STABLE CARBON ISOTOPE COMPOSITION OF METHANE RELEASED TO THE ATMOSPHERE IN THE UPPER SILESIAN COAL BASIN

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Introduction

Methane is the second most important greenhouse gas. Methane emissions associated with coal production (mining, transport and storage on the surface) are responsible for approximately 12% of the global anthropogenic flux of this gas to the atmosphere (Saunois et al., 2016). In regions of active coal mining methane emissions may lead to elevated concentration of this gas in the near-ground atmosphere.

Upper Silesian Coal Basin (USCB) is the largest active coal mining region in Europe. Emissions of methane associated with mining activities in USCB fluctuated during the period 2001-2011 between ca. 6.46×10^8 and 7.35×10^8 m^3yr^{-1} (Patyńska, 2014). This flux constituted ca 36% of the total methane flux to the atmosphere emitted from the territory of Poland (KOBiZE, 2018).

This work was focused on quantification of stable carbon isotope composition of methane ($\delta^{13}C$) associated with coal mining activities in USCB. The carbon isotope composition of methane is a powerful tool to constrain the global methane budget (e.g. Schaefer et al., 2016). It is also widely used to decipher the origin of methane.

Samples and methods

To constrain $\delta^{13}C(CH_4)$ released to the atmosphere in USCB, two types of measurements were performed: (i) sampling of air released from ventilation shafts of the mines and isotope analyses of CH_4 in the laboratory, and (ii) indirect method based on mobile measurements of CH_4 concentration and $\delta^{13}C(CH_4)$ in the vicinity of mines. The later method is based on two-component mixing approach where CH_4 present in regional atmosphere is mixed with CH_4 released from the mines thus modifying its atmospheric concentration and isotope composition. The $\delta^{13}C$ of methane being released to the atmosphere by a local source (mine) was calculated from linear relationship between $\delta^{13}C(CH_4)$ in the mixture and the reciprocal of the measured CH_4 concentration. Ground-level measurements were supplemented by extraction of near-surface soil gas and analyses of $\delta^{13}C(CH_4)$ (Sechman et al., 2017). Analyses of $\delta^{13}C$ of CH_4 retrieved from different levels in selected methane mines were also included in the assessment (Kotarba, 2001, and unpublished data).

Ground-level field campaigns were conducted over the 2013-2018 period and comprised in total 12 mines located in two areas of the USCB: (i) north and north-western region, and (ii) southern region. Those two regions differ greatly in terms of CH_4 emissions - major methane-bearing mines are located in the southern part of USCB. The $\delta^{13}C$ of CH_4 in the near-surface zone (depth interval 1.2-1.5m b.g.l.) was analysed along four transects covering southern part of USCB (Sechman et al., 2017). Ground-level measurements of atmospheric CH_4 were performed with laser-based instrument (Picarro 2201-*i*). CH_4 retrieved from soil gas and methane collected in mines were analysed using Finnigan Delta Plus isotope ratio mass spectrometer.

Results

The results of carbon isotope analyses of methane encountered in the USCB are summarized in Table 1. They represent three types of methane: (i) methane emissions into the atmosphere measured on the ground, (ii) methane in the near-surface zone, and (iii) methane in mines.

Table 1. *Overview of stable carbon isotope composition of various forms of methane encountered in the USCB.*

Type of methane sources	Average $\delta^{13}C$ (‰)	Method
Methane emissions from mines		
North and north-western region of USCB: (mines: Staszic, Śląsk, Wieczorek, Wujek, Sośnica)	-48.5±1.1	sampling in vicinity of mines (Keeling plot); 17 campaigns.
Southern region of USCB:		
(mines: Brzeszcze, Krupiński)	-57.9±5.6	sampling in vicinity of mines (Keeling plot); 5 campaigns.
(mines: Borynia, Brzeszcze (3 shafts), Marcel (2 shafts), Pniówek (3 shafts), Jastrzębie (2 shafts))	-50.1±3.9	air samples collected from ventilation shafts and analysed in the lab.
Methane from near-surface zone		
Southern region of USCB (Sechman et al., 2018):	-59.8±5.4	15 soil gas samples collected along four transects
Coal-bed methane in mines		
Southern region of USCB (Kotarba, 2001; unpublished data)		54 coal-bed gas samples collected from 4-6 meter long holes drilled in the virgin parts of the coal deposits.
Depth interval: 0–500 m b.g.l. (25)	-69.9±7.5	
500–1000 m b.g.l. (39)	-61.3±11.2	11 gas samples collected from wells drilled from the surface
1300 m b.g.l. (1)	-43.4±0.2	

Conclusions

Extensive survey of carbon isotope composition of methane releases from the region of USCB revealed relatively narrow range of $\delta^{13}C$ values, with the grand average around -50 ‰. Interestingly, this value is much lower than that adopted by Schaefer et al. (2016) for the releases associated with coal mining and natural gas and oil production (-35‰). Methane extracted from near-surface zone reveals substantially more negative $\delta^{13}C$ values, around -59.8‰, pointing to its microbial and partly thermogenic, coal-bed origins. More negative $\delta^{13}C$ values can be also traced in the upper 500 meters of the surveyed mines, in contrast to less negative $\delta^{13}C$ values observed at greater depths. Highest $\delta^{13}C$ values of methane (around -43‰) were found in boreholes at depths of ca. 1300 m a.g.l.

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