

Final report

Determination of the atmospheric transport, concentration and deposition of radionuclides and particulate matter by in situ measurements and model simulations

This research project is a joint project in the cooperation between
(1) Department of Meteorology, Eötvös Loránd University (ELU), and
(2) Institute of Radiochemistry and Radioecology, University of Pannonia (UP)

Background

An increased interest of air pollution monitoring and modelling, as well as the analyses of the various effects of air pollution on the environment has been observed in the last few decades. As more accidental (nuclear and industrial) and natural emission events (volcano's eruptions, forest fires etc.) have occurred in different parts of the world, there is an increased demand both on the part of scientific society and population for the understanding and effective prediction of the environmental, social or economical effects of possible accidental releases and other atmospheric environmental problems.

Fast and accurate estimations of air pollutant dispersion by state-of-the-art models may reduce the harmful effects of an environmental problem or an accidental release. Based on the results of sophisticated model calculations, the decision makers could make important arrangements, which can protect human health, or even save human lives. At the same time, these models could also be appropriate for further analyses of atmospheric transport processes. Therefore, continuous developments of dispersion models and widespread investigations with them are essential.

Two of the most important components of air pollution are particulate matter and radionuclides. Elevated concentrations of particulate matter – emitted mainly from transport and residential heating – can cause several health problems. The other serious impact for the environment and human health is caused by different radionuclides. Radionuclides, if released to the environment, may reach the human body through several transfer pathways and the soil-plant-animal pathway is considered as one of the important routes through which radionuclides can enter the human body.

The main aims of this project were to analyse the atmospheric transport processes, concentration fields and deposition mechanisms of radionuclides and particulate matter emitted to the atmosphere by various continuous and accidental sources by high spatiotemporal resolution in situ measurements and state-of-the-art model simulations.

Project tasks

The activities in the project can be divided into 4 main sections.

1. Definition and sensitivity analyses of input parameters for dispersion and food chain models

A literature survey was carried out to assess the available data on soil-plant, plant-animal and food-human transfer factors, including numerous domestic and international publications. It has been identified that in case of Sr-90 and Cs-137 there are orders of magnitude differences in the reported values between specific publications. This made it necessary to use location-specific data at the given site. During the design of measurement procedures for Sr-90 and Cs-137 in different matrixes (soil, plant and samples of animal origin) using beta- and gamma-

spectrometry methods, the preliminary experiments revealed that in case of these two artificial radionuclides, the direct classical methods for measuring radioactivity (beta- and gamma-spectrometry) do not have the detection limits required for the reliable determination of the radionuclide concentrations present in the samples. Accordingly, the stable isotopes of the radionuclides' elements (Sr-84 and Cs-133) were used in the measurements required to determine the transfer factors. Samples were taken from different geographical points such as Japan, Vietnam, Kazakhstan and Hungary.

2. Air quality analyses by different models and measurements

During the project, air quality analysis was carried out using various model simulations and processing of measured data.

Due to increasing issues of air pollution in urban areas, continuous research is being conducted to upgrade models, which can predict the distribution of pollutants and thus enable timely interventions to mitigate their negative effects. To support these efforts, traffic data from an integrated transport model was used to drive the COPERT traffic emission model and the WRF-Chem atmospheric chemistry model. With reliable macroscopic traffic data from the Budapest region, traffic state estimations were calculated for every fifteen minutes of the day using dynamic assignment with predefined and time-varying static demand matrices.

A downscaling method using 24-hour forecasts from 9 independent air quality models of the Copernicus Atmosphere Monitoring Service (CAMS) was introduced to improve PM_{2.5} predictions in Budapest in the heating season.

PM_{2.5} concentrations were detected with high temporal resolution at different sites (downtown and suburb region of Budapest). DustTrak TSI 8532 monitors, and IQAir - AirVisual Pro Air Quality Monitors were used for this investigation, whereby both indoor and outdoor air quality were analysed.

Due to the pandemic situation associated with the Covid-19 virus, we have expanded our research to examine the effects of reduced traffic emissions on urban air quality related to the decreasing mobility in the spring of 2020.

3. Development of a new coupled dispersion and a food chain model

In the recent period, our eating habits and the quality of the food we eat have changed significantly. One reason for this is globalization, which allows us to deliver any food anywhere in the world. The other reason is that the quality of food has changed, we also consume other types of food, and the ratios between each type of food have changed. For this reason, it is also necessary to modify the parameters in food-chain models (type and amount of food, seasonality etc.). We examined long-term statistical analyses of the eating habits and used them to determine a list of the most important and sensitive foods. In addition to food, we also investigated which part of the population is most affected by the change, which are the most sensitive groups. The food-chain model was coupled with a Lagrangian atmospheric dispersion model to simulate advection and turbulent diffusion using the puff modeling approach. Meteorological data for dispersion simulations was obtained from GFS and AROME numerical weather prediction models run by NOAA and Hungarian Meteorological Service, respectively.

4. Mobile measurements of air pollutants

During different periods and in different weather conditions, measurement campaigns were carried out to map the average particulate matter (PM_{2.5}, PM₁₀) exposure on cycling lanes in the downtown of Budapest. Measurements were made with DustTrak TSI 8532 portable aerosol

monitoring systems with high temporal and spatial resolution. Our investigation focused on the spatial variation of particulate matter in different urban environments, such as street canyons, urban green areas or high- and low traffic roads and bridges across the Danube River. The accuracy of the instruments was also tested against measurements from reference instruments during a calibration campaign.

The summary of our measurements and model simulations during the project can be found in Table 1. and 2, respectively.

Table 1. Measuring activities during the project

Measured values	Measurement method	Project results
Sr-90 activity concentration in different matrixes	LSC (Liquid scintillation technique)	Baigazinov et al., 2020 Dung et al., 2021
Cs-137 activity concentration in different matrixes	HPGe gamma-spectrometry	Baigazinov et al., 2020 Mamyrbayeva et al., 2020 Dung et al., 2020 Dung et al., 2021 Mamyrbayeva et al., 2021
Particulate matter (PM2.5, PM10) concentration	TSI DustTrakII Aerosol monitor 8532	Csapó és Mészáros, 2019 Mészáros, 2022 Tordai és Mészáros, 2023
Particulate matter (PM2.5) concentration	AirVisual Pro monitors	Bushra and Mészáros, 2019a,b Bushra et al., 2020 Mészáros, 2022
Microclimatological characteristics in different urban areas	Testo 635-2 sensors	Tordai és Mészáros, 2023
Particulate matter (PM2.5) concentration	AirVisual Pro monitors	Bushra and Mészáros, 2019a,b Bushra et al., 2020 Mészáros, 2022
Particulate matter (PM2.5, PM10) concentration	Database from Hungarian Air Quality Network	Varga-Balogh et al., 2021

Table 2. Modelling activities during the project

Modelling activity	Applied method	Project results
PM2.5 prediction in Budapest	CAMS (Copernicus Atmospheric Monitoring Service) models (9 models)	Varga-Balogh et al., 2020a Varga-Balogh et al., 2022 Leelőssy et al., 2022
PM10 prediction in Budapest	Data fusion method, CAMS model	Varga-Balogh et al., 2020b
Urban air quality forecast	WRF-Chem model	Szanyi, 2020 (thesis) Kovács et al., 2019a Kovács et al., 2021
Analysing the effects of different emission sources on air pollution	SHERPA model	Horváth et al., 2023
Modelling the dispersion of radionuclides	Newly developed combined transport and food-chain model	Socio-economic application: a simulation model for Paks NPP2

Summary of the results

1. Definition and sensitivity analyses of input parameters for dispersion and food chain models

It was found that the active Cs deposition on the ground is present in the form of granular non-ionic materials that are not adsorbed or trapped by minerals in the soil, but instead physically adhere to the rough surfaces of the soil mineral particles. Granular Cs can be transferred among media, such as soils and plants. The physical properties and dynamic behaviour of the granular Cs is expected to be helpful in considering methods for decontamination of soil, litter and other media. Our results can serve as initial data for modelling the transport/deposition of radionuclides. Since the granular Cs is not soluble in water, and it does not become ionic Cs, it cannot be trapped into soil minerals, but physically adheres to the rough surfaces of soil mineral particles. This means that the ^{134}Cs and ^{137}Cs in the contaminated soils would not be available to be adsorbed into soils. Therefore, it can be transferred among media, such as soils and plants (Mamyrbayeva et al., 2020).

Biotransfer-factors (^{90}Sr , ^{137}Cs) were determined based on the measurement results. Due to the comparatively high detection limits of the classical radiochemical methods and measurement techniques, neutron activation analysis (NAA) was used to determine strontium and caesium transfer factors. All samples were dried and ashed during the preparation for the NAA measurements. In the NAA measurement, the gamma radiation from the $^{84}\text{Sr}(n,\gamma)^{85}\text{Sr}$ reaction and the $^{133}\text{Cs}(n,\gamma)^{134}\text{Cs}$ reaction was used to determine the strontium and caesium concentrations, respectively. After determining the stable isotope concentrations, the radioactive isotope concentrations were calculated, followed by determining the location specific transfer factors (Baigazinov, et al., 2020).

The results were compared with the data found in the international literature. The comparison has revealed that in modelling, the input parameters are of crucial importance, since the biotransfer-factor of a specific radionuclide is greatly dependent on the location-specific parameters (soil type, weather etc.) (Mamyrbayeva et al., 2021).

Analysis of the observed results was carried out to prepare for using in the modelling of the dispersion of continuous and accidental emissions to estimate the annual exposure effecting the population. Deposition processes in the modelling studies, particularly for in-cloud rainout processes, can be elaborated on the basis of the premise that sulphate aerosol is the major transport medium for radio-caesium, because sulphate has been studied intensively as one of the most abundant substances of cloud condensation nuclei and as a key component of precipitation chemistry (Akata, et al., 2019).

2. Air quality analyses using different models and measurements

The COPERT vehicular emission model of average speeds was applied to provide the emission factors, so that the macroscopic emissions for the traffic network could be calculated. As a next step, the WRF-Chem online coupled weather and atmospheric chemistry model was adapted to estimate atmospheric dispersion of pollutants. As a result, it can be stated that combining macroscopic road traffic modelling with atmospheric models can enhance the estimation efficiency of urban air pollution (Kovács et al., 2021).

For urban monitoring sites, air quality forecasting with atmospheric chemistry transport models have limitations due to the complexity of air pollution sources in cities. In the winter, urban heating is the main source of air pollutants in Hungary. Winter stagnation events with low wind speed can increase concentrations and cause the deviation of air quality guidelines within the city. For Budapest, a time-dependent downscaling method was used to predict the daily mean of $\text{PM}_{2.5}$ concentration for the heating seasons in 2018–2021. Nine individual

models of the Copernicus Atmosphere Monitoring Service (CAMS) were compared to PM_{2.5} data from six urban measurement sites in Budapest. Downscaled predictions were produced by the linear combination of the CAMS models using spatially constant and time-dependent weights fit on the previous 10-days long period. Predictions from the model fusion were more efficient in smog episodes and had similar overall efficiency to the bias-corrected ensemble. The fusion of the CAMS models leads to a more accurate forecast of wintertime PM_{2.5} peaks in urban monitoring sites of Budapest than using any of the individual models (Varga-Balogh et al., 2020a).

Similarly to other countries, the first wave of the COVID pandemic induced a collapse of mobility in Hungary during the spring of 2020. From the environmental perspective, the obtained road traffic reduction of 20–50% could be regarded as an undesired traffic regulation experiment. Air quality impacts within Hungary were evaluated based on data from 52 monitoring sites measuring concentrations of pollutants NO_x, O₃, and PM₁₀. Air pollution during the lockdown was compared to the same period (February–June) in the reference years 2014–2019. The large spatial heterogeneity of the air quality response was explored. The emission reduction coincided with the extreme weather of 2020, characterized by unusually warm pre-lockdown February and spring drought. The anomalously low pre-lockdown air pollution was further reduced (NO_x) or increased (PM₁₀) during the restrictions. PM₁₀ response was modest and largely weather-driven (Varga-Balogh et al., 2021).

Besides the complex dispersion model simulations, the SHERPA source-receptor model was also used to investigate the contribution of each emission sector to the domestic PM₁₀ concentration values. We also investigated the effects of local and remote sources on air quality (Horváth, 2022; Horváth et al., 2023).

An intercomparison of different air quality sensors were carried out in November and December 2020 at the Observatory of the Hungarian Meteorological Service (Gilice tér, Budapest). Based on the result we have demonstrated the reliability and applicability of different low-cost PM sensors (Mészáros, 2022).

The results are potentially applicable for future planning of smog alerts and other emergency systems.

3. Development of a new coupled dispersion and food chain model

Based on the former investigation and sensitivity analyses, a detailed dispersion model was developed to estimate the transport and deposition processes of radionuclides. A puff modelling method was applied to simulate atmospheric dispersion on the 1–30 km scale. Meteorological data was obtained from the GFS and AROME numerical weather prediction models at 0.5° and 2.5 km horizontal resolution, respectively. Grid-scale puff advection and turbulent diffusion was simulated with a Lagrangian approach. Subgrid-scale diffusion was simulated with a Gaussian approach. Dry deposition was calculated assuming a component-specific constant deposition velocity. The food-chain model was coupled through component-specific dose factors estimating food-chain doses from the deposited activity at each gridpoint. Besides that, external (cloud) dose was estimated from air concentrations, ground dose from deposited activity and thyroid dose from deposited ¹³¹I activity.

Examining the factors available in different databases, we managed to define and support the most sensitive variables of the food-chain model, which are the Tf and Cr values, with a PRCC analysis. We would also like to confirm that location-specific (specific continent and region) knowledge of these helps to produce more accurate model results. Laboratory tests were extended to ⁴⁰K, ²¹⁰Po, ²²⁶Ra, ²³⁸U, ²⁴¹Am radioisotopes. Accumulation from soil to plant and from plant to animal was investigated. The individual transfer factors (Tf) and concentration ratios (Cr) were determined from the results. In this way, we can further refine the value of the

internal dose (ingestion dose), which serves as one of the output data of our food chain model. We compared our results with the currently available databases, so we were able to refine our developed model in a location-specific manner. We were also able to validate the model by running several scenarios (Duogn et al., 2020; Mamyrbayeva et al., 2020).

For the development of a new food chain model the deposition coefficients (for ^{90}Sr and ^{137}Cs) and sensitivity study of parameters in several models were carried out. An important task was the determination of the bioaccumulation factors (for ^{90}Sr and ^{137}Cs) in different plants and animals. Bioaccumulation factors (BAF) are one of the important parameters to evaluate the uptake of radionuclides from soil/water/air to plants or animals. It is the ratio of radionuclide concentration in the plant/animal to the concentration of radionuclides in soil/water/air per unit mass. Knowledge on the transfer of ^{137}Cs and ^{90}Sr radionuclides to livestock products is relatively available from both laboratory and field studies, therefore we compared our results with the data from these studies. The comparison showed that our results are of the same order of magnitude as the literature values (Baigazinov et al., 2020).

4. Mobile measurements of air pollutants

Mobile particulate matter measurements have also been carried out along different routes in the downtown of Budapest. These measurements can provide better temporal and spatial resolution of air quality in a specified region of the city than the data from fixed, automatic measuring network. The spatial distributions of the measured concentrations clearly show the impacts of roads, parks and the well ventilated Danube valley (Tordai és Mészáros, 2023). Furthermore we have found some hot-spot areas, where we have measured peak concentrations due to the crossroads. We have found that the built environment has a major impact on the spatial patterns of urban air pollution (Csapó és Mészáros, 2019). Even within a short distance, large concentration deviations can occur, which are sometimes not described by data from stationary air pollution monitoring stations. Results show large spatial variability of concentration values, underlining the importance of car-free streets, urban vegetation and the good ventilation.

The spatial distribution of air pollution conditions within the city can also be described by air quality models, but the actual effects of built-in conditions and air pollution along individual routes can be reproduced primarily by mobile measurements (Mészáros, 2022). Measurement results can help to refine the input parameters of environmental models.

Conclusion

Given the complexity of the field, researchers from the two universities (ELU and UP) decided to collaborate on this project. This collaboration between meteorologists and radiologists has provided a number of synergistic benefits. The collaboration between the two universities, which have extensive experience in the field, has provided a unique research potential to complete the research work and a guarantee for further progress in improving knowledge on air pollution and its effects, as well as to induce new researches.

Our measurements have been used to analyse in detail the spatial and temporal variation of air pollutants. The sensitivity of the different model inputs was examined. The spatial and temporal distribution of air pollution were analysed using model simulations at different scales. An integrated dispersion and food chain model was developed to study the complex contamination pathways of aerosol particles and radionuclides. The results may be useful for further model development.

Our results were presented at national and international scientific conferences and in SCI journals (see reference list). Students and PhD students were also involved both in the

measurements and in the model simulations. This allowed students to theses and doctoral dissertations on the subject.

Conference participations

In the following we summarize the conference participations and the presented results during the project (Table 3.).

Table 3. Participations in national and international conferences

Conference	Site	Date	Project results
EGU General Assembly Conference 2019	Vienna, Austria	2019.04.07–12	Balogh et al., 2019
EGU General Assembly Conference 2019	Vienna, Austria	2019.04.07–12	Kovács et al., 2019b
45 th Meteorological Scientific Days	Budapest, HU	2019.10.14–15.	Mészáros et al., 2019
EGU General Assembly Conference 2020	online	2020.05.04–08.	Varga-Balogh et al., 2020b
VII. Terrestrial Radioisotopes in Environment International Conference on Environmental Protection	Veszprém, HU	2020.05.18–22.	Baigazinov et al., 2020
46 th Meteorological Scientific Days	Budapest, HU	2020.11.19.	Leelőssy et al., 2020
16 th Carpathian Basin Conference for Environmental Sciences	Budapest, HU	2021.03.30–04.01.	Bushra et al., 2021a
Third National Conference on Radiation Awareness and Detection in Natural Environment	online	2021.03.18–20.	Mamyrbayeva et al., 2021
EGU General Assembly 2021	online	2021.04.19–30.	Bushra et al., 2021b
10 th International Conference on Radiation in Various Fields of Research	Herceg Novi, Montenegro	2022.06.13–17.	Baiganizov et a., 2023
XV. Hungarian Aerosol Conference	Hévíz, HU	2022.09.21–23.	Mészáros, 2022
21 st International Conference on Harmonisation within Atmospheric Dispersion Modelling for Regulatory Purposes	Aveiro, Portugal	2022.09.27–30.	Varga-Balogh et al., 2022
VIII. Terrestrial Radioisotopes in Environment International Conference on Environmental Protection	Vonyarcvashegy, HU	2022.10.04–07.	Baiganizov et a., 2023
48 th Meteorological Scientific Days	Budapest, HU	2022.11.17–18.	Leelőssy et al., 2022
18 th Carpathian Basin Conference for Environmental Sciences	Szeged, HU	2023.05.17–19.	Horváth et al., 2023

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(our selected publication during the project):

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