



**4th International Workshop
on Uncertainty in Atmospheric Emissions**
7-9 October 2015, Krakow, Poland

PROCEEDINGS



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About the Workshop

The assessment of greenhouse gases and air pollutants (indirect GHGs) emitted to and removed from the atmosphere is high on the political and scientific agendas. Building on the UN climate process, the international community strives to address the long-term challenge of climate change collectively and comprehensively, and to take concrete and timely action that proves sustainable and robust in the future. Under the umbrella of the UN Framework Convention on Climate Change, mainly developed country parties to the Convention have, since the mid-1990s, published annual or periodic inventories of emissions and removals, and continued to do so after the Kyoto Protocol to the Convention ceased in 2012. Policymakers use these inventories to develop strategies and policies for emission reductions and to track the progress of those strategies and policies. Where formal commitments to limit emissions exist, regulatory agencies and corporations rely on emission inventories to establish compliance records.

However, as increasing international concern and cooperation aim at policy-oriented solutions to the climate change problem, a number of issues circulating around uncertainty have come to the fore, which were undervalued or left unmentioned at the time of the Kyoto Protocol but require adequate recognition under a workable and legislated successor agreement. Accounting and verification of emissions in space and time, compliance with emission reduction commitments, risk of exceeding future temperature targets, evaluating effects of mitigation versus adaptation versus intensity of induced impacts at home and elsewhere, and accounting of traded emission permits are to name but a few.

The *4th International Workshop on Uncertainty in Atmospheric Emissions* is jointly organized by the *Systems Research Institute of the Polish Academy of Sciences*, the Austrian-based *International Institute for Applied Systems Analysis*, and the *Lviv Polytechnic National University*. The 4th Uncertainty Workshop follows up and expands on the scope of the earlier Uncertainty Workshops – the *1st Workshop* in 2004 in Warsaw, Poland; the *2nd Workshop* in 2007 in Laxenburg, Austria; and the *3rd Workshop* in 2010 in Lviv, Ukraine.

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Urban metabolism: atmospheric loads and fluxes of major greenhouse gases (CO₂, CH₄) in Krakow, southern Poland

Mirosław Zimnoch¹, Jarosław Necki¹, Lukasz Chmura^{1,2}, Alina Jasek¹, Michał Galkowski¹, Tadeusz. Kuc¹, Zbigniew Gorczyca¹, Jakub Bartyzel¹, Kazimierz Rozanski¹

1 - AGH University of Science and Technology, Krakow, Poland

2 - Institute of Meteorology and Water Management, National Research Institute, IMGW-PIB
Branch of Krakow, Krakow, Poland
zimnoch@agh.edu.pl

Abstract

Emissions of carbon dioxide from fossil fuel burning constitute an important component of atmospheric carbon budget, on both global and regional scales. In urban settings across Europe anthropogenic CO₂ burden of local atmospheres is particularly visible. Methane is the second most important anthropogenic greenhouse gas. In southern Poland, apart of CH₄ emissions associated with agricultural sector and coal production, other sources of anthropogenic methane including leakages from urban gas supply networks and numerous landfills in the region are also abundant. We are presenting a brief overview of long-term investigations aimed at quantification of atmospheric loads and fluxes of CO₂ and CH₄ in Krakow agglomeration using atmospheric observations combined with ground-level measurements and modeling.

Keywords: carbon dioxide, methane, urban metabolism, greenhouse gases

1. Introduction

Urban centers play an important role in the global greenhouse gas emissions. More than 70% of anthropogenic sources of CO₂ are located in urban areas [1]. The European continent, with its dense transportation network, developed industrial infrastructure and high population density is currently responsible for more than 25 % of the global fossil fuel CO₂ emissions [2]. Urban agglomerations are also important source of methane. It has been estimated that aging natural gas distribution networks are leaking at the rate of up to 8% of the total gas consumption [3].

Here we present the results of long-term investigations aimed at quantification of atmospheric loads and fluxes of CO₂ and CH₄ in Krakow agglomeration using atmospheric observations combined with ground-level measurements and modeling. In the first part we demonstrate how measurements of atmospheric mixing ratios of carbon dioxide, combined with analyses of its carbon isotope composition (¹³C/¹²C and ¹⁴C/¹²C ratios) can be used to make partitioning of the local atmospheric CO₂ budget and quantifying the contribution of fossil fuel-derived CO₂ in the local atmosphere. In the second part, we present two independent methods of assessing surface fluxes of CO₂ and CH₄ originating from distributed sources.

Figure 1 shows schematic diagram of carbon dioxide and methane budget in the urban atmosphere. Typical for urban regions are generally elevated concentrations of CO₂ and CH₄ in the local atmosphere, when compared to remote, clean areas [4]. These local loads are caused by intense surface emissions of CO₂ and CH₄ from both point and distributed sources. When combined with diurnal variations in the intensity of vertical mixing of the lower atmosphere, they often lead to characteristic variations in

atmospheric concentrations of CO₂ and CH₄ in urban regions, with high levels of those gases recorded during the night and reduced concentrations during mid-day and afternoon hours.

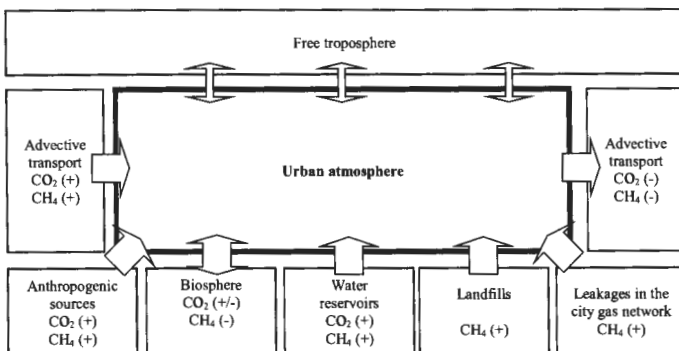


Figure 1. Schematic diagram of carbon dioxide and methane budget for urban areas. Sources and sinks of CO₂ and CH₄ are marked by '+' and '-' sign, respectively.

2. Site description

Krakow, the second largest city in Poland (approx. 800 000 inhabitants) is located in the Vistula river valley. Characteristic features of the local climate are significant percentage of calm periods (ca. 36%) and frequent temperature inversions, sometimes extending over several days. The average wind speed for the period 2005-2009 was around 3.3 m s⁻¹. West and south-west direction of surface winds prevail. Westerly circulation is generally connected with stronger winds (wind speeds above 4 m s⁻¹). Monthly air temperature at the site reveals a distinct seasonal cycle, with summer maximum (July-August) reaching ca. 20-25°C and winter minimum (January-February) between ca. -5 and 0°C.

Atmospheric measurement site (red dot on Figure 2) was situated on the University campus located in the western part of the city (50°04'N, 19°55'E, 220 m a.s.l.), bordering recreation and sports grounds. Air intake for atmospheric measurements was located on the roof of the Faculty building, ca. 20 meters above the local ground.

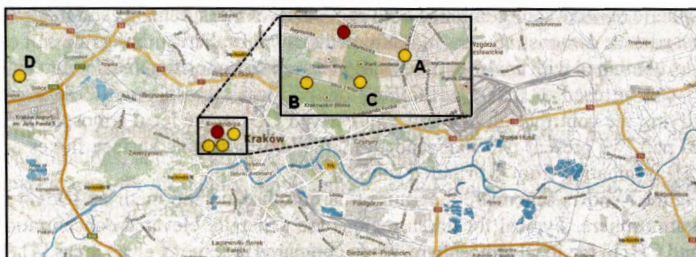


Figure 2 Location of the measurement points. Red dot represents location of atmospheric measurements, yellow points represent sites where measurements of soil fluxes of CO₂ and CH₄ were conducted.

Four sites located within the limits of Krakow agglomeration were chosen for measurements of soil fluxes of CO₂ and CH₄ (Figure 2): (i) site A (50°03'51"N, 19°55'26"E, 209 m a.s.l.) located in immediate vicinity of one of the city's major streets, (ii) site B (50°03'41"N, 19°54'08"E, 203 m a.s.l.) placed within urban meadow Blonia – 48 ha grassland recreation area in the city center with no car traffic in the direct neighborhood, (iii) site C (50°03'39"N, 19°55'08"E, 205 m a.s.l.) placed in the city park between sites A and B, and (iv) site D (50°05'24"N, 19°48'07"E, 247 m a.s.l.) located in the home garden on the outskirts of the city, in the upwind distance of ca. 11km west from other points. Site D was considered a reference site, not directly influenced by the city.

3. Methods

3.1 Atmospheric loads and partitioning of CO₂ budget

Regular measurements of atmospheric mixing ratios of CO₂ and CH₄ were performed using gas chromatograph HP6890 equipped with Shin Carbon ST column, Ni catalyst and FID detector [5]. Typical uncertainty of mixing ratio measurements was in the order of 0.1 μmol mol⁻¹ and 5 nmol mol⁻¹, for CO₂ and CH₄, respectively. In addition, during the period 2007-2009 several diurnal measurement campaigns aimed at collection of air samples for mixing ratio and isotopic analysis of CO₂ were performed. For each campaign 5 to 7 spot air samples were collected in 4-hour intervals. Carbene dioxide was cryogenically extracted from the collected air samples for further isotope analyses using IRMS and AMS techniques to determine the isotopic composition of carbon (δ¹³C and Δ¹⁴C) [6,7]. The measured ¹³C/¹²C ratios are expressed in delta notation on the VPDB scale [8], while the radiocarbon content is expressed in capital delta notation relative to Oxalic Acid standard and normalized for the year 1950 [9]. Overall uncertainty of isotope measurements was in the order of 0.2‰ for δ¹³C and 0.7 ‰ for Δ¹⁴C.

The measured atmospheric CO₂ mixing ratio at a given site (C_{meas}) consists of three components: (i) regional background component (C_{bg}) which provides the bulk of the atmospheric CO₂ load, (ii) biogenic component (C_{bio}), and (iii) fossil-fuel derived component (C_{foss}). When the carbon isotope composition of atmospheric CO₂ is measured in addition to atmospheric mixing ratio, the following mass and isotope balance equations can be formulated:

$$C_{meas} = C_{bg} + C_{bio} + C_{foss} \quad (1)$$

$$(\delta^{13}C_{meas} + 1)C_{meas} = (\delta^{13}C_{bg} + 1)C_{bg} + (\delta^{13}C_{bio} + 1)C_{bio} + (\delta^{13}C_{foss} + 1)C_{foss} \quad (2)$$

$$(\Delta^{14}C + 1)C_{meas} = (\Delta^{14}C_{bg} + 1)C_{bg} + (\Delta^{14}C_{bio} + 1)C_{bio} + (\Delta^{14}C_{foss} + 1)C_{foss} \quad (3)$$

Equations 1-3 enable partitioning of the measured CO₂ load in the local atmosphere into fossil-fuel derived, biogenic and regional background components (see [4] for details).

3.2 Direct measurements of surface fluxes

Soil CO₂ and CH₄ fluxes were measured using closed-dynamic chamber system coupled with Picarro G2101-i trace gas analyser. CO₂ concentration measurements inside the chamber were performed every second. The chamber method is a widely used

technique for measuring gas exchange between soil surface and the atmosphere [10,11,12].

The chamber system used in this work was designed and built in the Faculty of Physics and Applied Computer Science. The system consists of three stainless steel chambers of cylindrical shape, coupled with a collar hammered to the depth of ca. 5 cm into the soil and equipped with water seal to avoid gas leakages. The system was equipped with a flow regulator stabilizing the air flow through each chamber, pressure, temperature and relative humidity sensors installed in each chamber and a drying agent (magnesium perchlorate, $Mg(ClO_4)_2$) used for drying the air collected in the glass flasks for isotope analyses. In order to measure the rate of changes of CO_2 and CH_4 concentration inside the chamber, each chamber was sequentially connected to the analyzer during the measurement period. One path was equipped with flask ports allowing to take air samples for further analyses of isotopic composition of CO_2 using IRMS technique. The flux of CO_2 and CH_4 was calculated using the following expression:

$$f = \frac{p \cdot V \cdot \frac{dC}{dt}}{R \cdot T \cdot S} \quad (4)$$

where:

f - molar flux density ($mmol \ m^{-2} \ h^{-1}$),

p - atmospheric air pressure (Pa),

dC/dt - rate of concentration change of the given gas inside the chamber ($mmol \ mol^{-1} \ h^{-1}$),

R - universal gas constant ($kg \ m^2 \ s^{-2} \ mol^{-1} \ K^{-1}$),

T - ambient air temperature (K)

V and S - chamber volume (m^3) and soil surface area covered by the chamber (m^2),

3.3 Indirect assessment of surface fluxes

During the day, when thermal convection operates in the lower atmosphere, trace gases emitted from the surface are dissolved in a large volume of well-mixed layer within the Planetary Boundary Layer (PBL), leading to relatively low concentrations of those gases observed close to the ground. Inversion of vertical temperature profile in the lower atmosphere during late afternoon leads to drastic reduction of the intensity of vertical mixing and subsequent accumulation of trace gases in near-ground atmosphere.

Diurnal changes in the dynamics of vertical mixing within PBL in combination with nocturnal boundary layer (NBL) height measurements can be applied for the assessment of surface fluxes of selected trace gases. The rate of nocturnal increase of atmospheric concentrations of trace gases is controlled by the mixing layer height according to mass balance equation:

$$H \frac{d\langle C \rangle}{dt} = F_{in} - F_{out} \quad (5)$$

where:

H - height of the mixing layer

$\langle C \rangle$ - mean concentration

F_{in} - surface flux

F_{out} - flux associated with removal processes (horizontal and vertical transport). For nights with low wind speed ($< 1 \text{ m s}^{-1}$) and the adopted frequency of measurements, this term can be neglected.

During stable atmospheric conditions with low wind speeds, a distinct vertical gradient of trace gas concentration is established within the PBL. As the measurements are performed close to the surface, at the height of ca. 20 m, a correction factor relating the average increase of the concentration within the PBL ($d\langle C \rangle / dt$) to the increase of this concentration observed close to the ground is required:

$$F_{in} = \frac{H}{k} \frac{dC_{surf}}{dt} \quad (6)$$

where:

H - mixing layer height

k - correction factor

C_{surf} - concentration of the measured trace gas at the adopted measurement height.

The correction factor k was calculated using numerical simulation of vertical profiles of atmospheric ^{222}Rn using EMEP atmospheric model [13]. Radon is a natural radioactive noble gas emitted from soils. The soil radon flux is mainly controlled by concentration of ^{226}Ra and in some extent by physical properties of the soil. This fact enables the use of ^{222}Rn in the assessment of surface fluxes of other trace gases such as CO_2 and CH_4 . The NBL height H was monitored using VHS sodar (Version 3) built by the Krakow Branch of the Institute of Meteorology and Water Management. Detailed description of the sodar system can be found in [14] whereas flux assessment methodology is described in [15].

4. Results and discussion

4.1 Atmospheric loads and partitioning of CO_2 budget

Figure 3a shows an example of atmospheric loads of CO_2 and CH_4 in Krakow during the month of June 2007. Shown are daily means of the respective mixing ratios. For comparison, Figure 3a contains also daily means of CO_2 and CH_4 recorded at Kasprowy Wierch station, ca. 100 km south of Krakow, located on top of the Kasprowy Wierch mountain in the Polish Tatras (1989 m a.s.l.). This station can be considered a regional background station, free of local influences [5]. It is apparent from Figure 3a that urban atmosphere of Krakow is characterized by elevated concentrations of the measured trace gases. The excess concentrations with respect to Kasprowy Wierch can be substantial; for instance, at June 15th the daily mean CO_2 concentration in Krakow was around 462 ppm, to be compared with ca. 383 ppm at Kasprowy Wierch. Also methane revealed elevated concentration: ca. 2.33 ppm to be compared with 1.85 ppm at Kasprowy Wierch.

Figure 3b shows the results of event sampling performed on 20/21 June 2007. Shown are the values of three parameters: CO_2 mixing ratio, $\delta^{13}\text{C}$ and radiocarbon content of CO_2 . Distinct diurnal variability of CO_2 mixing ratios is observed, with the maximum (417 ppm) recorded late in the night and minimum (391 ppm) in the afternoon. Initial increase of CO_2 mixing ratio is accompanied by drop of $\delta^{13}\text{C}$ and increase of $\Delta^{14}\text{C}$. Later on, during the day 21th June significantly lower values of $\Delta^{14}\text{C}$ are recorded.

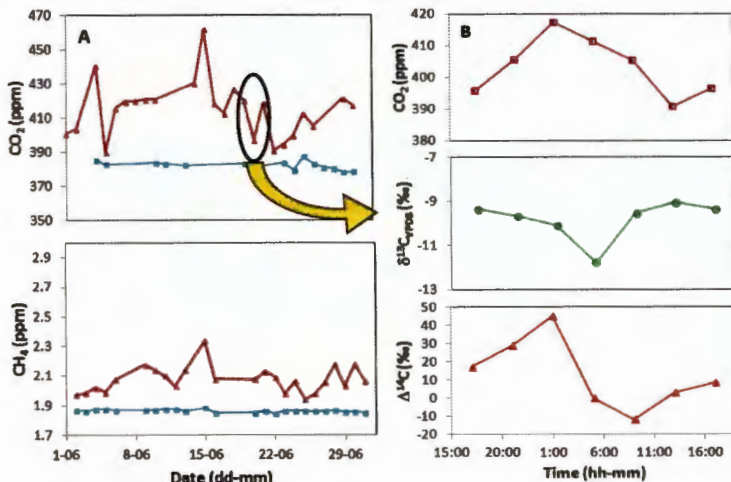


Figure 3. (A) Daily means of CO₂ and CH₄ mixing ratios recorded at Krakow and Kasprowy Wierch stations during June 2007. (B) concentration and carbon isotope composition of atmospheric CO₂ recorded during measurement campaign carried out between 20 and 21 June 2007 in Krakow.

The data shown in Figure 3b were used to calculate the budget of CO₂ in the local atmosphere of Krakow during 20/21 June 2007. Equations 1-3 were used to derive temporal evolution of individual components of this budget (fossil-fuel derived, biogenic and regional CO₂ background component). The results are presented in Figure 4. The calculated contributions indicate a dominant role of biogenic CO₂ emissions during night hours. During day time anthropogenic emissions dominate. The calculated background mixing ratios of CO₂ are in good agreement with the measurements performed at this time period at Kasprowy Wierch station [4].

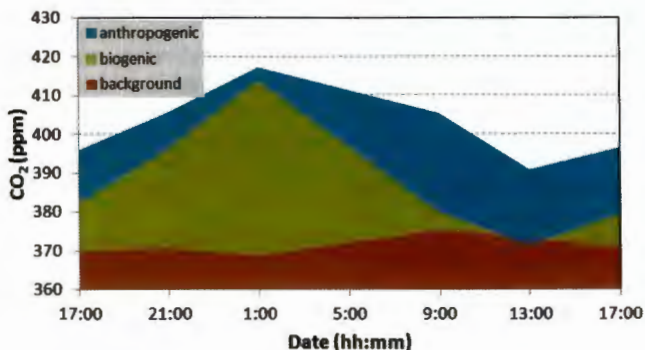


Figure 4. Partitioning of atmospheric CO₂ load over Krakow during 20/21 June 2007 into fossil-fuel derived, biogenic and regional CO₂ background component.

4.2 Direct measurements of surface fluxes

Figure 5 summarizes soil CO₂ fluxes measured at four different sites in Krakow between July 2009 and June 2013. All investigated sites reveal a strong seasonality of CO₂ emissions induced by seasonal vegetation cycle and seasonal variations of soil parameters (temperature, water content).

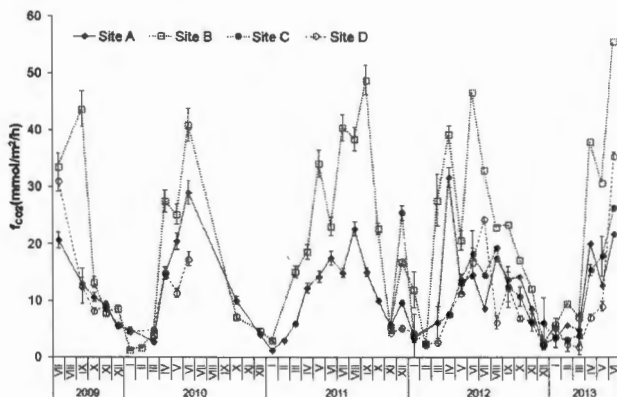


Figure 5. Seasonal variations of soil CO₂ fluxes measured at four selected ecosystem sites in Krakow agglomeration (A-D - cf. Figure 2), differing in degree of anthropogenic influence.

Minimum values of the CO₂ fluxes were recorded during winter months (December, January and February). Low soil temperatures limit respiration activity, while higher water contents reduce permeability of the soil during this season. Snow cover, if present, may further reduce transport of CO₂ between the soil and the atmosphere during winter months. Typical CO₂ fluxes measured during winter varied between approximately 1 and 5 mmol m⁻² h⁻¹ for all sites. During the vegetation period (April to October) the CO₂ fluxes increase significantly, reaching maximum values of 25-30 mmol m⁻² h⁻¹ at site A, C and D and 40-50 mmol m⁻² h⁻¹ at site B. Timing of the CO₂ flux maxima varies from year to year. The mean value of soil CO₂ flux calculated for entire observation period and all four sites reaches 16.2 mmol m⁻² h⁻¹ and is comparable to the mean local fossil fuel CO₂ flux, which is approximately 17.8 mmol m⁻² h⁻¹ (2008) [2]. Measurements of methane fluxes revealed small negative numbers (between -1 and -2 μmol m⁻² h⁻¹ for all urban sites and close to zero for the reference site D) indicating that soils in the study area are a weak sink of methane. This fact suggests that apparent source of methane leading to night-time maxima frequently observed in atmospheric CH₄ record available for Krakow station, has entirely anthropogenic origin.

4.3 Indirect assessment of surface fluxes

Calculations of surface night-time fluxes of CO₂ and CH₄ based on simplified atmospheric budget of these gases (cf. section 3.3) were performed for the period from May 2005 to May 2009, for nights with wind speed lower than 1m s⁻¹ (for details see [15]). The calculated mean monthly surface fluxes of CO₂ (Figure 6) reveal distinct

seasonality, with a minimum of ca $2 \text{ mmol m}^{-2} \text{ h}^{-1}$ occurring during winter time (December to March) and a maximum of ca. $20 \text{ mmol m}^{-2} \text{ h}^{-1}$ in summer (July and August). This seasonality is also seen in direct measurements of soil CO_2 fluxes (cf. Figure 5) and is modulated by the biospheric component, originating from soil respiration. Relative uncertainty of the monthly mean CO_2 fluxes derived from simplified atmospheric budget varies between ca. 1 and 87%.

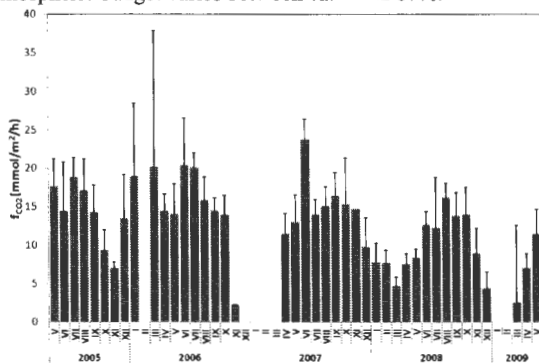


Figure 6. Monthly means of surface-averaged CO_2 flux for Krakow city derived from the atmospheric balance method.

Contrary to the CO_2 flux, the surface fluxes of CH_4 (Figure 7) do not reveal any distinct seasonality and, in contrast to chamber measurements, are distinctly positive. The monthly means of the calculated CH_4 flux scatter between ca. 50 and $200 \mu\text{mol m}^{-2} \text{ h}^{-1}$, with relative uncertainty of the mean values varying between 10 and 227%. The mean CH_4 flux calculated for the whole analyzed period (May 2005 – December 2008) is equal $97.2 \pm 5.4 \mu\text{mol m}^{-2} \text{ h}^{-1}$. The quoted uncertainty represents one standard deviation of the mean value. Multiplying the mean CH_4 flux by the surface area of the city (326.8 km^2), leads to the total mean annual emission of methane into the atmosphere in Krakow in the order of $(6.2 \pm 0.4) \times 10^6 \text{ m}^3 \text{ yr}^{-1}$. Leakages of the city gas network are thought to be the main source of this methane.

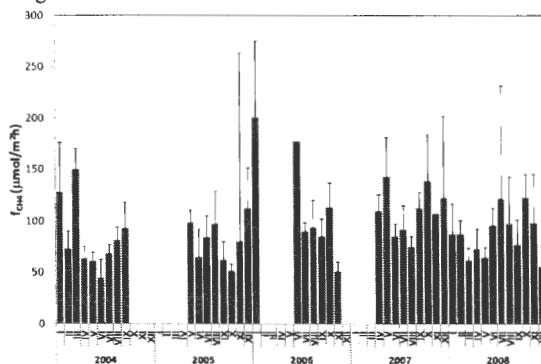


Figure 7. Monthly means of surface-averaged CH_4 flux for Krakow derived from the atmospheric balance method.

5. Summary

Urban centers are important elements of the global carbon cycle. Their importance will likely increase in the foreseeable future, with the continuing transformation of the world population from rural to urban. It is therefore important to gain a deeper understanding of carbon cycling in the urban environment in order to design appropriate strategies to control emissions of major greenhouse gases into the atmosphere.

An overview of long-term investigations aimed at quantification of atmospheric loads and fluxes of CO₂ and CH₄ in Krakow agglomeration using atmospheric observations combined with ground-level measurements and modeling, presented above, reveals the potential of such approach for quantifying atmospheric loads and surface fluxes of major greenhouse gases in complex urban environment, thus constituting an independent verification tool for greenhouse gas emissions reported in the framework of international agreements by signatory countries.

Acknowledgements

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