

# **IMPROVING THE GREENHOUSE GAS BUDGET ESTIMATION FOR HUNGARY USING TALL TOWER OBSERVATIONS**

(OTKA K129118/K141839)

Summary report

Principal investigator: László Haszpra

## **Introduction**

The determination of the atmospheric greenhouse gases (GHG) budget and the clarification of the contributing processes are essential for the prediction of the future climate and the mitigation of climate change. Due to the obligations arising from the international conventions, the precise determination of the national GHG budgets, verified by measurements, is becoming increasingly important as well. The project aimed to contribute to a better understanding of the atmospheric GHG budgets and to improve the GHG emission estimates on local, regional, national, and continental scales. For this purpose, the monitoring infrastructure, which had been constructed partly using a former OTKA grant (K109764), was used.

## **Difficulties and changes during the project**

The project (K129118) was granted to the Hungarian Meteorological Service (HMS), however, the law prohibited the principal investigator from continuing this work at HMS, a government office, beyond the official retirement age. Therefore, the project was transferred to the Geodetic and Geophysical Institute (GGI) of the Research Centre of Astronomy and Earth Sciences (RCAES). When GGI left RCAES in 2021, the project was inherited by the Institute for Earth Physics and Space Sciences and received a new project identification number K141839. For this reason, the publications produced by this project wear the identification number either K129118 or K141839. The organizations followed different financial rules, which upset the original budget plan; the changes temporarily slowed down the administrative processes but did not significantly affect the research work.

The project originally planned to hire a young scientist (PhD student or postdoc) but no interested person was found. This led to a certain temporal redistribution of the tasks relative to the original work plan.

Although the COVID-19 pandemic caused serious difficulties, the maintenance of the measurements could be succeeded. However, the dissemination of the results at scientific conferences was hindered by travel restrictions.

## **Results**

### *Measurements*

The study of greenhouse gas emissions, the focus of this research topic, can only be based on reliable atmospheric measurements. The project involved continuous, high-precision measurements of carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>), nitrous oxide (N<sub>2</sub>O), and carbon monoxide (CO) concentrations, as well as surface-atmosphere fluxes of CO<sub>2</sub>, N<sub>2</sub>O and CO at the tall-tower monitoring site at Hegyhátsál. The measurement program was complemented by methane stable-isotope measurements. It was found that different measurement protocols have detectable effects on the input data of the emission calculation models.

In most cases, several intakes are installed along the tall towers to give information also on the vertical concentration profile of the components considered. Typically, a single gas analyzer is used, and the intake points are sequentially connected to the instrument. It involves that the continuous concentration signal is sampled only for discrete short periods at each intake point, which does not allow for a perfect reconstruction of the original concentration variation. This increases the uncertainty of the calculated hourly averages, which are typically used by the atmospheric transport and budget models for emission estimation. The purpose of the study was to give the data users an impression of the potential magnitude of this kind of uncertainty, as well as how it depends on the length of the sampling period at each intake. Based on the study we suggested increasing the sampling frequency as much as technically feasible to provide measurement data for emission estimation with the lowest possible uncertainties. The results were first presented to the European “tall-tower” scientific community and finally published in *Atmospheric Measurement Technique*, a Q1 category scientific journal (Haszpra and Prácser, 2020; 2021).

CO<sub>2</sub> concentration measurements started at Hegyhátsál in 1994. By the end of the present project, a 30-year-long data series was available, which already allowed detailed statistical analyses. Within the general upward trend of 2.17  $\mu\text{mol mol}^{-1} \text{ year}^{-1}$ , we detected an above-average growth rate in the summer nighttime concentrations presumably caused by the significant warming trend during the measurement period. The atmospheric CO<sub>2</sub> concentration shows a remarkable seasonal cycle with winter maximum and summer minimum due to the cyclic activity of the vegetation. The length of the summer season, when the CO<sub>2</sub> concentration is negative relative to the annual average was increasing between 1994 and 2023. Defining its beginning by the day of the year when the CO<sub>2</sub> concentration becomes negative relative to the annual average (spring zero-crossing), the beginning of the summer season is getting earlier by  $-0.38 \pm 0.16 \text{ day year}^{-1}$ . Although this linear trend is statistically significant at a reasonable level of probability ( $p=0.024$ ), the data series shows two distinct periods. Until the end of the 2000s the advance was fast ( $-0.96 \pm 0.41 \text{ day year}^{-1}$ ;  $p=0.033$ ) followed by stagnation  $0.08 \pm 0.32 \text{ day year}^{-1}$ ;  $p=0.81$ ). The earlier timing of the spring zero-crossing indicates the earlier start of the growing season. It seems that the spring onset of leaf unfolding used to be temperature-limited has become light-limited in the represented region by now. Although the temperature has been steadily increasing, the available light limits the further advancement of the beginning of the growing season. The fluctuation in the growth rate of the CO<sub>2</sub> concentration is much wider than that in the anthropogenic emission. This is caused by the interannual variation in biospheric activity affected by global processes. The growth rate at Hegyhátsál shows a fairly strong correlation ( $r=0.44$ ) with the El Niño – Southern Oscillation phenomenon with a 6-7 months lag time indicating the global nature of the biospheric effects. The partial results, combined with the measurements carried out at K-pusztá between 1981 and 1999 were presented at two conferences (Haszpra, 2022; 2023). The detailed study focusing only on the high-precision measurements at Hegyhátsál has been submitted to the Q1 category journal, *Atmospheric Measurement Techniques* (Haszpra, 2024). The manuscript is still under review. We evaluated the three-year-long atmospheric methane mole fraction and  $\delta^{13}\text{C}_{\text{CH}_4}$  isotope ratio measurement campaigns, which were performed at Hegyhátsál. The results were compared with the data reported by two NOAA atmospheric monitoring sites (Mace Head, Ireland, and Zeppelin, Swabard) to determine the continental methane excess and the relative isotopic shift. For the interpretation of the similarities and differences, atmospheric back-trajectory analysis was used. The Hungarian monitoring station can be separated from the coastal and polar areas based on the mole fraction results having higher maxima and seasonal amplitude, but the  $\delta^{13}\text{C}_{\text{CH}_4}$  results match well with the NOAA stations' results. The mean CH<sub>4</sub> mole fraction value

is about 80 ppb higher than the comparable marine background, and values above 2,000 ppb were also frequently observed. The mean  $\delta^{13}\text{C}_{\text{CH}_4}$  value of  $-47.5 \pm 0.3\text{‰}$  ( $2\sigma$ ) was comparable to values at all three monitoring sites, but specific pollution events were detected at Hegyhátsál. Concentration-weighted trajectory modeling, meteorological parameters, stable carbon isotopic composition ( $\delta^{13}\text{C}_{\text{CH}_4}$ ), and Miller-Tans analysis show that the main factors influencing  $\text{CH}_4$  at Hegyhátsál, apart from diurnal and seasonal changes in the planetary boundary layer, are emissions from residential heating and industrial  $\text{CH}_4$  emissions during the winter. The results were published in *Journal of Geophysical Research: Atmosphere*, a D1 category scientific journal (Varga et al., 2021).

### *Emission calculations*

From time to time stagnant weather conditions can form over the Pannonian/Carpathian Basin in winter. Cold air piles up in the Basin preventing the vertical mixing of the atmosphere. During a cold-air pool episode emissions from the surface accumulate in the boundary layer of the atmosphere for days gradually increasing the atmospheric concentration. Taking advantage of three cold-air pool episodes in January-February 2017, and the  $\text{CO}_2$  concentration measurements on the tower, we determined the average  $\text{CO}_2$  emissions of the tens of thousands of  $\text{km}^2$  area represented by the monitoring site using a simple boundary layer budget model. The model gave a carbon dioxide emission of  $6.3 \text{ g m}^{-2} \text{ day}^{-1}$  for the footprint area of the measurements on average for the period of the episodes. The  $6.7\text{--}13.8 \text{ nmol } \mu\text{mol}^{-1}$ ,  $0.15\text{--}0.31 \text{ nmol } \mu\text{mol}^{-1}$  and  $15.0\text{--}25.8 \text{ nmol } \mu\text{mol}^{-1}$  concentration ratios for  $\text{CH}_4\text{:CO}_2$ ,  $\text{N}_2\text{O:CO}_2$  and  $\text{CO:CO}_2$ , respectively, correspond to  $15.3\text{--}31.7 \text{ mg m}^{-2} \text{ day}^{-1}$  methane,  $0.9\text{--}2.0 \text{ mg m}^{-2} \text{ day}^{-1}$  nitrous oxide and  $60.0\text{--}103.4 \text{ mg m}^{-2} \text{ day}^{-1}$  carbon monoxide emissions for the region. The study, which we published in *Environmental Sciences Europe* (Haszpra et al., 2019), a D1 category scientific journal, indicated the high share of biomass burning in residential heating in rural environments that results in high carbon monoxide emissions relative to those of carbon dioxide. It also indicated that the actual emission factor for nitrous oxide may exceed the range given in the inventory compilation guidelines, which should be taken into account in reporting. The study called attention to the winter residential heating. As a continuation of the work, we specifically determined the emissions of Hegyhátsál village located close to the monitoring tower. For the estimation of the emissions, we used a footprint model developed for the identification of the source areas, and the direct surface-atmosphere flux measurements

available from the tower. The recorded emission densities, dominantly resulting from residential heating, are 3.5, 0.043, and 72  $\mu\text{g m}^{-2} \text{s}^{-1}$  for CO, N<sub>2</sub>O, and CO<sub>2</sub>, respectively. While the measured CO and CO<sub>2</sub> emissions are comparable to those calculated using the assumed energy consumption and applying the according emission factors, this study also showed that the nitrous oxide emissions exceed the “officially” expected value by an order of magnitude. This may indicate that the nitrous oxide emissions are significantly underestimated in the emission inventories, and modifications in the methodology of emission calculations are necessary. The study was published in *Atmospheric Measurement Techniques* (Haszpra et al., 2022), a Q1 category scientific journal, and the results were communicated with the National Emission Inventory Group of Hungary. The compilation of the national GHG inventories has to follow a strict international guideline, therefore our findings cannot change the methodology immediately. However, several similar research projects are ongoing in Europe, which may lead to the modification of the guidelines.

The tall-tower CO<sub>2</sub> surface-atmosphere exchange measurements also allowed the estimation of the CO<sub>2</sub> exchange of a typical agricultural region surrounding the tower. The measurements were started in 1997, so the long data series also allowed trend estimations. Long-term, the mean net ecosystem exchange (NEE) of -170  $\text{gC m}^{-2} \text{year}^{-1}$  revealed that the complex agricultural region was a net carbon sink from the atmospheric perspective. Trend analysis indicates that NEE decreased (i.e. became more negative, which means a stronger sink) by 12  $\text{gC m}^{-2} \text{year}^{-1}$ , which may be explained by improved agrotechnology and climate change. Net biome production (NBP) was estimated using crop census data and assumptions about the management practices that affect lateral carbon flux. The long-term mean NBP was -30  $\text{gC m}^{-2} \text{year}^{-1}$ , which indicates that the soils may be losing carbon, though this loss is within the range of uncertainty of the measurements ( $\sim 50 \text{ gC m}^{-2} \text{year}^{-1}$ ). Our results also indicated that environmental conditions in spring have a major role in the annual carbon balance in this region. The study based on a uniquely long data series was published in *Agricultural and Forest Meteorology* (Barcza et al., 2020), a D1 category scientific journal.

### *Contribution to international emission estimations*

It was hypothesized for a long time that the increasingly frequent severe droughts strongly reduce the CO<sub>2</sub> uptake by vegetation. In addition to scattered studies from different countries, it was also indicated by our local studies at Hegyhátsál (Barcza et al., 2020). Persistent droughts

are spatially extended phenomena, which should also be reflected in the concentration measurements. First, we studied it in Polish-Hungarian cooperation using the measurements at Kasprowy Wierch (KAS) and Hegyhátsál (HUN). The results were submitted to *Atmospheric Chemistry and Physics Discussion* (Chumura et al., 2020), which is the preprint version of the D1 category *Atmospheric Chemistry and Physics*. We presented evidence of an increased impact of droughts on the annual cycle of carbon dioxide over Central-Eastern Europe, based on long-term observations (1995-2018) of mixing ratios conducted at the two sites. Analyses of the smoothed, detrended annual cycles from both sites revealed a gradual reduction of annual amplitudes towards lower magnitudes, with simultaneous reductions of annual maxima (KAS:  $-0.13 \pm 0.05$  ppm year<sup>-1</sup>, HUN:  $-0.08 \pm 0.12$  ppm year<sup>-1</sup>) and increases of minima (KAS:  $0.09 \pm 0.04$  ppm year<sup>-1</sup>, HUN:  $0.08 \pm 0.08$  ppm year<sup>-1</sup>). The study proved that the spatial scale chosen was too small to achieve solid, statistically significant results. Therefore the Hungarian results were divided into two parts: the analysis of the temporal variation of the CO<sub>2</sub> concentration was included in the evaluation of the multi-decadal data series mentioned before (Haszpra, 2024), while the drought-oriented results were incorporated into a pan-European drought study performed by a large research group including us and most of the main contributors of the above mentioned Polish works. The study was published in Series B of *Philosophical Transactions*, a D1 category journal of the Royal Society (Ramonet et al., 2020). The main finding of the study was that the usual summer minimum in CO<sub>2</sub> due to the surface carbon uptake was reduced as much as by 1.4 ppm in 2018 in the territory of Europe most affected by the drought, which means a significantly reduced carbon uptake by the vegetation.

By combining atmospheric concentration measurements with atmospheric transport models the location of the sources of the substance studied and their yields can be determined. This type of the so-called inverse transport models has been intensively developed during the past decade, because they may be the main tool for the determination of national emissions in the near future. We contributed to a project led by Japanese scientists developing a high-resolution ( $0.1^\circ \times 0.1^\circ$ ) inverse model for the determination of CO<sub>2</sub> emissions. The model was published in *Atmospheric Chemistry and Physics*, a D1 category journal of the European Geophysical Union (Maksyutov et al., 2021). Due to the high number of coauthors and supporting projects, the funding organizations could not be listed in the paper. However, the participating monitoring stations are listed, and the persons contributing substantially to the study can be found among the co-authors.

## Publication activity

The project resulted in five D1/Q1 and two Q1 publications, of which one D1/Q1 does not explicitly mention the project. One additional potential Q1 publication is still pending; the manuscript is under review.

Climate change caused primarily by the anthropogenic emissions of greenhouse gases, which were the focus of the present project, is one of the biggest challenges humankind faces. Successful mitigation and adaptation require that everybody, regardless of educational background, understands the processes, the actions, and the consequences. Scientific educational papers and lectures play an essential role in this kind of knowledge dissemination. Such papers, internet publications, and lectures usually cannot explicitly refer to the OTKA's support, therefore OTKA does not appreciate this activity. The final report of the present project lists only seven such publications (*Magyar Tudomány, Léggör, websites*) that are the most directly related to the OTKA topic. These publications mainly focus on the importance of greenhouse gas measurements in climate management, and on methane, which has a growing importance in climate change mitigation efforts.

## References

- Barcza, Z., Kern, A., Davis, K. J., Haszpra, L., 2020. Analysis of the 21-years long carbon flux dataset from a Central European tall tower site. *Agricultural and Forest Meteorology* 290, 108027. doi: 10.1016/j.agrformet.2020.108027. **D1**
- Chmura, L., Galkowski, M., Sekula, P., Zimnoch, M., Necki, J., Bartyzel, J., Zieba, D., Rozanski, K., Wolkowicz, W., Dlugokencky, E., Haszpra, L., 2020. Signs of reduced biospheric activity with progressing global warming: evidence from long-term records of atmospheric CO<sub>2</sub> mixing ratios in Central-Eastern Europe. *Atmospheric Physics and Chemistry Discussion*, 10.5194/acp-2019-748
- Haszpra L., 2022: 41 years of atmospheric carbon dioxide monitoring in Hungary. *21th WMO/IAEA Meeting on Carbon Dioxide, Other Greenhouse Gases and Related Measurement Techniques (GGMT-2022)*, Wageningen, the Netherlands, 19-21 September, 2022. <https://www.ggmt2022.online/wp-content/uploads/ggmt2022/posters/Haszpra-poster84.pdf>

- Haszpra, L., 2023: Four decades of atmospheric CO<sub>2</sub> measurements in Hungary. *51th Global Monitoring Annual Conference*, Boulder, Colorado, U.S.A., 23-24 May 2023. <https://gml.noaa.gov/annualconference/abs.php?refnum=13-230418-B>
- Haszpra, L., 2024: Multi-decadal atmospheric carbon dioxide measurements in Central Europe, Hungary. *Atmospheric Measurement Techniques* (under review)
- Haszpra, L. and Prácser, E., 2020: Technical note on tall-tower measurements. *ICOS Science Conference*, Utrecht, 15-17 September, 2020.
- Haszpra, L. and Prácser, E., 2021: Uncertainty of the hourly average concentration values derived from non-continuous measurements. *Atmospheric Measurement Techniques* 14, 3561–3571, <https://doi.org/10.5194/amt-14-3561-2021>. **Q1**
- Haszpra, L., Ferenczi, Z., Barcza, Z., 2019. Estimation of greenhouse gas emission factors based on observed covariance of CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>O and CO mole fractions. *Environmental Sciences Europe* 31, 95. doi: 10.1186/s12302-019-0277-y. **D1**
- Haszpra, L., Barcza, Z., Ferenczi, Z., Kern, A., Kljun, N., 2022: Real-world wintertime CO, N<sub>2</sub>O and CO<sub>2</sub> emissions of a Central European village. *Atmospheric Measurement Techniques* 15, 5019–5031, <https://doi.org/10.5194/amt-15-5019-2022>. **Q1**
- Maksyutov, S., Oda, T., Saito, M., Janardanan, R., Belikov, D., Kaiser, J. W., Zhuravlev, R., Ganshin, A., Valsala, V. K., Andrews, A., Chmura, L., Dlugokencky, E., Haszpra, L., Langenfelds, R. L., Machida, T., Nakazawa, T., Ramonet, M., Sweeney, C., Worthy, D., 2021: Technical note: A high-resolution inverse modelling technique for estimating surface CO<sub>2</sub> fluxes based on the NIES-TM – FLEXPART coupled transport model and its adjoint. *Atmospheric Chemistry and Physics* 21, 1245-1266. <https://doi.org/10.5194/acp-2020-251>. **D1**
- Ramonet, M., Ciais, P., Apadula, F., Bartyzal, J., Bastos, A., Bergamaschi, P., Blanc, P. E., Brunner, D., Caracciolo di Torchiariolo, L., Calzolari, F., Chen, H., Chmura, L., Colomb, A., Conil, S., Cristofanelli, P., Cuevas, E., Curcoll, R., Delmotte, M., di Sarra, A., Emmenegger, L., Forster, G., Frumau, A., Gerbig, C., Gheusi, F., Hammer, S., Haszpra, L., Hatakka, J., Hazan, L., Heliasz, M., Henne, S., Hensen, A., Hermansen, O., Keronen, P., Kivi, R., Komínková, K., Kubistin, D., Laurent, O., Laurila, T., Lavric, J. V., Lehner, I., Lehtinen, K. E. J., Leskinen, A., Leuenberger, M., Levin, I., Lindauer, M., Lopez, M., Lund Myhre, C., Mammarella, I., Manca, G., Manning, A., Marek, M. V., Marklund, P., Martin, D., Meinhardt, F., Mihalopoulos, N., Mölder, M., Morgui, J. A., Necki, J., O'Doherty, S., O'Dowd, C., Ottosson, M., Philippon, C., Piacentino, S., Pichon, J. M., Plass-Duelmer, C., Resovsky, A., Rivier, L., Rodó, X., Sha, M. K.,



Scheeren, H. A., Sferlazzo, D., Spain, T. G., Stanley, K. M., Steinbacher, M., Trisolino, P., Vermeulen, A., Vítková, G., Weyrauch, D., Xueref-Remy, I., Yala, K., Yver Kwok, C., 2020: The fingerprint of the summer 2018 drought in Europe on ground-based atmospheric CO<sub>2</sub> measurements. *Philosophical Transactions B*. 375, 20190513. doi:10.1098/rstb.2019.0513. **D1**

Varga, T., Fisher, R. E., France, J. L., Haszpra, L., Jull, A. J. T., Lowry, D., Major, I., Molnár, M., Nisbet, E. G., László, E., 2021: Identification of potential methane source regions in Europe using  $\delta^{13}\text{CCH}_4$  measurements and trajectory modeling. *J. of Geophysical Research: Atmospheres* 126, e2020JD033963. <https://doi.org/10.1029/2020JD033963>. **D1**