Joint Bratislava–Prague studies of radiocarbon and uranium in the environment using accelerator mass spectrometry and radiometric methods

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Abstract A research program has been established between Bratislava and Prague groups to study natural and anthropogenic radionuclides in the environment using both Accelerator Mass Spectrometry (AMS) and radiometric methods. The first studies have focused on ^{14}C activity variations in the atmosphere and biosphere with the aim to evaluate an impact of Czech and Slovak Nuclear Power Plants (NPP) on the environment, and on the development of AMS technique for investigation of actinides (mainly uranium isotopes) in the environment.

Keywords Environmental radioactivity · Atmosphere · Tree rings · Radiocarbon · Uranium · Accelerator mass spectrometry

Dedicated to the memory of Prof. Petr Beneš.

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Introduction

Accelerator Mass Spectrometry (AMS) represents a revolution in analytical technologies for measurement of long-lived radionuclides when instead of waiting for decay products of radioactive nuclides (e.g. alpha-, beta-, or gamma-rays), they are analyzed directly mass-spectrometrically as they would be stable [1, 2]. For some long-lived radionuclides this change has improved the sensitivity of analysis by more than five orders of magnitude [3, 4]. These recent developments which also include other mass spectrometry methods (e.g. ICPMS [5]) have had great impact on applications of radionuclides as tracers of environmental processes which because of the lack of suitable samples (e.g. when too big samples were required), or because of limited analytical sensitivities, could not be realized in the past [2, 6]. In this way, new investigations in all environmental compartments, e.g. in the atmosphere, biosphere and hydrosphere, as well as a coupling of interactions in the geosphere could be carried out [7]. The AMS opened new investigations in isotope oceanography [8, 9], in climate change studies [10], in space science studies [11], and environmental research [12]. The recent developments in the AMS technologies may also be illustrated by another change in the philosophy of analysis represented by a transfer from a bulk sample analysis to specific compound analysis of stable and radioactive isotopes, very often using coupled analytical systems, (e.g. coupling of GC/MS with AMS) which will open new applications in environmental and biomedical sciences [13].

Although the AMS technology was originally developed for radiocarbon measurements, later applications included almost all long-lived radionuclides [1–5]. In this respect, specifically new isotopic tracers such as for example ^{236}U have attracted interests in terrestrial and marine

environment investigations [5, 8, 12], and naturally they have been part of the Prague–Bratislava collaboration project. Important part of these new developments included preparation of targets for ion sources of tandem accelerators with the aim to obtain maximum ion yields. In the analysis of ^{236}U, the separation precision, minimization of contamination, and sample handling are the state of art of every AMS laboratory. The natural isotopic ratio $^{236}U/^{238}U$ ranges from 10^{-10} to 10^{-14}, which causes natural ^{236}U practically undetectable by conventional techniques [14]. On the other hand, the anthropogenic ratios could be as high as 10^{-3}, what brings many challenges to sample preparation with natural isotopic ratios. The majority of published methods of AMS analysis deals with sample preparation as the crucial issue and the ^{236}U is also the case; because of its abundance every even small advantage in measurement could highly improve the final result. Measurements of low isotopic ratios of $^{236}U/^{238}U$ demand improvements in AMS techniques, which could also be seen in applications of new target matrices. Many current methods of sample preparation for $^{236}U/^{238}U$ isotopic ratio determinations terminate with the conversion of uranium into oxides. The isobaric interferences of more abundant $^{235}U^{17}O^-$ and $^{234}U^{18}O^-$ ions may, however, require improvements in the target technologies. Recently, the development of fluoride target matrix has been presented, which could increase the ion source yields. In the same time, using monoisotopic fluorine significantly decreases the isobaric interferences. This matrix has already been used in AMS measurements of other radionuclides, however, more studies are necessary to allow and optimize this method for routine measurements [15–17].

Environmental radionuclide studies around the Czech and Slovak Nuclear Power Plants (NPP) have been of interest of both Bratislava and Prague groups for many years [18–21]. Radiocarbon has been of specific interest because of its long half-life (5,730 years) and its radioecological significance, because it may have highest contribution to radiation doses of public from operation of NPPs [22]. Actinides (mainly uranium and plutonium isotopes) on the other hand play an important role in the case of nuclear accidents, or long-term storage of radioactive wastes [5, 8, 23].

Two main methods of ^{14}C monitoring around NPPs have been developed in the past. The most sensitive and precise one is a direct monitoring of atmospheric $^{14}CO_2$ together with measurement of CO_2 mixing ratio.[1] Utilising $^{14}CO_2$ activities[2] and CO_2 concentration data, the molar activity concentration can be calculated (i.e. $^{14}CO_2$ quantity per molar[3] unit of the air). Such robust parameter is connected only with quantity of $^{14}CO_2$ molecules in molar unit of air and cannot be influenced by Suess effect caused by dilution of ^{14}C abundance in carbon isotopic mixture as a result of local/regional fossil CO_2 emissions [21, 24]. In other words, CO_2 molar activity concentration cannot be locally influenced by releases of fossil CO_2, but only by emissions of $^{14}CO_2$. A disadvantage of the (direct) molar activity concentration monitoring is higher operation cost, and necessity of special sampling and monitoring instruments [25, 26].

A biota based monitoring is cheaper and can be applied on a bigger scales, the resulting parameter is, however, only the ^{14}C activity (i.e. abundance of ^{14}C in carbon isotopic mixture). Such parameter can be influenced by local/regional Suess effect, which causes a dilution of ^{14}C in carbon isotopic mixture by fossil CO_2 emissions. This monitoring method requires therefore a reference site with a local Suess effect similar to that of a NPP site. When a suitable monitoring is applied, periods of biomass accumulation should be contemplated carefully. Biota samples, growing during spring/autumn, could represent well varying ^{14}C levels as a result of different microclimatic conditions.

Recent developments in accelerator technologies and their applications in environmental sciences have been an inspiration for creation of a Centre for Nuclear and Accelerator Technologies (CENTA) at the Comenius University in Bratislava. The centre consists of a tandem accelerator designed for nuclear reaction studies and for ion beam analysis of environmental and material samples. A special interest has been devoted to AMS studies of long-lived radionuclides in environmental and life sciences, including evaluation of ^{14}C levels around NPPs. A Prague–Bratislava program has been established with the aim to use the AMS technology for investigations of radionuclide variations in the environment. This paper is focusing on radiocarbon variation studies around the Czech (Temelín—2×1000 MW$_{el}$ and Dukovany—4×440 MW$_{el}$) and Slovak (Jaslovské Bohunice—4×440 MW$_{el}$) PWR based NPPs (Fig. 1) using radiometric and AMS techniques. We compare radiocarbon concentrations measured in atmospheric carbon dioxide with those in biota samples with the aim to study long-term contributions of NPPs on ^{14}C levels in the environment. The development of a new method of target preparation for analysis of uranium isotopes in the environment by AMS is also presented.

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[1] CO_2 mixing ratio means number of CO_2 mol per one mole of air.

[2] Following Stuiver-Polach convention ^{14}C (or $^{14}CO_2$) activity is a reporting of ^{14}C abundance in carbon isotopic mixture, where 0 ‰ of $\Delta^{14}C$ is about 0.226 Bq of ^{14}C per gram of carbon isotopic mixture [27, 28].

[3] Molar units of air can also be replaced by normalized volume or weight units [25].

Fig. 1 Radiocarbon sampling sites in the Czech and Slovak Republic (*A* Žlkovce–Jaslovské Bohunice NPP, *B* Dukovany NPP, *C* Temelín NPP, *D* Prague–Bulovka, *E* background sampling site at Košetice, *F* Bratislava)

Experimental

Radiocarbon (sampling and measurement)

Bratislava group

Long-term monitoring of ^{14}C in atmospheric carbon dioxide has been carried out monthly in Žlkovce, a monitoring station located close to the Jaslovské Bohunice NPP [29, 30], and at the Bratislava university campus [31]. The method is based on bubbling the air through NaOH solution, precipitation of $BaCO_3$ in the laboratory, liberation of CO_2 by H_3PO_4, and finally preparation of CH_4 (over a ruthenium catalyst) as a counting gas of proportional counters. A detailed description of sampling, preparation of gases and counting methods has already been described [32, 33].

Core sampling of tree rings from a lime tree (*Tillia cordata*) was also carried out in Žlkovce. Tree rings after separation under a microscope were dried, chemically cleaned (acid-basic-acid), combusted to CO_2, and finally graphite targets were prepared for AMS measurements, which were carried out in the VERA laboratory of the Vienna University [34, 35].

Prague group

Monitoring of atmospheric $^{14}CO_2$ is performed in the Czech Republic at two sites: Prague–Bulovka and Košetice. The site in Prague is near the border of the urban agglomeration of Prague, in the vicinity of a heavily used motorway; therefore, a local load from fossil fuel combustion can be expected there. The Košetice site is situated in the area of the Meteorological Observatory (a part of the

Czech Hydrometeorological Institute). The surroundings of the Košetice site have an agricultural-forestry character, without significant local sources of fossil CO_2. Monthly CO_2 samples from both sites were collected by bubbling air through a 0.7 M NaOH solution in 1.9 L flasks (the final amount of carbonates did not exceed 30 % of the saturation capacity of the solution). Subsequently, samples were processed in laboratory by acid carbonate decomposition using H_3PO_4.

In the period of 2007–2008 samples of *Urtica dioica* were collected in the surroundings of Dukovany NPP–EDU and Temelín NPP–ETE (18 samples, without a preferred wind direction). Stinging nettle or common nettle is an herbaceous perennial flowering plant having an exiguous annual biomass supply and a thin root system. Vegetation period of this plant in the Central Europe is between the end of March and beginning of November. Distances of sampling points were in the interval of 0.7–1.2 km, where maximal ^{14}C activity surplus can be expected. Reference samples (rEDU and rETE, 11 samples, 10–20 km from a given NPP) were collected in localities with estimated densities of roads and traffic loads similar to those in the vicinity of NPPs. Biota samples were washed with 10 % HCl and distilled water, dried at 105 °C, homogenized, and combusted to produce CO_2.

Samples of resulting CO_2 (both from exposed NaOH solution and from combustion of biota samples) were after purification processed by benzene synthesis [20, 21]. ^{14}C activity was measured by liquid scintillation spectrometer Quantulus 1220 in 3–mL low-background Teflon vials.

Oxalic acid NIST standard reference material SRM 4990-C was used for calibration by both the Bratislava and Prague groups. Resulting ^{14}C data are reported as $\Delta^{14}C$ (in ‰), following Stuiver-Polach convention [27].

Uranium targets chemistry

In the analysis of $^{236}U/^{238}U$ mass ratios in environmental samples, the final sample preparation step has been usually a conversion of uranium into its oxides. Another possibility could be a conversion of uranium dioxide into anhydrous uranium tetrafluoride, which may therefore offer a higher isobaric suppression for molecular ions, together with a higher accuracy and sensitivity of uranium isotope analysis.

For the preparation of uranium tetrafluoride, uranyl nitrate, hydrofluoric and hydrochloric acid, ammonium hydrogen difluoride, and hydrazine hydrate solution (24–26 %) of p. a. commercial preparation were used. The preparation of uranium tetrafluoride was carried out following the published procedures [17, 36, 37].

Fig. 2 Time series of atmospheric $^{14}CO_2$: Prague–Bulovka (local and regional Suess effect), Košetice (regional Suess effect), and Jungfraujoch (clean-air Alpine monitoring station). Interannual decrease of ^{14}C activities is visible in all time series [38, 39]
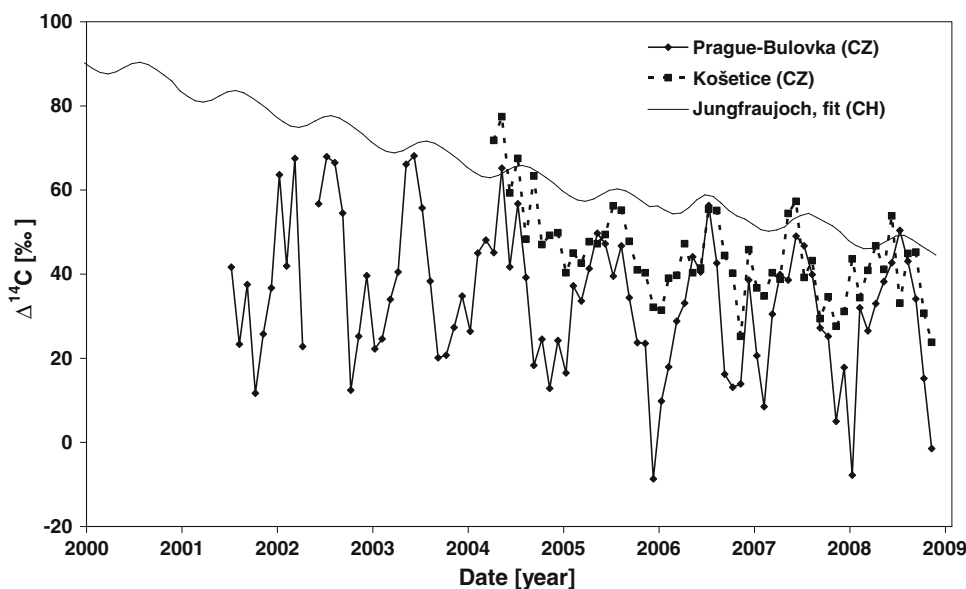

Table 1 Basic statistical parameters of ^{14}C levels in biota samples collected in the vicinity of NPPs Dukovany (EDU), Temelin (ETE), and in corresponding reference localities (rEDU, rETE), indicating a negligible impact of NPPs

	EDU	rEDU	ETE	rETE
Average	37.9	33.2	35.4	30.0
Median	35.5	33.4	34.5	30.3
Standard deviation	7.2	4.7	4.6	2.4
Variation	52.2	22.1	21.3	5.9
Number of observations	10	6	8	5
Observed maximum	52.7	39.7	42.9	32.5
Observed minimum	30.0	26.6	28.8	27.0

Activities are reported in ‰ of $\Delta^{14}C$ [27]

Results and discussion

Radiocarbon around Czech NPPs

Time behaviour of atmospheric $^{14}CO_2$ activity can be characterized by an interannual decrease since the beginning of 1990s, which is caused by global Suess effect [38, 39]. Seasonal fluctuations with minima during cold parts of the year are amplified by local and regional Suess effect, what is evident from the time series of atmospheric $^{14}CO_2$ in Prague–Bulovka (Fig. 2). The observed $^{14}CO_2$ activities approximate to reference values from clean-air high mountain station Jungfraujoch during summer months [39].

Basic statistical parameters of ^{14}C monitoring in the vicinity of NPPs and in reference localities utilising nettle plant (*Urtica dioica*) samples (EDU, ETE, rEDU, rETE) are reported in Table 1. The results from each type of area

were compared utilizing *t* test (unpaired, probability of first kind of observation error 5 %).

The observed difference of 5.4 ‰ in $\Delta^{14}C$ (ETE−rETE) for nettle sampling is in a good agreement with observed difference of 4.8 ‰ of $\Delta^{14}C$ (significant for 5 % probability of the first kind of observation error) obtained on the base of previous sampling campaign based on leaves of deciduous trees collection in years 2002–2005. Application of nettle plants as a sampling material also seems to reduce variations of ^{14}C activities on half in comparison with tree leaves sampled during our previous campaign [40].

The observed variations in ^{14}C activities for each type of locality are due to fluctuations in local Suess effect, which can be well visible during winter months (e.g., heating of houses, increased occurrence of atmospheric inversions). Another source of variations can be due to a relatively short time interval of biomass accumulation in tree leaves (about 4–5 weeks in April and May), when the activity of atmospheric $^{14}CO_2$ changes relatively quickly (Fig. 2). The biomass accumulation in tree leaves also depends on local microclimatic conditions (atmospheric precipitations, soil moisture, and sunlight exposure), which can cause shifts in the period of atmospheric $^{14}CO_2$ intake by tree leaves [40–42].

Radiocarbon around Slovak NPPs

Another possible sampling material for ^{14}C monitoring around NPPs is represented by tree rings. Such type of sampling enables to reconstruct ^{14}C chronology in a given locality, usually for several decades. To reach one-year time resolution, an analytical method based on AMS measurement (in which a sufficient sample of carbon
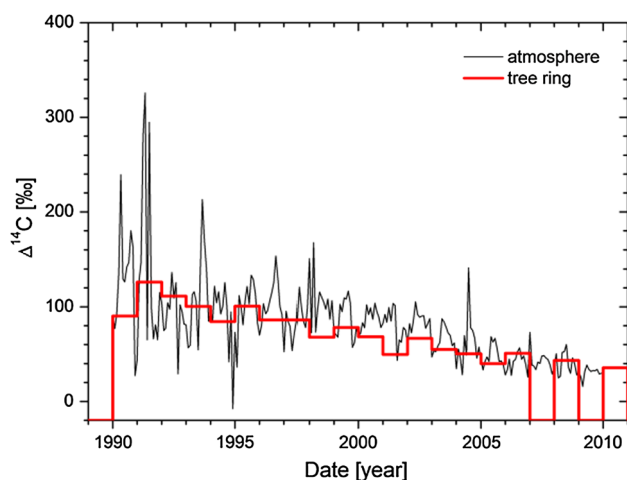
Fig. 3 Record of relative ^{14}C activities in tree rings, compared with monthly measurements of ^{14}C in atmospheric CO_2 at the Žlkovce site

Fig. 4 Comparison of uranium oxide and fluoride ion currents in the Faraday cup

quantity is only a few milligrams) is necessary. Results of ^{14}C activity in tree rings sequence lime tree (*Tillia cordata*) collected at the Žlkovce monitoring station are presented in Fig. 3. Another advantage of the AMS based method of ^{14}C activity determination is a smaller uncertainty of measurements (reduced by about a factor of two).

Due to wood biomass accumulation from May to September, a small decrease in the comparison with maximum activities of atmospheric [14]CO_2 (during the summer period) can be expected in tree ring samples [24], what is also documented in Fig. 3. Nevertheless, possible local variations of ^{14}C activities caused by microclimatic differences (a time shift in biomass ingrowth) can be reduced for tree rings compared to leaves and other biota samples.

The atmospheric ^{14}C record in the Žlkovce monitoring station (Fig. 3) indicates large variations. The deep winter minima are due to the local Suess effect, while during a year several ^{14}C emissions from the Bohunice NPP can be identified. However, the total impact of the Bohunice NPP on ^{14}C levels in Žlkovce, as represented both by the atmospheric [14]CO_2 and the tree-ring record, is negligible. The observed ^{14}C levels are comparable with the high-altitude clean air station at Jungfraujoch, as well as with the surface air station at Košetice (Fig. 2).

The ^{14}C tree-ring record is well averaging atmospheric ^{14}C concentrations in CO_2, documenting its suitability for long-term monitoring of ^{14}C levels around NPPs.

Development of uranium targets for AMS

In the analysis of [236]U/[238]U mass ratios in environmental samples using AMS, the final sample preparation step is usually a conversion of uranium into its oxides. Thereafter, with such oxide target matrix, the UO^- molecular ions are used for the beam production. Recently, Wang et al. [17]
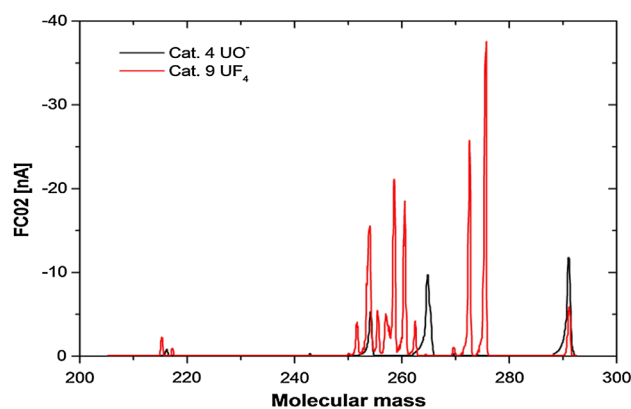
studied preparation of fluoride matrix in the [236]U/[238]U ratio determination, and showed a feasible method of conversion of uranium dioxide into anhydrous uranium tetrafluoride. In comparison with uranium oxide matrices, uranium fluoride targets contain no oxygen and hydrogen, and may therefore offer a higher isobaric suppression for molecular ions together with a higher accuracy and sensitivity of uranium isotope analysis. However, the preparation of anhydrous UF_4 targets is more complicated than the preparation of uranium oxide targets, and the preparation procedure is associated with hazardous evaporation of concentrated hydrofluoric acid.

When reproducing the procedure of anhydrous uranium tetrafluoride preparation published by Wang et al. [17], the sample mass scan of AMS measurement clearly showed several peaks related to oxide molecular ions (Fig. 4). The ion current from only one UF_2^- sample was in average higher by about 50 % than the UO^- current from the uranium oxide samples. The targets were completely sputtered away, and the estimated ionization yields of UO^- and UF_2^- were of the order of 10^{-3}. However, the improved procedure of the UF_4 targets production could provide even higher ionization yields.

In order to prepare pure anhydrous uranium tetrafluoride, two more preparation procedures have been added into this research. Yeamans [36] published a simpler preparation procedure consisting of a contacting uranium dioxide with ammonium bifluoride, and a consequent combustion at the temperature around 400 °C. Sahoo and Satapathy [37] published probably the most suitable procedure among the others, because the entering uranium compounds could be in the +VI oxidation state. In this publication, the uranyl compound is mixed with hydrazine bifluoride and combusted at 400 °C. The uranium reduction takes place during the temperature increase. Samples of anhydrous uranium tetrafluoride were prepared using all three methods and then measured by X-ray powder

diffraction. In every spectrum the uranium oxide diffraction lines of low intensities can be easily identified, and in addition, the diffraction patterns of uranium dioxide showed also the lines of metaschoepite (($(UO_2)_{4-}O(OH)_6(H_2O)_5$)) in which uranium is partially oxidised to higher oxidation state. In order to improve the purity of uranium tetrafluoride samples, a new uranium dioxide sample will be prepared and the influence of reducing atmosphere during the combustion will be also studied.

Conclusions

The main results presented in this paper may be summarized as follows:

(i) Two biota-sampling campaigns around the NPPs in the Czech Republic (2002–2005, leaves of deciduous trees; 2007–2008, nettle plants) indicate an excess of ^{14}C levels close to the border of statistical significance. The ^{14}C levels in nettle samples collected in the vicinity of the Temelín NPP were about 5.4 ‰ above the average reference background value. Recently tree-ring samples around the Czech NPPs have also been collected, which are at present under analysis. We are also considering a regular ^{14}C activity monitoring in atmospheric $^{14}CO_2$ around NPPs.

(ii) The ^{14}C tree-ring record at the Žlkovce monitoring station (located close to the Bohunice NPP, Slovakia) has been well averaging atmospheric ^{14}C concentrations in CO_2, documenting its suitability for long-term monitoring of ^{14}C levels around NPPs. The observed ^{14}C levels were comparable with those observed at clean-air monitoring stations at Jungfraujoch (a high-altitude station in Swiss Alps) and at Košetice (a low-altitude station in Czech Republic).

(iii) The development of uranium tetrafluoride targets for AMS analysis of $^{236}U/^{238}U$ mass ratio in environmental samples has shown promising results, but more work is needed to clearly demonstrate advantages of this technique.

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