels.

Monthly SO_2 emission also does not depend on the volume of total combusted fuels and the sulphur load brought in with fuels, including alternative fuels as well (Fig. 3.3-3.4). Despite that trend lines have been added in Figures 3.1-3.4, the low correlation coefficients obtained confirm that there is no linear dependency between the examined parameters. This agrees with test results obtained by, among others Cones at al. [35], who have not proved any impact of the amount of combusted wastes and their chlorine and sulphur content on the level of HCl and SO_2 emissions from a cement kiln.

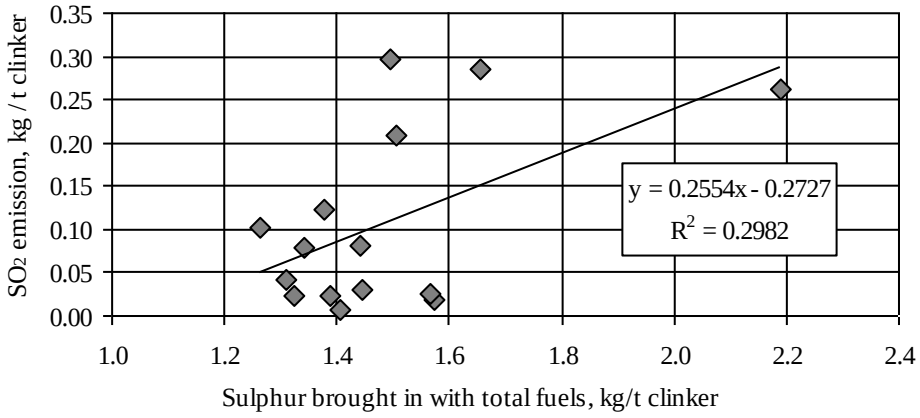

Fig. 3.3. SO_2 emission in comparison to the total sulphur load brought in the process with total fuels.

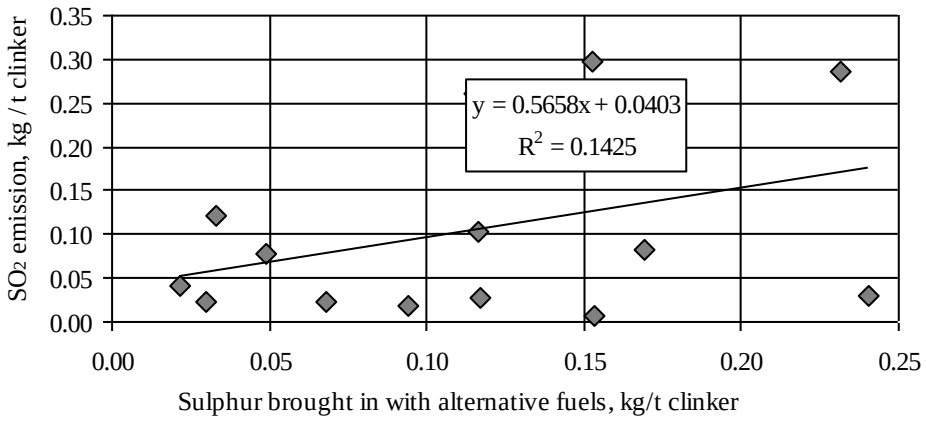

Fig. 3.4. SO_2 emission in comparison to the total sulphur load brought in the process with alternative fuels.

3. Summary

Co-combustion of wastes in a cement kiln, profitable from an economic point of view (saving considerable amounts of fossil fuels), does not cause any substantial differences in the volume of emitted basic air pollutants.

Analyses carried out for a cement kiln operated using the dry method prove that the replacement of approximately 10-20 % of coal (combusted in the main burner and calciner burners) with alternative fuels, bringing in chlorine or sulphur amounting to up to 0.25 kg/t clinker, does not cause any noticeable increase in HCl and SO_2 emissions into the air.

Wastes and fuels obtained from waste may deliver a considerable volume of chlorine compared to conventional fuel. An especially important fact is that there are no dependencies between the amount of chlorine brought into the production process with alternative fuels of this type and HCl emission. Therefore, not the problem of emission into the air but the production technology considerations should constitute the key factor limiting the amount of chlorine being brought in. This is due to the possibility for easily satisfying the HCl emission standards specified for the waste co-combustion installation in a cement kiln. Assuming that the chlorine brought in by fuels is the only source of HCl emission from this process, the degree of its retention in the examined kiln reached approximately 70-95 %.

Moreover, the completed analyses prove that the mean level of fuel sulphur absorption in raw material flour considerably exceeds 90 %, in some months reaching even up to ca. 99 %, which is a higher value than specified in the BAT reference documents for the cement industry [31]. Absorption of gaseous substances (including SO_2) in a cement kiln

environment leads to their bonding in the solid phase (clinker and cement dust). As a result of that, the efficiency of this process is an important factor affecting the level of emissions of these substances in the gaseous phase. On the other hand, the fact has been confirmed that volatile sulphur content in raw materials is decisive for SO_2 emission, not its content in fuels. Probably, a high variability as regards the amount of sulphur delivered in raw materials is also the reason for the lack of correlation between SO_2 emission and clinker production volume in a given month.

The emissions of the remaining examined substances (CO_2, CO, NO_x, TOC and dust) is also not significantly dependent on the amount of combusted alternative fuels. Relatively, the highest Pearson's coefficient of linear correlation (approximately 0.4) between the amount of combusted alternative fuels and the emission volume referring to 1 metric ton of clinker was obtained for CO and dust. However, these correlations are very faint, and they do not improve much when considering the total amount of combusted fuels. On the other hand, taking into account the data related to the period of a month, a strong linear correlation between the production volume and the total emission into the air occurs only in the case of CO_2, CO and NO_x.

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