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THE INFLUENCE OF SHALE ROCK FRACTURING EQUIPMENT OPERATION ON ATMOSPHERIC AIR QUALITY

WPŁYW PRACY URZĄDZEŃ DO SZCZELINOWANIA SKAŁ ŁUPKOWYCH NA JAKOŚĆ POWIETRZA ATMOSFERYCZNEGO W TRAKCIE WYKONYWANIA ZABIEGU

The hydraulic fracturing jobs performed on shale rocks are connected with atmospheric emissions of dusts and exhaust gases from high-power motors supplying pump aggregates used for fracturing operations and from other technological devices. The total power of motors driving technological systems depends on the specific character of deposit and well and may range between a dozen to tens of thousands kW. An exemplary set of technological systems used for frac jobs is presented in figure 1. The following substances are emitted to the atmosphere during engine operation, e.g. nitrogen oxides (NO_x), sulfur dio-xide (SO₂), carbon oxide (CO), dust PM10, ammonia, benzo(a)pyrene (B(a)P), benzene, toluene, xylene, formaldehyde, acetaldehyde, acrolein. As a consequence admissible concentrations of these substances in air can be exceeded.

The influence of dust and gaseous emissions accompanying shale rock fracturing jobs is addressed in this paper. Model analyses were performed. An exemplary model of a process used for simulating propagation of atmospheric emissions in a specified calculation area $(1,150 \text{ m} \times 1,150 \text{ m})$ were based on the analysis of hydraulic fracturing jobs performed in wells in Poland and abroad. For making calculations more actual, the model was located in the Gdańsk area and was ascribed its typical meteorological and orographic parameters. In the center of this area a rig site $150 \text{ m} \times 150 \text{ m}$ was distinguished. The emission field was generated by 12 high-power engines supplying pump aggregates, 1680 kW each. The time of work of particular engines was established for 52 hrs (13 frac jobs, each lasting 4 hrs). It was assumed that all engines will operate simultaneously and using 100% of their power.

Attention was paid to the correct modelling of the real emission field. Technical parameters of motors and the applied fuels were characterized. Emission indices were worked out by, e.g. U.S. Environmental Protection Agency or European Environment Agency.

The calculations of air pollutions from analyzed motors were performed with a mathematical modelling method using Gaussian plum.

The results of calculations could be used for evaluating spatial distribution of maximum 1 hour concentrations (S_1), incidence of exceeding admissible 1 hour concentration values ($P(D_1)$), percentile 99.8 or 99.726 from 1 hour concentrations and average concentrations (S_a) for selected most important for the air quality contaminants, i.e. NO_x (as NO_2), SO_2 , CO, PM10, benzo(a)pyrene, benzene, toluene,

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xylene, formaldehyde, acetaldehyde and acrolein. The results of calculated air concentrations of selected substances on the rig border are listed in table 9, whereas spatial distributions of NO_x and PM10 concentrations in figures 3 to 8. The analysis of the obtained results did not reveal cases of exceeding Polish emission standards. However, nitrogen oxide (NO_x) or dust PM10 can be expected to exceed these values, e.g. in a situation when the total power installed in motors driving technological systems in the course of hydraulic fracking will be higher than assumed in the analyses.

The results of calculations show to a significant impact of nitrogen oxides (NO_x) and dust PM10 emissions on air quality. The risk that emission standards are exceeded beyond the rig area is conditioned both by technological factors (total power of operating motors, parameters of combusted fuel, reduced emission technologies applied to engines, duration of frac jobs, etc.) and a number of external factors, e.g. meteorological and orographic factors or high level of emitted substances in air within the rig area.

Keywords: gas prospecting, shale gas, atmospheric emissions, environmental impact

Proces hydraulicznego szczelinowania skał łupkowych wiąże się z emisją do powietrza zanieczyszczeń pyłowo-gazowych z silników wysokoprężnych dużej mocy napędzających agregaty pompowe do szczelinowania skał oraz inne urządzenia technologiczne. Łączna moc silników napędzających urządzenia technologiczne uzależniona jest od specyfiki złoża oraz specyfiki odwiertu i waha się od kilkunastu do kilkudziesięciu tysięcy kW. Przykładowy zestaw urządzeń technologicznych wykorzystywanych w procesie szczelinowania zamieszczono na rysunku 1. Podczas pracy silników do atmosfery emitowane są między innymi następujące substancje: tlenki azotu (NO_x), ditlenek siarki (SO₂), tlenek węgla (CO), pył PM10, amoniak, benzo(a)piren (B(a)P), benzen, toluen, ksylen, formaldehyd, acetaldehyd, akroleina. Skutkiem emisji mogą być przekroczenia dopuszczalnych, określonych prawem wartości stężeń substancji w powietrzu.

W artykule skoncentrowano się na ocenie wpływu na jakość powietrza atmosferycznego emisji substancji pyłowo-gazowych towarzyszących procesowi szczelinowania skał łupkowych. Badania miały charakter modelowy. W oparciu o analizę przebiegu szeregu procesów szczelinowania hydraulicznego wykonywanych na wiertniach zlokalizowanych w Polsce i za granicą zbudowano przykładowy model procesu, który posłużył do wykonania symulacji propagacji zanieczyszczeń emitowanych substancji w powietrzu w zadeklarowanym obszarze obliczeniowym stanowiącym teren o wymiarach 1150 m × 1150 m. Dla urealnienia wyników obliczeń umiejscowiono model badanego obszaru w okolicach Gdańska i przypisano mu charakterystyczne dla tego rejonu parametry meteorologiczne i orograficzne. W środku analizowanego obszaru zlokalizowano teren wiertni o wymiarach 150 m x 150 m. Pole emisji kształtowane było przez 12 silników wysokoprężnych napędzających agregaty pompowe, każdy o mocy 1680 kW. Czas pracy pojedynczego silnika ustalono na 52 godziny w roku (13 zabiegów szczelinowania, każdy trwający 4 godziny). W obliczeniach założono, że podczas trwania szczelinowania wszystkie silniki będą pracowały równocześnie i będą obciążone w 100 %.

W artykule dużo uwagi poświęcono poprawnemu zamodelowaniu rzeczywistego pola emisji. Scharakteryzowano parametry techniczne silników oraz zużywanych przez nie paliw. Zaadoptowano na potrzeby obliczeń modelowych wskaźniki emisji opracowane między innymi przez Amerykańską Agencję ds. Środowiska, czy Europejską Agencję ds. Środowiska.

Obliczenia propagacji w powietrzu zanieczyszczeń pochodzących z analizowanych silników wykonano metodą modelowania matematycznego wykorzystując w tym celu model smugi Gaussa.

Wyniki obliczeń pozwoliły na ocenę rozkładu przestrzennego stężeń maksymalnych 1-godzinnych (S₁), częstości przekroczeń wartości dopuszczalnej stężeń 1-godzinnych (P(D₁)), percentyla 99,8 lub 99,726 ze stężeń 1-godzinnych oraz stężeń średniorocznych (S_a) dla wybranych, najbardziej istotnych z punktu widzenia wpływu na jakość powietrza zanieczyszczeń tj.: NO_x (jako NO₂), SO₂, CO, PM10, benzo(a) pirenu, benzenu, toluenu, ksylenu, formaldehydu, aldehydu octowego oraz akroleiny. Wyniki obliczeń stężeń wybranych substancji w powietrzu na granicy wiertni zestawiono w tabeli 9, natomiast rozkłady przestrzenne stężeń NO_x oraz PM10 przedstawiono na rysunkach 3-8. Analiza wyników obliczeń nie wykazała wprawdzie dla analizowanego przypadku występowania przekroczeń obowiązujących w Polsce standardów imisyjnych, ale w przypadku takich zanieczyszczeń jak tlenki azotu (NO_x) czy pył PM10 istnieje realne zagrożenie wystąpienia takich przekroczeń, między innymi w sytuacji, gdy łączna moc

zainstalowana w silnikach napędzających urządzenia technologiczne w czasie procesu szczelinowania będzie wyraźnie większa niż przyjęta w opisanych badaniach.

Wnioski z przeprowadzonych obliczeń wskazują na istotny wpływ na jakość powietrza emisji tlenków azotu (NO_x) i pyłu PM10. Ryzyko wystąpienia przekroczeń obowiązujących standardów imisyjnych poza granicą wiertni jest uwarunkowane zarówno czynnikami technologicznymi (łączna moc pracujących silników, parametry spalanego paliwa, zastosowane w silnikach techniki ograniczenia emisji, czas trwania szczelinowania itp.) jak i szeregiem czynników zewnętrznych takich jak czynniki meteorologiczne, orograficzne czy wysoki poziom tła emitowanych substancji w powietrzu w lokalizacji wiertni.

Słowa kluczowe: poszukiwania gazu, gaz z łupków, emisja do powietrza, oddziaływanie na środowisko

1. Introduction

Shale gas deposits are developed with multilateral wells located at a depth of a few thousand meters. For obtaining gas inflow a few to a dozen of hydraulic fracturing jobs are performed in the horizontal sections, i.e. prepared fracturing fluid and sand are injected at high pressure thanks to which the fracture can be supported. Specialist high-pressure equipment of total power ranging between a dozen to tens thousand kW is used for this type of procedure. In the course of these jobs the powering engines emit contaminations and noise (Macuda, 2010).

After fracturing is complete, the frac fluid is removed from the well and reservoir tests are performed to assess the natural gas resources. Gas recovered to the surface is most frequently combusted in flare stacks or separated from post-frac flowback fluid and stored (Nagy & Siemek, 2011).

The risk analysis of drilling works and frac jobs presented in the report (AEA Technology plc, 2012) reveals that the hydraulic fracturing operation has the biggest influence on the quality of air in the neighborhood of the rig area in the course of developing shale gas deposits. Great amounts of exhaust gases are emitted from high-power engines feeding particular elements responsible for the injection of fracturing fluid to the well. In this process the following substances may be emitted to the atmosphere, e.g. nitrogen dioxide (NO₂), sulphur dioxide (SO₂), carbon oxide (CO), dust PM10, ammonia, benzo(a)pyrene (B(a)P), benzene, toluene, xylene, formal-dehyde, acetaldehyde, acrolein.

2. Characteristic of sources of atmospheric emissions from shale rock hydraulic fracturing processes

The process of hydraulic fracturing of shale rocks is connected with emissions of dusts and gases from high-pressure high-power engines supplying technological systems. These are most frequently diesel engines (more rarely CNG, LPG) which are used for powering, e.g. pump aggregate, space aggregate, blender, hydration unit, chemical dosing unit, nitrogen pump, postreaction fluid pump, energy generator, fire pump, etc. An exemplary frac system of 17,600 KM is presented in Fig. 1.

The highest emissions were observed for pump aggregates as engines are usually used for powering this type of equipment and their power exceeds that total power of all engines powering the remaining elements involved in the hydraulic fracturing job.





Fig. 1. Exemplary schematic of a hydraulic fracturing system of 17,600 KM (ZRG Krosno, 2013)

Engines used for pump aggregates are selected mainly in view of their power, which should be sufficient to support the hydraulic fracturing operation at the pump outlet and provide sufficient efficiency of the frac fluid injection. The operator should also account for the emission class of engines used in pump aggregates as being closely connected with the amount of dust and gases emitted to the atmosphere and so the impact of hydraulic fracturing job on the quality of air beyond the rig area.

An exemplary pump aggregate is equipped with a high-pressure four-cycle and twelve cylinder engine of 1,678 kW (2250 KM), maximum rotational torque 9,370 Nm and 1,900 rpm. Diesel oil is most frequently used here. Pump aggregates are usually mounted on specially prepared autoplatforms giving them full mobility. Exhaust gases are discharged to the atmosphere through a pipe of diameter 0.2 m to 0.4 m, depending on the aggregate type, and usually disposed at a height of 4 m a.s. Depending of the year of production and engine version they may be equipped with emission reduction systems. This question is regulated by homologation acts for engines introduced to the market, though it must be remembered that these regulations may have regional character to a great extent (e.g. UE, U.S.A., Japan, etc.) For instance, industrial rigs and compressors equipped with self-ignition systems of net power above 19 kW but less than 560 kW, operating at a constant rotational speed are classified as offroad mobile machines, according to § 3 section 2 pt. 1 of Minister of Economy and Labor Regulation of 19 August 2005 *about detailed research on combustion engines in view of limiting generated gaseous and*

Constant development of engines in view of their efficiency and restrictive emission standards found its expression in the EU Parliament Directive 2012/46/UE presenting guidelines of homologating engines in view of emission limit values: stages IIIA, IIIB and IV (DKE, 2012). The transposition of this directive to the Polish law should be effective by 21 December 2013. Similar regulations are valid in the U.S.A. where engines supplying offroad machines have emission standards (Tier 1-4) established by U.S. EPA. Emission limits for engines of power exceeding 560 kW, and so the ones most frequently used for shale gas prospecting, are presented in table 1.

TABLE 1

	Emission limits in g/kWh				
Contaminant	Tier 1	Tier 2	Tier 4 Interim	Tier 4	
	2000-2005	2006-2010	2011-2014	+2015	
СО	11.4	3.5	3.5	3.5	
HC	1.3	-	0.4	0.19	
NMHC+NO _x	-	6.4	—	-	
NO _x	9.2	-	0.67	0.67	
PM	0.54	0.2	0.1	0.04	

Emission limits of high-pressure engines > 560 kW used, e.g. in pump aggregates (U.S. EPA, 1996)

3. Methodics of assessing air emissions by pump aggregates

When evaluating atmospheric emissions from high-pressure engines supplying pump aggregates the following parameters should be taken into account: type and capacity of engine, type of fuel, work characteristic (fixed-speed, variable speed), fuel consumption, cleaning or emission control systems, actual emission standard. These data are usually given in technical specification of the engine or its homologation certificate. Producers tend to give results of their own analyses indicating maximum emissions of only basic contaminants, e.g. NO_x , SO_2 , CO, HC, PM10 and PM2.5. The emission indices to be used when determining the quality and quantity of air pollutions emitted by engines are given by the U.S. Environmental Protection Agency in AP-42 and by European Environmental Agency (EEA) in a database worked out within the project EMEP/ CORINAIR (U.S. EPA, 1996, 2013; EIG, 2006). The emission indices coming from AP-42 (U.S. EPA, 2006), referring to stationary engines of over 560 kW are presented in table 2.

		, ,		,	
		Fuel – diesel oil			
		Emission index	Emission index referred	Quality of	
Contaminant	Emission control	referred to power of	to energy introduced to	emission index	
		engine at output	engine with fuel		
		[g/kWh]	[ng/J]	~	
	no	14.592	1,376	В	
NO _x	Delayed mixture	7 904	817	В	
	injection	7.501	017		
CO	no	3.344	365.5	С	
SO _x ^{b)}	no	4.918 * S	434.300 · S	В	
CO ₂ ^{c)}	no	705.280	70,950	В	
PM	Delayed mixture	0.426	13.0	Р	
	injection	0.420	45.0	Б	
TOC (as CH ₄)	no	0.429	38.7	С	
Methane	no	d)	d)	Е	
NMLZO	no	d)	d)	Е	

Emission indices for stationary diesel engines of over 560 kW (U.S. EPA, 2006)

 a) Quality of emission index evaluation according to scale: A – excellent, B – above average, C – average, D – below average, E – unsatisfactory, F – no (no documented analyses made)

^{b)} Index calculated for maximum sulphur content in diesel oil S [%] = 0.001%

c) Assumed 100% conversion of C making up fuel to CO₂

^{d)} Based on analyses made on only one engine it was stated that TOC measured in exhaust gases is based on methane in 9% and nonmethane volatile organic compounds (NVOC) in 91%

The analysis of data in table 2 reveals that SO_2 emission depends on the percentage of sulphur (S) in combusted fuel. According to PN-EN 590 the sulphur content in diesel oil in Poland cannot exceed 0.001 wt% (PKN, 2011). The limit percent participation of sulphur in light and heavy heating oil as well as oil used by inland ships is specified by a Regulation of the Minister of Economy of 4 January 2007 about qualitative requirements for sulphur content in oils, types of installations and conditions in which heavy heating oils will be used (Official Journal no. 4. item 30. of 4 January 2007) (RMG, 2007). According to the regulation, sulphur content per mass of oil cannot exceed:

- 1) 0.1% in light heating oil, labelled as CN 2710 19 45 and 2710 19 49 since 1 January 2008,
- 1% in heavy heating oil, labelled as CN 2710 19 51, 2710 19 61, 2710 19 63, 2710 19 65 and 2710 19 69,
- 3) 0.1% in oils for inland ships since 1 January 2010.

In Poland heavy oils can be combusted (as quoted in pt. 2) without meeting requirements related to the spoken of regulation except for combustion of fuels for energy generation purposes which has to be given permit for discharging gases and dusts to the atmosphere, or an integrated permit. Multipurpose engines used for shale gas prospecting are supplied by commonly available diesel oil in which the admissible sulphur mass does not exceed 0.001%.

High-pressure engines generate aerosols of various chemical composition to the atmosphere. The qualitative and quantitative indices of atmospheric aerosol emissions from a stationary high-pressure engine of over 560 kW, which has not been equipped with emission control systems, are presented in table 3.

Contamination	Emission index in reference to energy introduced to engine with fuel [ng/J]
Range of determined solid fractions ^a):	
< 1 µm	20.554
< 3 µm	20.597
< 10 µm	21.328
Total amount of determined solid particles	26.66
Condensed liquid particles	3.311
PM10 ^{b)}	24.639
Total dust ^{c)}	29.971

Aerosol emissions from stationary high-pressure engine of over 560 kW, not equipped with emission control systems (U.S. EPA, 2006)

^{a)} Fraction range expressed as aerodynamic particle diameters

^{b)} Sum of total content of determined dust particles of aerodynamic diameter <10 µm and condensed liquid particles in the same fraction range

c) Sum of total content of determined dust particles and condensed liquid particles

High-pressure and high-power engine emits volatile organic compounds (VOC) and polycyclic aromatic hydrocarbons (PAH). Emission indices for selected representatives of both these groups of compounds are presented in tables 4 and 5, respectively (U.S. EPA, 2006).

TABLE 4

Emission indices of selected organic compounds from stationary high-pressure engine of over 560 kW, not equipped with emission control systems

Contaminant	Emission index referred to energy introduced with fuel to engine [ng/J]		
Benzene	0.33368		
Toluene	0.12083		
Xylene	0.08299		
Propylene	1.19970		
Formaldehyde	0.03393		
Aceltaldehyde	0.01084		
Acrolein	0.00339		

TABLE 5

Emission indices of selected polycyclic aromatic hydrocarbons from stationary high-pressure engine of over 560 kW, not equipped with emission control systems

Contaminant	Emission index referred to energy introduced with fuel to engine [ng/J]		
1	2		
Naphtalene	5.59E-02		
Acenaphtalene	3.97E-03		
Acenaphten	2.01E-03		
Fluorene	5.50E-03		

TABLE 3

1	2			
Phenanthrene	1.75E-02			
Anthracene	5.29E-04			
Fluoranthene	1.73E-03			
Pyrene	1.60E-03			
Benzo(a)anthracene	2.67E-04			
Chrysene	6.58E-04			
Benzo(b)fluoranthene	4.77E-04			
Benzo(k)fluoranthene	9.37E-05			
Benzo(a)pyrene	1.11E-04			
(Indeno(1,2,3-cd)pyrene	1.78E-04			
Dibenz(a,h)anthracene	1.49E-04			
Benzo(g,h,l)perylene	2.39E-04			
Total WWA	9.12E-02			

High-pressure and high-power engines can be equipped with various control systems reducing dust-gaseous emissions. The most frequently applied techniques and control systems for reducing atmospheric emissions from high-pressure engines of over 560 kW are presented in table 6.

TABLE 6

	Deremotor	Fuel – diesel oil		
Emission control	connected with control method	Degree of NO _x emission reduction [%]	Changes in specific fuel use ^{a)} [%]	
	10%	No data	No data	
Lower engine load	20%	< 20	4	
	25%	5-23	1-5	
	2°	< 20	4	
Delay fuel injection stage	4°	< 40	4	
	8°	28-45	2-8	
Introduce additional cir	3%	No data	No data	
	±10%	7-8	3	
Water injection	50%	25-35	2-4	
Selective catalytic reduction method (SCR)		80-95	0	

Degree to which NO_x emission and fuel consumption can be reduced with the use of selected emission control methods (U.S. EPA, 2006)

^{a)} Specific use of fuel means quantity of fuel used in a unit of time at a given power of engine

When the engine is not equipped with any emission control system, then emission should be evaluated with the use of indices listed in tables 2 to 5. The amount of emission can be calculated on the basis of information about the power of engine in kW or quantity and type of combusted fuel per hour. Otherwise, the emission should be corrected by the control degree typical of a given control system/technology as specified in table 6. Exhaust emission generated by engines which meet emission limits should be calculated with the use of proper emission limit values as listed in table 1.

4. Modeling the effect of dust-gaseous emissions from hydraulic fracturing jobs in shale rocks on air quality

The influence of dust-gaseous emissions from hydraulic fracturing jobs on air quality was evaluated on the basis of the following assumptions. The study area of 1,150 m × 1,150 m was localized in Gdańsk region. The rig site 150 m × 150 m was in the center of it. The entire area was covered with a network of receivers (50 m calculation step). The assumed emission field stayed under the influence of 12 pump aggregates, supplied by high-pressure engines of 1,680 kW each, supplied by diesel oil of calorific value 42,500 kJ/kg and sulphur content 0.001%. Fuel consumption by one engine was assumed as of 300 dm³/h as defined on the basis of its technical specification. The amount of dust-gaseous emissions was calculated for each engine on the basis of emission indices recommended for this type of engine in AP-42 (U.S. EPA, 2006). The aim of the analysis in view, a lower favourable emission variant was assumed, i.e. all engines used in the hydraulic fracturing process neither satisfy the emission requirements and limits, nor have emission control systems installed. It was additionally assumed for dust emitted from engines that dust of diameter under 10 μ m (PM10) will constitute 100% of total dust. The emission values calculated for one engine are presented in tables 7 and 8.

TABLE 7

~			
Contaminant	Emission* [kg/h]		
NO _x	14.5615		
СО	3.8679		
SO ₂	0.0046		
PM10	0.4550		
Benzene	3.53E-03		
Toluene	1.28E-03		
Xylene	8.78E-04		
Formaldehyde	3.59E-04		
Acetaldehyde	1.15E-04		
Acrolein	3.59E-05		

Selected emissions from high-pressure engine of 1,680 kW supplying pump aggregate

* Emission determined on the basis of chemical energy introduced with fuel to the engine. Emission was calculated for fuel of calorific value 42,500 kJ/dm³ and sulphur content 0.001%.

TABLE 8

PAH emissions from high-pressure engine of 1,680 kW supplying pump aggregate

Contaminant	Emission WWA* [kg/h]		
1	2		
Naphtalene	2.54E-01		
Acenaphtalene	1.81E-02		
Acenaphtene	9.16E-03		
Fluorene	2.50E-02		
Phenanthren	7.98E-02		
Anthracene	2.41E-03		

1	2
Fluorantene	7.89E-03
Pyrene	7.26E-03
Benzo(a)anthracene	1.22E-03
Chrysene	2.99E-03
Benzo(b)fluorantene	2.17E-03
Benzo(k)fluorantene	4.27E-04
Benzo(a)pyrene	5.03E-04
(Indeno(1,2,3-cd)pyrene	8.10E-04
Dibenz(a,h)anthracene	6.77E-04
Benzo(g,h,l)perylene	1.09E-03
Total WWA	4.15E-01

* Emission determined on the basis of chemical energy introduced with fuel to the engine. Emission was calculated for fuel of calorific value 42,500 kJ/dm³ and sulphur content 0.001 %.

For the correct description of emission variability in a function of time it was assumed that the total time of operation of each engine during the frac job in a given site will be 52 hrs, which results from the fact that 13 hydraulic fracturing jobs are performed in one horizontal wells, when each job lasts 4 hours. It was also assumed that all engines would operate simultaneously during fracturing procedure and would be 100% changed. The exhaust gases emitted by engines are discharged to the atmosphere with exhaust pipes (vertical, open emitters) the outlets of which are disposed 4 m above the surface and the emitter diameter is 0.35 m. The assumed average temperature of exhaust gases equals to 773 K, whereas exhaust gases leave the outlet at a rate of 46 m/s. No other emission sources were accounted for in calculations, e.g. engines powering blenders, hydration unit, etc. due to the fact that power of these engines is many times lower as compared to the ones used for pump aggregates.

Dispersion of contaminations from analyzed sources was calculated with a mathematical model based of Gaussian plum. The model is based on an analytical solution of differential equation of exhaust gases dispersion in a moving gaseous medium:

$$\frac{dS}{dt} = \frac{d}{dt} \left(\frac{s_y^2}{2} \right) \frac{d^2 S}{dy^2} + \frac{d}{dt} \left(\frac{s_z^2}{2} \right) \frac{d^2 S}{dz^2}$$
(1)

where:

- S concentration of contaminants at reception point,
- t time in which contaminants move from the emission place to the reception point,
- σ_y Pasquill dispersion coefficient along y axis in the Cartesian coordinates system x, y, z for point of time t,
- σ_z Pasquill dispersion coefficient along z axis in the Cartesian coordinates system x, y, z for point of time t.

The solution of equation (1) for 1 hour emission air concentrations is obtained for the following boundary conditions:

1) source of emission of mass intensity E is disposed at a point with coordinates: t = 0,

y = 0 and z = 0 (here the point emitter at geometrical height *h* is introduced),

- 2) on the surface the contaminations (gaseous substance) are not absorbed which means that for z = 0 the contamination stream equals to zero (quantity of contaminations reaching the surface equals to the amount of contaminations which 'bounced' from the surface),
- 3) concentration of contaminations at an infinitely distant place from its source $(t, y, z \rightarrow \infty)$ equals to zero.

The solution of equation (1) at the assumed boundary conditions takes the following form:

$$S_{xyz} = \frac{E}{2 \pi \overline{u} \sigma_y \sigma_z} \exp\left(-\frac{y^2}{2 \sigma_y^2}\right) \left\{ \exp\left(-\frac{(z-H)^2}{2 \sigma_z^2}\right) + \exp\left(-\frac{(z-H)^2}{2 \sigma_z^2}\right) \right\}$$
(2)

where:

- S_{xyz} concentration of (gaseous) contamination at point (x, y, z) [mg/m³],
 - E contamination emission [mg/s],
 - H effective height of monitor [m]: H = h + Dh, (Dh elevation of gases over the emitter of geometrical height h [m]),
 - \overline{u} average wind velocity in an air layer from z = h to z = H [m/s].

Equations (1) and (2) are valid on the assumption that: air is treated as incompressible medium, air movement is considered only in the horizontal direction (following the positive orientation of x axis), concentration field (S), and basic atmospheric factors are constant in time. The model also assumes that the distribution of contamination concentrations is a normal distribution with variances σ_x , σ_y , σ_z .

This simple calculation model for the purpose of evaluated impact of post-frac emission on air quality could be applied thanks to the adequacy of the model as far as the description of effects taking place at a local scale in an area of a simple orography and with the available statistics of meteorology data for the analyzed region are concerned.

A detailed description of the assumed methodics can be found in a Regulation of the Minister of Environment of 26 January 2010 about reference values for some substances in air (Official Journal no. 16, item 87) (RMŚ, 2010).

According to the assumed methodics, the calculation area was ascribed an average aerodynamic roughness of surface $z_0 = 0.35$ m. As far as meteorology data are concerned, the annual wind rose plot for Gdańsk was assumed (Fig. 2). It was statistically analyzed on the basis of meteorology data covering a 10-year period.

Calculations of dispersion of contaminants coming from pump aggregates in the course of hydraulic fracturing jobs allowed for evaluating spatial distribution of maximum 1 hour concentrations (S_1), incidence of exceeding 1 hour admissible concentrations ($P(D_1)$), percentile 99.8 and 99.726 from 1 hour concentrations and average annual concentrations (S_a) for selected contaminations which are most important for air quality, i.e.: NO_x (as NO₂), SO₂, CO, PM10, benzo(a) pyrene, benzene, toluene, xylene, formaldehyde, acetaldehyde and acrolein. For better evaluation of the effect of hydraulic fracturing on air quality the concentrations of selected substances in air were calculated on the rig area boundary (blue square in figs. 3 to 8), being boundaries of the lease/property of an operator performing prospecting drilling jobs. The results of calculations of concentrations of selected substances in air on the rig border are presented in table 9.



Fig. 2. Annual wind rose plot for Gdańsk

TABLE 9

	Adr (RM	nissible v Ś. 2010, 2	alue 2012)	Γ	Maximum value calculated at rig border			
Substance	D ₁	Da	P(D ₁)	S ₁	Percentile 99.8/ (99.726) z S ₁	Incidence of exceeding P(D ₁)	S _a	
	$[\mu g/m^3]$	$[\mu g/m^3]$	[%]	$[\mu g/m^3]$	[µg/m ³]	[%]	[µg/m ³]	
NO ₂	200	40	0.200	34330.9	10.7	0.17	15.9	
SO ₂	350	20	0.274	10.8	(0)	0	0.0050	
CO	30000	-	0.200	9119.1	2.8	0	4.2	
PM10	280	40	0.200	536.4	0.17	0.03	0.25	
B(a)p	0.012	0.001	0.200	0.0028	0	0	0	
Benzene	30	5	0.200	8.3	0.00259	0	0.0039	
Toluene	100	10	0.200	3.0	0.00094	0	0.0014	
Xylenemined by	100	10	0.200	2.07	0.00065	0	0.00096	
Formaldehyde	50	4	0.200	0.84	0.00026	0	0.00039	
Acetaldehyde	20	2.5	0.200	0.27	0.000080	0	0.00013	
Acrolein	10	0.9	0.200	0.085	0.000030	0	0.000040	

Concentrations of selected substances in air on the rig border

The analysis of data in table 9 reveals that none of the analyzed substances exceeds admissible 1 hour concentrations (D₁), average concentrations (D_a) in air and also incidence of exceeding admissible values $P(D_1)$. The most noxious for the air is nitrogen dioxide whose incidence of exceeding admissible concentrations D₁ on the boundary of the rig area is lower than the admissible value of 0.2 % of the year. This contamination also has very high momentary concentrations reaching as much as 34,331 µg/m³. Despite the fact that the 1 hour NO₂ concentration in air periodically exceeds the admissible value of D₁ over times 170, the duration of this episode

is short enough over the year's perspective that the admissible value determined by the percentil 99.8 is not exceeded. The average annual concentrations of nitrogen dioxide make up only ca. 40% of total admissible average concentration D_a . The spatial distribution of calculated 1 hour concentrations (S₁), incidence of exceeding of these values (P(D₁) and average concentrations (S_a) NO₂ are presented in figures 3 to 5.



Fig. 3. Spatial distribution of maximum 1 hour NO_x concentrations recalculated for NO_2 caused by emissions generated by hydraulic fracturing jobs [mg/m³]



Fig. 4. Spatial distribution of incidence of exceeding admissible 1 hour No_x, recalculated for NO₂ generated during hydraulic fracturing jobs [%]



Fig. 5. Spatial distribution of average NO_x concentrations recalculated for NO₂ caused by emission generated by the hydraulic fracturing job $[\mu g/m^3]$

Apart from nitrogen dioxide, another contamination, whose concentration at the rig area boundary was highest was dust PM10. Its maximum 1 hour concentration exceeded the admissible standard D_1 nearly twice, whereas the incidence of exceeding admissible concentrations D_1 was 0.03% of the year, i.e. immision standards have not been exceeded in this case. For illustrating the effect of dust PM10 emission from the hydraulic fracturing job on air the spatial distributions of calculated PM10 immision parameters are presented in figures 7 to 8.



Fig. 6. Spatial distribution of maximum 1 hour concentrations of PM10 caused by emission from the analyzed hydraulic fracturing job [mg/m³]

Fig. 7. Spatial distribution of admissible incidence of exceeding 1 hour concentrations of PM10 caused by emission from the analyzed hydraulic fracturing job [%]

All the remaining substances analyzed for their effect on air quality should be considered to be of little importance from the point of view of potential risk of air contamination. Only maximum 1 hour concentrations of carbon oxide and benzene approach the boundary of 30% of admissible value D_1 . Due to the usually short time in which hydraulic fracturing jobs are performed over the year, the average concentrations of emitted contaminations will not reach levels leading to the exceeding of admissible values D_a . The highest maximum of the substance concentration in air for all analyzed meteorological cases was always observed at a distance of 52 m from the source of emission, at wind speed of 1 m/s and steady state of equilibrium of the atmosphere.



Fig. 8. Spatial distribution of average annual concentrations of PM10 caused by emissions from analyzed hydraulic fracturing job $[\mu g/m^3]$

5. Conclusions

The hydraulic fracturing process is connected with the risk of occurrence of high and even excessive concentrations of contaminants in air as a result of exhaust gases produced by pump aggregates and other technological systems supporting the hydraulic fracturing jobs. Nitrogen dioxide and dust turn out to be critical air contaminants which may exceed the admissible standards. The risk of excessive concentrations of a given contaminant in air beyond the rig area mainly depends on the summaric power of engines used in the hydraulic fracturing jobs, extent to which they meet the emission limits and minimum distance of emission sources from the rig area boundaries.

The results of modelling of propagation of contaminants from 12 high-pressure engines of 1,680 kW each, which have not been equipped with modern emission control systems reveal that:

- 1) in Poland the immision standards beyond the rig area in are not exceeded,
- there is a potential danger of not meeting all environmental requirements regarding NO₂ concentrations (to a less degree in reference to PM10) in air on the rig area boundary, if:
 - 1. quantity of installed power of engined powering pump aggregates for hydraulic fracturing was higher than in the analyzed case,
 - 2. duration of fracturing job was longer than assumed in calculations,
 - 3. level of NO₂ background within the rig site was higher than 10 % of D_a ,
 - 4. poor quality fuel was combusted, e.g. heavy heating oil,
 - 5. other important sources of NO₂ and PM10 emissions were operating simultaneously in the rig area, synergically affecting air quality,
- owing to the shortness of emission exceeding of average annual concentrations (D_a) is not likely.

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