

## Methane concentration and isotope composition analyses over the Silesian Coal Basin, Poland.

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Methane emissions from one of European regions associated with coal excavation industry constitute an important component of the continental anthropogenic flux of this gas into the atmosphere. The region of Silesian Coal Basin (SCB), located in the Silesian province, Poland has approximately 1.5 x 1.5 degrees of latitudinal and longitudinal dimension and is responsible for between 450 – 1350 Gg CH<sub>4</sub> of atmospheric methane releases annually, making it one of the most significant sources of this gas in Europe. Large discrepancies between the reported values of CH<sub>4</sub> emission rate are due to the different reporting methodology applied by the data providers.

In this region, active or restructuring coal mining methane emissions may lead to elevated concentrations of this gas in near-ground atmosphere. Observed methane mixing ratio in pbl during nighttime over the specific areas of SCB is elevated by up to 50ppm, on the public roads in vicinity of the mine shafts. Apart from CH<sub>4</sub> emissions associated with coal production, other sources of anthropogenic methane are also abundant in SCB. These include city gas networks leakages that enrich the air by up to 5ppm (in the city centers) and numerous landfills. Most of the landfills, not yet equipped with appropriate CH<sub>4</sub> uptake installations, also contribute to substantial anthropogenic flux of this gas to the atmosphere. Values of methane mixing ratio recorded during the in-situ measurements close to the landfill sites reached 15ppm.

To test the impact of emissions of anthropogenic methane in the region we launched a dedicated study aimed at characterizing spatial and temporal variability of CH<sub>4</sub> mixing ratios in near-ground atmosphere. Measurements of CH<sub>4</sub> mixing ratios were performed along latitudinal transects (ca. 50°N), typically extending from ca. 15°E to 20°E, covering the Upper Silesia and bordering regions. Occasionally, transects were extended to entire southern Poland and northern Slovakia. The measurement campaigns were performed repeatedly during the time periods: 1998-2000, 2004 -2013 and again in 2014 and 2015 under the InGOS TNA projects. Different analytical techniques were applied for direct measurements of methane concentration. At the beginning only spot samples were collected. In later years the sampling was supplemented by in-situ, quasi-continuous measurements of CH<sub>4</sub> mixing ratios using portable CH<sub>4</sub> analyzer (Gascorder) mounted on a car. Finally direct methane concentrations were obtained with Picarro CRDS analyzer. Since 2013 the sampling program has included collection of air samples in flasks, which were then analyzed for CH<sub>3</sub>D in the IMAU (Utrecht University) laboratory. Also, for the most interesting location, bag samples were collected and sent to the RHUL (London University) laboratory for determination of <sup>13</sup>CH<sub>4</sub> abundance. The measured CH<sub>4</sub> concentrations were compared with regional, free-tropospheric concentrations of methane obtained from high-altitude mountain station located ca 150 km south-east of the study area (Kasprowy Wierch, Tatra Mountains).

The transects of methane concentration over Silesian area, assisted by analysis of its stable isotopic composition has been performed in all of the locations where active and closed mining shafts are positioned, and most of the landfills and cities under the different meteorological and synoptic conditions to provide a base for efficient future verification of modelled distribution of methane sources.