CH_4 and CO_2 monitoring in the air of underground coal mines in southern Brazil and GHG emission estimation

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Abstract

This study aims to assess methane (CH_4) and carbon dioxide (CO_2) concentrations in the ventilation systems of two coal mines (A and B) in the Santa Catarina coal deposit in southern Brazil (Paraná Basin, Bonito Formation), and estimate their greenhouse gas (GHG) emissions. The highest CH_4 levels (1.8%) were recorded in strong methane emanation areas in mine A, below the lower explosive limit (5%). The IPCC-recommended methods significantly overestimated the methane emission (up to 80%) when compared to the experimental data measured for each mine. Application of an alternative method made it possible to estimate direct CO_2 emissions, indicating that CO_2 accounted for 22 to 77% of total GHG emissions. Carbon dioxide emissions are generally not included in GHG emission inventories, indicating that the coal industry underestimates the contribution of this gas. As such, it is recommended that the methodology used for these calculations be revised and that specific emission factors be applied for each mine. In order to improve the accuracy of inventories, more sampling needs to be carried out in all operational and abandoned mines.

keywords: underground coal mines, emissions, greenhouse gases.

1. Introduction

Different gases are released during the coal extraction process, including hydrocarbons (methane, ethane, propane, butane and n-propane), carbon dioxide, nitrogen, and helium, among others. The composition of gases in the underground atmosphere is related to factors such as the breaking up of rocks and coal extraction, the decomposition of inorganic substances, underground water, equipment operation and ventilation systems, among others (Zipf and Mohamed, 2010). Methane (CH_4) can be released in coal mines in a variety of forms and concentrations. As methane emerges from the cracks and layers of coal, it mixes with the ventilating air in a gradual process of dilution, decreasing from concentrations of 15% to 5%, known as flammability or explosive limits (Kissel, 2006). Given the security risks involved, methane levels in coal mines must be constantly monitored (McPherson, 1993). Carbon dioxide (CO_2) is emitted in coal mines through the decomposition of organic matter by microorganisms, which act in geological formation. It is also exhaled by mine workers, and released in the combustion processes of machinery used in the mines and explosives used in coal blasting (Games *et al*., 1978). Although the risks associated with the presence of CO_2 in mines is lower when compared to CH_4, high levels of carbon dioxide can be harmful to the health of workers (McPherson, 1993).

Methane (CH_4) and carbon dioxide (CO_2) are the principal greenhouse gases (Denman *et al*., 2007). Coal mining is a major source of CH_4 (Denman *et al*., 2007), which is 21 times more warning potent than CO_2 (IPCC, 2006). In Brazil, fugitive emissions from coal mining and beneficiation were estimated in two inventories (MCT, 2006; MCT, 2010). These estimations were based on generic emission factors for CH_4 in underground and surface mining and for CO_2 emitted only after mining (coal stockpile oxidation). The validity of these coefficients is questioned, particularly for mines with low GHG levels, where CH_4 emissions can be overestimated (Silva *et al*, 2010). In addition, applying the same coefficient factor to all mines in a country or region does not encourage the production and consumption of coal with less global warming potential (GWP).

Data about direct CO_2 emission from coal mining, and greenhouse gas (GHG) inventories are sparse, and generally only report the indirect emission related to the burning of fuel by equipment and post-mining coal stockpiles (Sloss, 2013). However, recently in GHG Emissions Inventory for South Africa (DEA, 2016) a country-specific CO_2 Emission Factor for coal mining in underground mines was proposed.

To our knowledge, in Brazil there are no studies on the composition of ambient air in the ventilation systems of these mines, which precludes an accurate assessment of risks and GHG emissions. In this respect, the present study aims to assess the composition of ambient air, focusing on CH_4 and CO_2, in underground coal mines located in the Santa Catarina coal basin in southern Brazil. The results of GHG monitoring are used to estimate gas emissions, applying different calculation methods, and comparing them with data from the literature on other coal mines around the world.

2. Materials and methods

2.1 Coal mines

Two underground coal mines from the Barro Branco and Bonito seams were studied (A and B), located in the Santa Catarina coal basin and belonging to Paraná Basin, containing coal from the Rio Bonito Formation. Mines A and B were chosen as representatives of the two currently mined coal seams, exhibiting different methane concentrations in accordance with a previous study carried out by the research group (Silva *et al.*, 2010). Mine A also contains areas with high CH_4 emissions, which have yet to be examined in detail and may be a potential accident risk. The main characteristics of coal extracted from mine A are: Rank (ASTM): high volatile C bituminous coal; ash: 33.7 wt%; volatile matter: 43.7 wt% daf; fixed carbon: 56.3 wt% daf; gross calorific value: 5,460 kcal kg^{-1}. Coal characteristics in mine B are: Rank (ASTM): medium volatile bituminous coal; ash: 45.9 wt%; volatile matter: 44.2 wt%; daf; fixed carbon: 55.8 wt% daf; gross calorific value: 3,880 kcal kg^{-1} (Silva *et al.*, 2010; Kalkreuth *et al.*, 2010). Run of mine (ROM) coal production in mines A and B in 2016 were 595,000 t $year^{-1}$ and 960,000 t $year^{-1}$, respectively.

2.2 Gas sampling and analyses

Gas samples were collected from the mines in four sampling campaigns, between 2014 and 2016. Sampling was carried out at the main entry and return points of the mines, in the ventilation intake and return airways, exhaust, and gas emanation areas, following validated procedures (Bonetti *et al.*, 2016). Two sampling containers were used: a borosilicate glass flask with two septa (PTFE/Silicone and Butyl rubber/PTFE, Supelco) and multilayer PE/aluminum sampling bags (Supelco). The concentration of the carbon dioxide and methane in air samples were determined following an optimized method using a gas chromatograph (PerkinElmer Clarus 580) equipped with a flame ionization detector (FID), methanator and a mega-bore Elite-Q Plot column. In addition to gas sampling, temperature (°C), atmospheric pressure (mbar) and wind speed (m s^{-1}) were measured using a weather meter (Kestrel® 4000NV).

2.3 Greenhouse gas emission estimates

The calculations used to estimate GHG emissions from these mines were based on methods developed by IPCC and compared to studies conducted by Harpalani and Prusty (2009). The first methodology applied (M1) to estimate annual CH4 emission was based on the Tier 1 method (Equation 1), developed by IPCC (2006):

$$E_{M1\ Tier\ 1}\ (Gg_{CH_4}) = EF \times TPC \times CF \qquad (1)$$

where EF is the emission factor (10 m^3 CH_4 t^{-1}, for low emission mines); TCP is the total coal production in t per $year^{-1}$; and CF is the conversion factor (0.67×10^{-6} Gg m^{-3}).

Due to the variability of gas levels along the ventilation airways of the mines, another method (M2) was used to determine annual CH_4 emissions based on Tier 3 (Equation 2), also developed by IPCC (2006) and presented by Irving and Tailakov (1999). The method was designed to replace Tier 1 in mines with low CH_4 levels (in our case 0.05% were used), considering the estimated ventilation airflow.

$$E_{M2\ Tier\ 3}\ (Gg_{CH_4}) = \frac{0.05}{100} \times TFD \times 365\ days \times CF \qquad (2)$$

where TDF is the total daily flow of gas at the ventilation outlet (m^3 day^{-1}).

The third methodology (M3) uses the average gas levels in the mine and airflow from the ventilation system (Equation 3). In this study, flow rates considered in this calculation were those at the ventilation outlets. Additionally, CO_2 monitoring at these points made it possible to estimate CO_2 emissions (Equation 4) using this methodology (M3).

$$E_{M3}\ (Gg_{CH_4}) = \Delta C\ (CH_4) \times TPF \times CF \qquad (3)$$

$$E_{M3}\ (Gg_{CO_2}) = \Delta C\ (CO_2) \times TPF \times CF \qquad (4)$$

where ΔC is the gas concentration variation between outlet and inlet air ventilation concentrations (ppm), TDF the total daily flow of gas at the ventilation outlet (m^3 day^{-1}) and CF the conversion factor. To estimate global emission, expressed as CO_2-equivalent, CO_2 and CH_4 emissions are multiplied by the global warming potential (GWP) of each gas (1 and 21, respectively, IPCC, 2006).

3. Results

3.1 CH_4 and CO_2 concentrations in underground coal mines

Table 1 shows the concentrations of the gases collected in the four sampling campaigns in mine A and B, which extracts coal from the Santa Catarina coal deposit in the Barro Branco and Bonito seams, respectively.

Methane was not detected (below LOD 4 ppm) at the entry points of mine A in any of the sampling campaigns. By contrast, CO_2 levels ranged from 751 to 992 ppm, similar to the values recorded in outdoor air around the mine (~800 ppm). The variation observed in the different campaigns may be related to intensity of the (external) emitting sources near the mine entrance, such as internal combustion equipment and vehicles. CH_4 and CO_2 levels increased along the ventilation airways, reaching significant values at the ventilation return and outlet points. A 30 to 85-fold increase was observed in methane levels, confirming significant emission of this gas during mining operations.

It is important to underscore that there were 170 and 2-fold increases in CH_4 (669 ppm) and CO_2 (1,913 ppm) concentrations, respectively, in this mine during the 2nd sampling campaign, following a detonation event. However, the highest levels in this study were recorded in the methane emanation areas, previously mapped by the mining company. In the 1st campaign, maximum levels of 18,006 ppm (1.8%) and 6,086 ppm were observed for CH_4 and CO_2, respectively, in the emanation areas. These were the highest concentrations measured for the two gases throughout the study. Two different emanation areas were monitored in the 2nd campaign, obtaining values of 1,137 to 3,523 ppm and 1,339 to 1,691 ppm for CH_4 and CO_2, respectively. The strongest emanation area, assessed in the 1st sampling campaign, was inaccessible during the 2nd campaign because it was partially depleted and flooded with water, apparently as a safety measure against the risk of explosion.

CH_4 and CO_2 levels also increased along the ventilation airways in mine B, but were less significant than those observed in mine A. Carbon dioxide content ranged from 718 to 894 ppm at ventilation entry points, close to the values recorded for external air. A 4 to 15-fold rise in methane concentration (<4 ppm to 13-54 ppm) at the ventilation outlet. Less significant CO_2 increases (1.5 to 3-fold) were observed in the ventilation airways, albeit higher than those recorded in mine A. As previously mentioned, the rise in CO_2 content may be due to a variety of other sources in the mines, which explains the differences recorded.

Table 1
Concentrations of CH_4 and CO_2 in different sites of the mines A and B in four sampling campaigns.

Sampling Sites	Mine A		Mine B	
	CH_4 (ppm)	CO_2 (ppm)	CH_4 (ppm)	CO_2 (ppm)
1st Campaign/2014				
Mine entry	<4[a]	849 ± 28	<4[a]	718 ± 21
Ventilation Circuit	117 ± 1	1,194 ± 8	96 ± 17	1,661 ± 21
	143 ± 21	1,226 ± 2	51 ± 4	1,626 ± 19
Emanation Areas	18,006 ± 865	6,086 ± 207		
Ventilation Outlet	213 ± 10	1,624 ± 25	54	1,748
2nd Campaign/2015				
Mine entry	<4 [a]	793 ± 100	<4[a]	756 ± 100
After detonation	669 ± 100	1,913 ± 365		
Emanation Areas	3,523 ± 69	1,339 ± 25		
	1,137 ± 332	1,691 ± 200		
Ventilation Outlet	143 ± 2	1,256 ± 947	34 ± 3	1,295 ± 168
3rd Campaign/2016				
Mine entry	<4 [a]	751 ± 52	<4[a]	741 ± 203
Ventilation Outlet	110 ± 41	1,012 ± 117	26 ± 0	1,375 ± 16
4th Campaign/2016				
Mine entry	<4 [a]	992 ± 168	<4 [a]	894 ± 117
Ventilation Outlet	338 ± 146	1,565 ± 174	13 ± 3	1,346 ± 143
Security Limits				
NR15 or NR22[b]	1%	3,900	1%	3,900
MSHA TLV[c]	n.a.[d]	5,000	n.a.[d]	5,000

[a]Limit of Detection (LOD) of CH_4; [b]CO_2 workplace tolerance limit (NR15, 2014) and CH_4 tolerance limit (NR 22, 2018) in Brazil; [c]Threshold Limit Values (TLV) by Mine Safety and Health Administration (MSHA, 2001) in the United States; [d]Not allowed.

No emanation areas were observed in mine B and the highest methane levels recorded at the ventilation outlet, were 5 to 9 times lower than those measured in mine A. The carbon dioxide content at the ventilation outlet varied from 1,295 to 1,748 ppm, similar to the levels obtained in mine A.

3.2 Greenhouse gas direct emission models on underground coal mines

Different methodologies were applied to estimate GHG emissions from the mines, obtaining the values shown in Table 2. The maximum and minimum levels are displayed, calculated based on the four sampling campaigns in mines A and B. In all cases, estimates were made using the three methods described in the experimental section. Methods M1 (Tier 1) and M2 (Tier 3) follow the procedure recommended by the IPCC, using generic emission factors based on coal production (10 m^3 CH4 t^{-1} of coal produced) and a fixed methane concentration emitted into the atmosphere. A low CH_4 concentration (500 ppm) was selected for the calculations, within the range recommended by IPCC, since Brazilian mines are not considered gassy (Silva *et al.*, 2010). Method M2 also uses a range of experimentally measured daily gas flows at the ventilation outlet (TDF). The M3 method differs from the others in that it is based on concentrations and flow rates measured in the field, for each of the mines studied (Table 1). Due to the variation observed, maximum and minimum concentrations of the gases analyzed were used.

There was a significant difference in CH_4 emissions between the three methods (Table 2). For mine A, range emissions of 3,984; 755-798 and 104-487 t CH_4 $year^{-1}$ were estimated by M1, M2 and M3, respectively. When compared to the maximum emission value estimated by M3, overestimation of emissions was 2 to 9 times greater for the other two methods. Differences were even more significant in mine B (20 to 115 times).

In this study, the CO_2 emitted by mining activities was also estimated. This is not normally done because there are no IPCC-recommended emission factors for carbon dioxide. However, since an increase in CO_2 levels was observed along the ventilation airways of the mines studied (Table1), it can be inferred that this gas is also generated during mining operations. The same calculation methodology used for methane in M3 was applied for CO_2 and the results are displayed in Table 2.

Mine	Methodology	Gas	Unit	Emission			
				Min.	%	Max.	%
A	Method M1 *(Tier 1)*	CH_4	t CH_4 $year^{-1}$	3,984		3,984	
	Method M2 *(Tier 3)*	CH_4	t CH_4 $year^{-1}$	755		798	
	Method M3	CH_4	t CH_4 $year^{-1}$	104		487	
		CH_4[a]	t CO_{2eq} $year^{-1}$	2,184	78	10,227	67
		CO_2	t CO_2 $year^{-1}$	626	22	5,133	33
	Total GHG emission		t CO_{2eq} $year^{-1}$	2,810	100	15,360	100
B	Method M1 *(Tier 1)*	CH_4	t CH_4 $year^{-1}$	6,432		6,432	
	Method M2 *(Tier 3)*	CH_4	t CH_4 $year^{-1}$	377		1,120	
	Method M3	CH_4	t CH_4 $year^{-1}$	28		56	
		CH_4[a]	t CO_{2eq} $year^{-1}$	588	30	1,185	23
		CO_2	t CO_2 $year^{-1}$	1,390	70	4,037	77
	Total GHG emission		t CO_{2eq} $year^{-1}$	1,978	100	5,222	100

[a] Methane emission estimated by method M3 was converted in CO_2-equivalent unit using CH_4 GWP of 21 (IPCC, 2006).

Table 2
Estimates of GHG emissions from underground mines by different methodologies.

4. Discussion

4.1 Effects of CO_2 and CH_4 in ambient air of underground coal mines

Variations in gas levels were recorded in both mines, being more significant for mine A, throughout the four sampling campaigns. For example, the methane levels measured at the ventilation outlet ranged from 110 ppm (3rd campaign) to 338 ppm (4th campaign). It is important to emphasize that these differences may be associated with fluctuations in the ventilation operation and coal production (Pinto *et al.*, 2003).

The higher concentrations of methane in the emanation areas were likely due to geological faults. A study conducted by Oliveira (2009) identified the occurrence of oily sandstone near the coal seam mined in mine A, generating hydrocarbons that can migrate into the coal layers through fractures, thereby increasing emissions.

Increases in CO_2 concentrations were less significant (1.4 to 2.1-fold); likely indicating different sources and production mechanisms for these two gases in these environments. In addition to mining activities, the rise in CO_2 levels is largely due to the exhaled breath of mine workers and the engines of equipment used inside the mine. Other potential sources of this gas are the decomposition of organic matter (wood from pillars) and spontaneous oxidation of coal by the ventilated air (Yuan and Smith, 2011). Decaying wood may contribute to the rise in CO_2 levels since it is widely used in this mine. On the other hand, there is no information on spontaneous coal fires in mines in the Santa Catarina coal deposit.

Methane is a flammable gas that is at risk of exploding when mixed with air at concentrations between 5 and 15% (Kissel, 2006). The maximum CH_4 level (1.8%), measured in an emanation area, was below the lower explosive limit. Therefore, it does not pose an immediate risk of explosion but according to NR22 (topic 22.28), methane concentration above 1% must not be permitted in underground mines. Continuous monitoring of these areas and emission sources is recommended. Generally, a secondary ventilation system is installed at these sites to lower methane levels in these environments and reduce the risk of accidents (Hartman *et al*., 2012). Brazilian regulatory standard NR15 (2014) stipulates a workplace exposure limit of 3,900 ppm for CO_2, including in underground mines. The American Mine Safety and Health Administration (MSHA, 2001) establishes a coal mine-specific threshold limit value (TLV) of 5,000 ppm for this gas. Carbon dioxide levels above these regulatory guidelines (6,086 ppm) were only recorded at one collection point (emanation area) during the 1st sampling campaign. This result confirms the need for better ventilation in CO_2 emanation areas in this mine.

Mine A mines the Barro Branco seam, which contains higher-ranking coal when compared to the Bonito seam mined by mine B, and probably a higher methane content. As stated earlier, other operating parameters, such as ventilation and coal production, can also contribute to the differences in CH_4 levels in these mines. With respect to safety thresholds, all the CH_4 concentrations recorded were below the explosive range (5-15%). Carbon dioxide levels were also below the limits stipulated by NR15 and MSHA.

These results suggest that CO_2 concentration is governed by similar sources in the two mines, such as workers' exhaled breath and internal combustion engines. By contrast, methane levels appear to be associated primarily with the geological characteristics of the coal seam mined.

4.2 Greenhouse gases emission estimation from brazilian coal mines

The significant differences on CH_4 emissions by the three methods evaluated were expected because the IPCC's emission factors were obtained using data from mines with different characteristics from those in Brazil (low-rank coal with a higher methane content). In the case of method M2, the methane level used in the calculations (500 ppm) was higher than the experimental values (Table 1) of 338 and 13 ppm obtained at the ventilation outlet (exhausts) during the fourth campaign in mines A and B, respectively.

A comparison of the methane emission results (Table 2) obtained by method M3 in mines A and B shows that the former emitted 4 to 9 times more methane than the latter. This corroborates the previous data, demonstrating the need to determine individual emission factors for each mine. It should be noted that methane emissions (t CH_4 year^{-1}) were converted into CO_2 equivalents (t CO_{2eq} year^{-1}) using the emission factor recommended by IPCC.

As observed for methane, the highest CO_2 emissions (5,133 t CO_2 year^{-1}) were recorded in mine A, corroborating the presence of different mechanisms/sources of CH_4 and CO_2 formation in the mines studied. Unfortunately, there are no IPCC recommended methods for estimating direct CO_2 emissions in underground coal mines. As previously stated, Cook (2013) highlighted the importance of direct CO_2 emissions from underground coal mines, which led to the inclusion of the gas in the latest GHG emissions report for South Africa (DEA, 2016). However, CO_2 contributions are small (7 %) when compared to CH_4 emissions, when reported on a CO_2-equivalent basis (Cook, 2013). By contrast, the results obtained here indicate that CO_2 accounted for 22 to 77% of GHG emissions by the Brazilian mines studied, due to their low methane emissions (Table 2).

As CO_2 emissions were not included in GHG emission inventories for the Brazilian coal sector (MCT, 2006, 2010), its values are underestimated. By contrast, according to method M3 estimates (Table 2), methane emissions may be overestimated. These facts raise significant doubts about the values calculated by IPCC methods (M1 and M2). Thus, these methods should be revised and specific emission factors used for each mine.

Table 3 shows a comparison of the CH_4 emissions obtained in this study and those recorded in underground mines in other countries. The mines selected for comparison exhibited low methane concentrations at the ventilation outlet (≤ 0.2%) and were considered non-gassy.

Harpalani (2009) studied two underground mines in India and reported CH_4 levels at the ventilation outlet of 0.02 to 0.2% and emissions between 740 and 6,342 t CH_4 year^{-1}. Lloyd and Cook (2005) analyzed CH_4 emissions in six underground coal mines in South Africa and found lower and less varied CH_4 levels (0.002 to 0.04%) than those recorded in the Indian mines. Emissions in some South African mines were significant (5,310 t CH_4 year^{-1}, Koornfontein mine) due to their high coal production (~5x10^6 tyear^{-1}).

By contrast, Su *et al*. (2011) reported the mean methane concentrations in several mines in China, grouped according to the country's different coal regions. Table 3 shows two of these groups (D1 and D2), which exhibit low methane emissions, with mean values between 0.06 and 0.12%. The average estimated CH_4 emission potential ranged from 4,754 to 5,427 t CH_4 year^{-1} between the groups, reflecting high coal production in the different mine groups. In another two groups of mines in India, Singh (2016) recorded average methane levels below 0.1% and emissions of 100,000 and 431,000 t CH_4 year^{-1} between groups, higher than the values reported for the Chinese groups. Methane concentrations in the Brazilian mines (0.001 to 0.03%) are comparable to those observed in the South African mines and slightly lower than those of the Indian and Chinese mines. Mine B displays one of the lowest mean methane levels (0.003%) among the mines listed in Table 3. On the other hand, the annual emissions of the Brazilian mines (28 to 487 t CH_4 year^{-1}) are significantly lower than those displayed by the others countries, with the exception of some South African mines. These findings corroborate the low concentrations obtained in the present study as well as the low coal production of the Brazilian mines (0.6 a 1x10^6 t year^{-1}).

The variation in coal production and total air volume at the ventilation outlet of the mines listed in Table 3 precluded a direct comparison with CH_4 emissions. As such, an emission factor (EF) was used, based on the mean CH_4 volume emitted (expressed in m^3) divided by coal production (t). The Indian mines exhibited higher EFs, particularly the Moonidih mine, with values far higher than its counterparts (up to 12.6 m^3 t^{-1}). The EF of Brazilian mine A (0.4 to 1.2 m^3 t^{-1}) is within the range reported for the Chinese and South African mines, whereas mine B shows the lowest EFs among all the mines studied, confirming the previously discussed results. It is important to emphasize that the coal extracted in mine A is from the Barro Branco seam, whose properties are superior (i.e. higher grade) to those of coal from the Bonito seam (mine B). This partially explains the EF values observed.

The IPCC (2006) recommends a generic EF of 10 $m^3\ t^{-1}$ for low emission mines. This value is used to calculate emission according to the Tier 1 method (M1) when more accurate data are not available. Only one of the Indian mines exhibited similar values to this generic EF, while values for the remainder were 6 to ~300 times lower. Given that most of the coal mined in some countries shows low CH_4 emission potential, using the recommended generic EF could significantly compromise the accuracy of national GHG emission inventories related to coal mining.

The results obtained in this study are an attempt to enhance GHG emission estimates. However, in order to improve the accuracy of inventories, more sampling needs to be carried out in all operational and abandoned mines.

Table 3
Methane emission by underground coal mines in different countries estimated using different methods.

Country	Mine	Estimation method	[CH_4] in VAM range %	[CH_4] in VAM average %	Methane Emission t CH_4 $year^{-1}$	Methane Emission million m^3 $year^{-1}$	EF calculated ($m^3\ t^{-1}$)	Reference
Brazil	Mine A	Tier 1			3,984	0.6	10	This study
		Tier 3		0.05	755-798	1.15 - 1.21		
		Alternative	0.01 - 0.03	0.02	104-487	0.15 - 0.73	0.40-1.22	
	Mine B	Tier 1			6,432	9.8	10	
		Tier 3		0.05	377 – 1,120	0.57 - 1.70		
		Alternative	0.0013 - 0.0055	0.0034	28 - 56	0.03 - 0.08	0.03 - 0.09	
India	Moonidih	Alternative	0.1 - 0.2	0.15	6,342	9.4	6.3-12.6	Harpalani, 2009
	Sudamdih		0.02 - 0.04	0.03	740	1.10	0,74 - 1.48	
	Low gassy mines	Tier 1	<0.1		431,000	634	10	Singh, 2016
	(Degree I)	Alternative	<0.1		100,000	149	2.91	
China	Mine Region D.1	Alternative	0.06 - 0.1	0.07	5,427	8.1	0.8	Su, 2011
	Mine Region D.2		0.06 - 0.12	0.09	4,754	7.1	0.7	
South Africa	Koornfontein	Alternative	0.04		5,310	7.9	0.70	Lloyd & Cook, 2005
	Twistdraai		0.01		1,716	2.6	1.01	
	Matla		0.01		1,092	1.6	0.41	
	Douglas		0.005		446	0.7	0.07	
	New Denmark		0.01		301	0.4	0.27	
	Boschmans		0.002		89	0.1	0.01	

VAM: Ventilation air methane; EF: Emission factor.

5. Conclusions

A significant variation in methane levels was observed not only between the mines studied, but also between sampling campaigns in a same mine. The highest CH_4 levels were recorded in strong methane emanation areas in mine A. Although these values were below the explosive range and therefore posed no immediate risk, levels were still high and continuous monitoring of both the area and emanation source is recommended. By contrast, CO_2 exposure limits were exceeded in some of the emanation areas, indicating the need for increasing ventilation at these sites.

Three methods for estimating greenhouse gas (GHG) emissions by underground coal mines were compared and the results obtained showed significant variation in methane emissions between

the mines studied. The IPCC recommended methods significantly overestimated methane emission when compared to the experimental data measured for each mine. The application of an alternative method (M3) made it possible to estimate direct CO_2 emissions from coal mining activities. Significant levels of this gas were recorded, demonstrating that CO_2 contributed to the total GHG emissions of the mines analyzed. Carbon dioxide emissions are generally not included in GHG emission inventories, indicating that the coal industry underestimates the contribution of this gas.

The results obtained here highlight the uncertainties involved in estimating emissions. As such, we recommend that the methodology used for these calculations be revised and that specific emission factors be applied for each mine. It is important to underscore that the results obtained in this study are an attempt to enhance GHG emission estimates. However, in order to improve the accuracy of inventories, more sampling needs to be carried out in all operational and abandoned mines.

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References

BONETTI, B., ABRUZZI, R. C., PIRES, M. J. R., DEDAVID, B. A., BITENCOURT, A. K., GOMES, C. J. B. Monitoring CH_4, CO_2 and CO concentrations on the air of the underground coal mines in Southern Brasil, In: WORLD MIN. CONGR., 24th, 2016. Rio de Janeiro. ***Annals...*** Rio de Janeiro: IBRAM, 2016, v. 1, p. 348-356.

COOK, A. P. Fugitive greenhouse gas emissions from South African coal mines. ***Executive Summary.*** Coaltech, South African, 2013.

DEA - DEPARTMENT OF ENVIRONMENTAL AFFAIRS OF SOUTH AFRICA, GHG Inventory for South Africa: 2000 – 2012, Report, 2016. http://www.saaqis.org.za/, accessed: July 2018.

DENMAN, K. L. et alii. 2007: Couplings between changes in the climate system and biogeochemistry. In: ***Climate Change 2007: the physical science basis. Contribution of working group i to the fourth assessment report of the IPCC.*** In: SOLOMON, S., D. QIN, M. MANNING, Z. CHEN, M. MARQUIS, K. B. Averyt, M. TIGNOR, MILLER, H.L. (Eds). Cambridge: University Press, Cambridge, United Kingdom and New York, NY, USA.

EPA, Greenhouse Gas Reporting Program 2014: Underground Coal Mines, https://www.epa.gov/ghgreporting/ghgrp-2014-underground-coal-mines. Acessed July 2018.

GAMES, L. M., HAYES, J. M., GUNSALUS, R. P. Methane-producing bacteria: natural fractionations of the stable carbon isotopes. ***Geochimica et Cosmochimica Acta.*** Oxford, v. 42, n. 8, p. 1295-1297, 1978.

HARPALANI, S., PRUSTY, B. K. ***Quantification of Ventilation Air Methane Emission from Gassy Underground Coal Mines in India.*** Washington, USA: Environmental Protection Agency (USEPA), 2009.

HARTMAN, H. L. et alii. ***Mine ventilation and air conditioning.*** (3ª Ed). New York: John Wiley & Sons, 2012. 752 p.

IPCC. Guidelines for National Greenhouse Gas Inventories. Fugitive Emissions. IPCC – Intergovernmental Panel on 27 Climate Change. 2006. v. 2, cap. 4. 78 p. http://www.ipcc-nggip.iges.or.jp/ Acessed July 2018.

IRVING, W., TAILAKOV, O. ***CH_4 emissions: coal mining and handling.*** Good Practice Guidance and Uncertainty Management in National Greenhouse Gas Inventories, 1999. 16 p. http://www.ipcc-nggip.iges.or.jp/ . Acessed July 2018.

KALKREUTH, W. et alii. Depositional setting, petrology and chemistry of Permian coals from the Paraná Basin: 2. South Santa Catarina Coalfield, Brazil. ***International Journal of Coal Geology,*** v. 84, n. 3, p. 213-236, 2010.

KISSELL, F. N. ***Handbook of methane control in mining.*** Department of Health and Human Services, 2006. 184 p.

LLOYD, P. J. D., COOK, A. Methane release from South African coalmines. ***Journal of the South African Institute of Mining and Metallurgy,*** v. 105, n. 7, p. 483-490, 2005.

McPHERSON, M. J. ***Subsurface ventilation and environmental engineering.*** New Delhi, India: Springer-Science & Business Media, 1993. 912 p.

MCT, FIRST BRAZILIAN INVENTORY. EMISSIONS OF GREENHOUSE GASES IN THE ENERGY SECTOR: Fugitive Emissions in Mining and Mineral Coal Processing. Brazilian Association of Mineral Coal (ABCM). Ministério da Ciência e Tecnologia. 2006, 79 p.

MCT, SECOND BRAZILIAN INVENTORY. EMISSIONS OF GREENHOUSE GASES IN THE ENERGY SECTOR: Fugitive Emissions in Mining and Mineral Coal Processing. Brazilian Association of Mineral Coal (ABCM). Ministério da Ciência eT. 2010, 82 p.

MSHA: Mine Safety and Health Administration (2001). Federal Register (n. 13, v. 66. 182 p) EUA.

NORMA REGULAMENTADORA: NR15 (2014). Atividades e Operações Insalubres (Portaria MTb nº 3.214) Brasília, Brasil, 82 p.
NORMA REGULAMENTADORA: NR22 (2014). Segurança e Saúde Ocupacional na Mineração, Ministério do Trabalho, Brasília, Brasil, 4 p.
OLIVEIRA, E. B. ***Non-conventional generation of hydrocarbons in the coal-mining region of Santa Catarina.*** Rio de Janeiro: Universidade do Estado do Rio de Janeiro, 2009. 103 p. (Master Thesis).
PINTO, P. C. et alii. Construção de um modelo computacional para o circuito de ventilação da Mina Esperança. ***REM - Rev. Esc. Minas,*** v. 56, n. 4, p. 243-248, 2003.
SILVA, R., et alii. Monitoring light hydrocarbons in Brazilian coal mines and in confined coal samples. ***International Journal of Coal Geology,*** v. 84, p. 269–275, 2010.
SINGH, A. K., KUMAR, J. Fugitive Methane emissions from Indian Coal Mining and handling activities: estimates, mitigation and opportunities for its utilization to generate clean energy. ***Energy Procedia,*** v. 90, p. 336-348, 2016.
SLOSS, L. ***Quantifying emissions from spontaneous combustion, CCC/224.*** IEA Clean Coal Centre: September 2013.
SPEIGHT, J. G. ***Handbook of coal analysis.*** (2ª Ed.) New Jersey: John Wiley & Sons, 2015. 367 p.
SU, S. et alii. Fugitive coal mine methane emissions at five mining areas in China. ***Atmospheric Environment,*** v. 45, n. 13, p. 2220-2232, 2011.
SÜFFERT, T., CAYE, B. R., DAEMON, R. F. ***Projeto Carvão Bonito Gaseificável.*** Porto Alegre, DNPM/CRPM, v. 1, 1977. 158 p. (Relatório Final).
XUEHAI, F. et alii. Prevention and origin of exceptional deleterious gas compositions in coal mine. ***Procedia Engineering,*** v. 26, p. 424-430, 2011.
YUAN, L., SMITH, A. C. CO and CO_2 emissions from spontaneous heating of coal under different ventilation rates. ***International Journal of Coal Geology,*** v. 88, n. 1, p. 24-30, 2011.
ZIPF, R. K., MOHAMED, K. M. Composition change model for sealed atmosphere in coal mines. In: UNITED STATES/NORTH AMERICAN MINE VENTILATION SYMP., 13th, 2010. In: HARDCASTLE, S., MCKINNON, D. (Ed.). Sudbury: Laurentian University, 2010. p. 493-500.

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