Research proposal

Arctic climate and atmospheric dust variations during the late glacial period from remnants of the Laurentide Ice Sheet

Background and objectives | Climatic records show that large, widespread, abrupt climate changes have occurred repeatedly and within decades in the recent geological past[1], including the last glaciation. Abrupt climate changes during this period were centered in the North Atlantic, which is a key region where vigorous wind systems encounter the southernmost extension of sea ice and oceanic currents with connections to the deep ocean via convection[2,3]. There is growing concern that increasing recent CO_2 forcing and global warming will lead to a global cascade of rapid regime shifts in the near future[4], which are difficult to model and predict and would be a global threat to civilisation. The reason for this lack of ability to predict tipping points is due to limited understanding of feedbacks in the climate system during abrupt events. In particular, the role of atmospheric dust, which has both direct and indirect influence on global climate through diverse physical and biogeochemical processes and feedbacks[5-7], is still relatively poorly known.

Abrupt climate changes of the last glaciation are preserved in Greenland Ice Sheet (GIS). High resolution ice core oxygen isotope records reveal that the generally cold glacial climate was perturbed by numerous rapid shifts to warmer interstadial conditions, called Dansgaard–Oeschger (D–O) events, over the last glaciation[8,9]. In Greenland, air temperatures during D–O events rose by 5–16 °C within decades[10,11], followed by a less rapid temperature decline back to stadial conditions. This D–O type climate variability is coupled with dust concentration and particle size variations, with rapid decrease in Ca^{2+} ions (a dust concentration proxy) at the onset of interstadials and slower run-up following their end[12]. These abrupt variations in dust characteristics were accounted for by changes in transit and atmospheric residence time, transport distance and dust source strength[13-15], indicating that the climate of continental sources and Greenland must have been coupled[16,17]. However, causal and response effects are unclear.

The effects of D–O events extended across much of the Northern Hemisphere[18] and were identified in well-dated terrestrial records such as speleothems in Europe and Asia[19-22]. Recent work led by the P.I. demonstrated close and potentially causative links between North Atlantic climate and European dust activity and paleotemperatures at D–O timescales[23,24], controlled by meridional displacements of the upper-tropospheric Atlantic jet. This implicates much wider abrupt changes in dustiness during D–O temperature variations, with the causal relationships and climate impacts remaining largely unclear. The only undisputed, high resolution records of hemispheric scale dustiness come from Greenland[25], but these records, which are key in understanding and interpreting dust-climate interactions on D–O timescales, are confined to the central part of the GIS. Clay mineralogy and Sr-Nd-Pb isotopic compositions of dust aerosols in central Greenland ice cores suggest a SE Asian origin over the LGM[26,27]. Despite wide acceptance of this model, the sources of dust are not unequivocally constrainable as yet and other potential dust sources to Greenland, including Africa[28-30] and central Europe[31], cannot be excluded based on expanded datasets. This is further corroborated by the latest, combined Hf-Sr-Nd isotope data of LGM dust samples collected by the P.I. around the northern hemisphere (NH)[32]. Such ambiguities mean we have a limited knowledge on wider glacial dust loading and present a major impediment in understanding what specific areas were emitting dust during abrupt events, the cause of these emissions, and the consequences for climate. This also implies that the major dust transport pathways and atmospheric circulation in these events is poorly constrained, preventing understanding of feedbacks between dust and climate and the role of different atmospheric patterns in dispersing dust. Furthermore, the reliance on Greenland ice core records exacerbates these problems as it is unclear how dust loading and composition was over a much wider area. Thus, it is essential to find another large scale dust and climate record in the NH. Ice marginal stable isotope and dust records from remnants of the Laurentide Ice Sheet (LIS) would have great potential in filling this knowledge gap.

In the Canadian Arctic, during retreat of the LIS many of the larger ice caps shrank but some survived, like the Barnes Ice Cap (BIC, Figure 1), which separated from the northeastern sector of the LIS 8500 years ago[33,34]. Recession of BIC during the late Holocene has left exposed formally deep ice layers of Pleistocene age along its margin[35,36] and at present, it is the only ice cap in the Canadian Arctic that allows direct sampling of relict Laurentide ice for paleoclimate research without deep-coring[33]. The BIC is geographically ideally located to the west of Greenland and ice stable isotope and dust proxy records of BIC would be crucial to answering unresolved questions of dust sources and transport to Greenland and the Arctic and gaining a better understanding of the transport characteristics and timing/amplitude of major dust events in relation to D–O variability. In obtaining dust concentration and source data from two (former) ice sheet areas, located far from plausible local sources, long range pathways of dust transport can be elucidated and the source information becomes clearer: certain pathways proposed for source areas of dust to GIS are not possible for LIS, and potentially vice versa. These paleodata would

be useful in informing model simulations of the LGM atmospheric circulation, would allow more precise paleoclimate interpretations of Greenland ice core records and better understanding of dust-climate feedbacks.

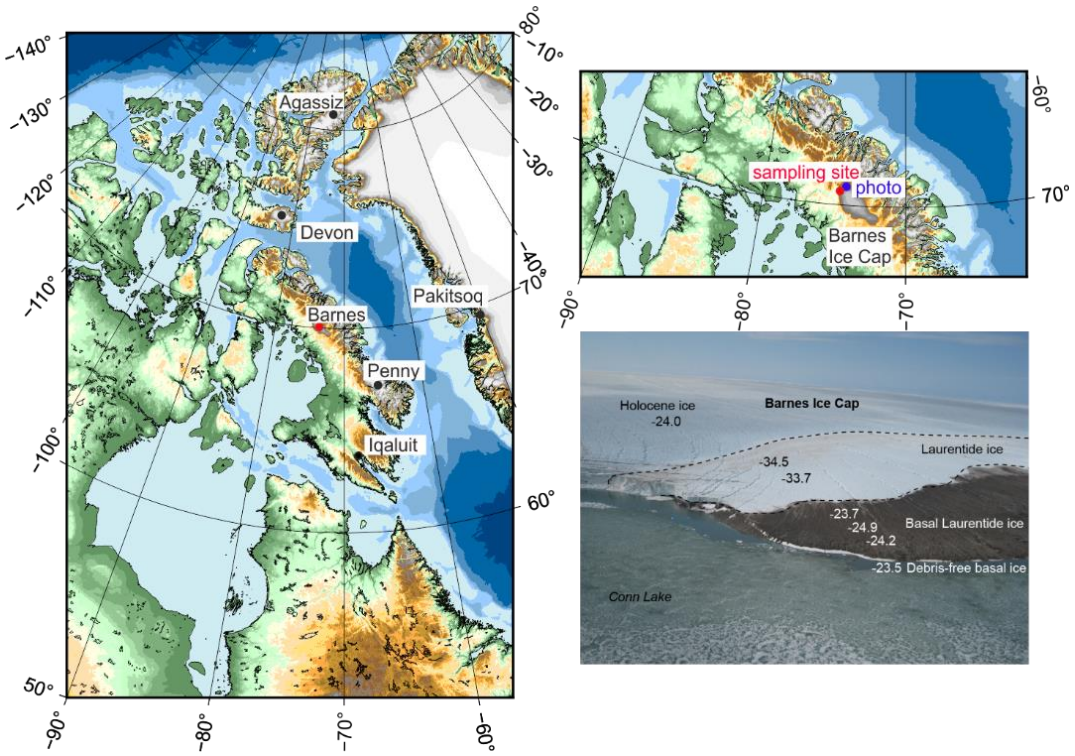

Figure 1. Maps showing the position of Barnes Ice Cap, Baffin Island, Canada and the location of planned sampling site. Nearby ice coring sites and travel locations are also shown. Photo courtesy: Gifford Miller.

This project aims to develop records of ice (water) stable isotopes ($\delta^{18}O/\delta D$ and d-excess), ionic aerosol concentrations, dust content, size distribution and source signatures along a transect across the ice margin of BIC. These proxy records, reflecting site temperature, oceanic source conditions of moisture, continental dust sources and activities and dust transport characteristics, would provide unique insights into abrupt climate changes and atmospheric circulation patterns over the LIS during the last glaciation. Surface sampling across a transect enables the collection of large masses of ice, allowing for direct, independent radiometric (^{14}C) dating of ice using dissolved organic carbon (DOC)[37], which has not been possible from polar ice cores so far due to limited amounts of available ice. The large sample sizes that can be obtained by surface sampling may also allow for precise determination of the Hf-Sr-Nd isotopic compositions of mineral dust for source fingerprinting, even for warmer interstadials and the Holocene. These periods were characterized by low dust concentrations, for which Nd-Hf isotope analyses of aerosols in ice cores are challenging or even impossible. The use of the Hf isotope tracer is especially important as Nd isotopes are not source diagnostic for dust in central Greenland ice cores[31].
While the specific sources of Greenland and wider hemispheric dust are debated, widespread stadial cooling and interstadial warming in the North Atlantic was associated with increased/decreased winter precipitation in the southwestern United States[38,39], driven by shifting polar jet positions[40]. Atmospheric reorganizations of this type, associated with D–O variability, may have alternately reactivated major dust sources in SE Asian (Chinese) deserts and loess areas during stadials[40] and drylands in continental North America up to Alaska[41-43]. We would like to directly test these hypotheses using the Hf-Sr-Nd isotopic and clay mineralogy fingerprints of dust recovered from BIC ice, as the SE Asian and North American continental sources have different mineralogical and isotopic fingerprints. Different signatures of dust in GIS versus LIS may support European/Saharan sources to GIS as these were less plausible for LIS. While not comparable in temporal resolution to central Greenland ice cores, records to be obtained from BIC margin hold a truly unique potential to provide information on climatic conditions during glacial-interglacial transitions and D–O warming/cooling in the continental interior of North America, with a major focus on the characteristics of dust transport to LIS and source region activity.

Methodology | This part of the proposal describes sampling, field work and the various methods to be applied in relation to specific aims of the project.

Sampling | The proposed sampling location is on the northwest margin of BIC (Figure 1). This site is selected because of the wide, relatively easy access to exposed white, bubble-rich ice of Pleistocene age (Figure 1) along the

gentle sloping ice margin (no hazards like crevasses). The white ice is underlain by gray-white ice, of which the lower parts are littered by rock debris[33]. Superimposed on the white ice, there are bubble-poor blue ice layers of early Holocene age. Before sampling, any slush or snow will be scraped off and ice samples will be collected from ~20 cm below the surface along a transect from the basal layers of white ice, through the white/blue ice contact and partly into blue ice. Before sample collection, we will measure the strike and dip of ice foliation on the transect to investigate if there are recumbent folds in the ice strata. Subsequent sampling will be performed in ~0.5 to 1 m horizontal resolution in white ice and ~2 m in blue ice for $\delta^{18}O/\delta D$ and aerosol concentration/size analyses. For ^{14}C analyses of DOC and dust isotopic composition, larger samples will be taken using a light-weight fuel-powered corer (30 cm diameter barrel). Given the remote location of BIC, we have to use airplane and helicopter to travel to the sampling site. Our first base will be Iqaluit (Figure 1), accessible by daily flights from Ottawa. From there, we will directly fly to Lewis Camp (70°24.3N, 74°55.1W; a landing strip in the northwest sector of BIC) using an aircraft to be chartered under logistical support with Canada's Polar Continental Shelf Project (PCSP). Since Lewis Camp is located 10 km away from the selected sampling site, we will either set up a smaller camp at the ice margin or organize daily helicopter drop-off/pick-up between the site and Lewis Camp. Due to safety and logistical reasons, three/four researchers and potentially two students would be involved in sampling, for which we plan 14 days during the late spring/early summer of 2023.

Stable O/H isotope analyses | To establish a chronological framework[33] and obtain paleoclimate information on both the site and the oceanic source region of paleoprecipitation the stable isotope compositions of ice will be measured. This is influenced by temperature at the accumulation site and also by humidity and wind speed at the oceanic source region[44,45]. Stable O/H isotopic compositions of melted ice ($\delta^{18}O/\delta D_{ice}$) will partly be analysed on-site using a novel, portable (briefcase-size) stable isotope analyser of Los Gatos Research (LGR) available from the University of Ottawa. For these measurements, the ice samples will be put in bags, which will then be melted on-site by immersion in warm water and analysed immediately. Another set of samples will be shipped to Europe and additional O/H stable isotope analyses will be done using a LGR Liquid Water Isotope Analyser in the stable isotope laboratory of the host institute (Budapest, Hungary).

Radiocarbon measurements | To establish a radiometric chronology for the transect profile, ^{14}C analysis of dissolved organic carbon (DOC) is planned. DOC is ~100 times more abundant than particulate organic carbon (POC) in ice. Thus, ^{14}C analysis of DOC will be performed for ~20 samples along the transect, mostly across the last glacial and early Holocene ice to improve the site chronology using an independent dataset of radiometric ages. Since the DOC content of ice is expected to be ~10 ppb C[46] and the ^{14}C analysis requires a total mass of ~10 μg C, ~1-2 kg of ice has to be sampled for ^{14}C of DOC. These samples will be collected using a corer with a 30 cm barrel from 20 cm below the surface to avoid contamination by modern organic particles. We expect that in-situ ^{14}C production by cosmic rays will not compromise the ^{14}C-dating of DOC. In a recent work on an alpine ice core Fang et al.[37] found that the estimated amount of in-situ produced ^{14}C in the DOC fraction is smaller than the analytical uncertainty for most samples. If the dating of the DOC fraction was compromised, the POC fraction will be used. The samples will be analysed primarily at the Paul Scherrer Institute (PSI), Switzerland[37,47] and as a double-check on validity, some duplicates will be run in the A.E. Lalonde AMS Radiocarbon Laboratory in Ottawa.

Analysis of aerosol size/concentration and ionic species | To derive information on dust source activity/strength, aerosol residence/transit time and to better understand atmospheric dust dynamics in relation to abrupt climate change, ionic species and aerosol size/concentration measurements will be done on discrete ice samples. These will be collected along the transect, packed in zip-lock plastic bags on-site and stored in insulated cooling boxes below the freezing point. These ice samples will be decontaminated by triple-rinsing in ultrapure Milli-Q water at the University of Ottawa, split into two and shipped to the Canadian Ice Core Lab (CICL), University of Alberta (http://uab.ca/cryo). In this laboratory the dust size and concentration measurements will be performed using a Beckman Coulter Multisizer 4e instrument, which measures the short-term changes in electrical impedance across a very small aperture (30 μm) through which particles suspended in a diluted electrolyte solution are forced to flow. Such changes are proportional to the particle volume, thus a particle volume-size distribution spectrum can be obtained for each sample, together with the dust concentration. Using this technique it is feasible to measure concentrations of a few ppb over a size spectrum of 400 log-size channels on less than 5 mL of sample. The high sensitivity, precision and accuracy of Coulter Counter measurements have made this a reference technique for eolian dust counting/sizing in polar ice[48]. Ionic species, including Cl^-, Br^-, SO_4^{2-}, NO_3^-, Na^+, NH_4^+, K^+, Mg^{2+}, and Ca^{2+}, will also be analysed at CICL using a Thermo Dionex ICS-5000+ DC ion chromatography system. Limits of detection for ionic species depend on the chromatographic conditions, sample volume, and the respective species, but are generally on the order of few tenths to a few μg/l, while the precision is ±10% on average[13,49,50].

Hf-Sr-Nd isotope analyses | To constrain major potential sources of dust deposited on the LIS, we will measure the Hf-Sr-Nd isotopic compositions of the aluminosilicate fractions of aerosols trapped in ice. Altogether 25 ice samples collected across the sampling transect at the northwestern part of BIC will be prepared, along with some 5-6 of basal, debris-rich ice. The large ice blocks (1-2 kg each) will be decontaminated by scraping of ice samples under laminar flow bench at -30 °C, and three subsequent washing in ultrapure Milli-Q water. The dried dust samples (and the <2 μm fractions of moraines) will be treated with acetic acid to remove carbonates and the remaining aluminosilicate fractions will be decomposed in closed Teflon vessels at 230 °C using ammonium bifluoride and HNO_3. Subsequently, elemental separations and purifications will be done using a column chemistry setup specifically designed to prepare Hf, Sr and Nd for isotopic measurements from low amount of dust, developed by the P.I. and the cooperative partner at the University of Vienna[51,52]. All the preparations will be done in clean laboratory environment at the University of Vienna, including the Sr-Nd isotope measurements using a Thermo Scientific Triton XT Multicollector Thermal Ionization Mass Spectrometer (TIMS). The Hf isotopic analyses are going to be done using a Thermo Scientific Neptune Plus Multicollector Inductively Coupled Mass Spectrometer (MC-ICPMS) at the Institute for Nuclear Research, Debrecen, Hungary.
One of the most important aspects of the proposed project is the geochemical fingerprinting of dust by combined Hf-Sr-Nd isotopic analyses, of which Hf isotopes are especially important tracers of ice core dust[31]. These measurements, however, can only be done in close cooperation between the Austrian and Hungarian partners. Laboratories and expertise at both sides are absolutely necessary to obtain high precision isotopic data from ice dust samples.

X-ray Diffraction measurements | To aid dust source discriminations, measurements of clay mineralogical compositions will be carried out. The distribution of clay minerals on the continents is a first-order function of weathering, itself largely a function of climate[53]. Beyond latitude-indicating species, any source area can have a characteristic spectrum of all the clay fraction and other fine fraction trace minerals, which partly reflect not only local lithology, but also a variety of climate and drainage characteristics. Mineralogical characterization of source areas and their distinction from each other is therefore a matter of comparing relative abundances of an entire suite of minerals, some of which may be diagnostic, but most of which are sufficiently common to be non-characteristic[26].
Dust recovered from large ice blocks collected primarily for Hf-Sr-Nd isotope measurements will be split. These sub-samples will be homogenised and saturated with K^+ and Mg^{2+} ions and oriented samples prepared. The oriented, K- and Mg-saturated samples will be analysed in air-dried state and after vapour solvation with either ethylene glycol or glycerol at 60°C for 24 h to identify expandable clay minerals (smectite and vermiculite). Additional K-saturated samples are going to be heated to 550°C to destroy kaolinite and expandable clay minerals[54]. The prepared clay samples will be analysed with a Panalytical PW 3040/60X'Pert PRO diffractometer (CuKα radiation, 40 kV, 40 mA, step size 0.0167, 5 s per step) at the University of Vienna. Some samples will be measured using a RIGAKU D/MAX RAPID II diffractometer, allowing for rapid micro area measurements of several 10 μm, at the Institute for Geological and Geochemical Research, Research Centre for Astronomy and Earth Sciences, Budapest, Hungary.

Expected results, significance and dissemination | Much of the information on abrupt climate changes over the last glaciation and Holocene of the Arctic and North Atlantic regions stem from high-resolution ice core records in Greenland. Ice cores drilled on the Penny and Agassiz Ice Caps in the Canadian Arctic mostly provided proxy climatic records of the Holocene[55-58] and partly the last 120 ka from Devon Island[59]. BIC preserves relict ice from the LIS with ice strata from the LGM ranging in $\delta^{18}O_{ice}$ from -41 to -35‰[34] and originating from the former Foxe Dome of the ice sheet[33,34,56]. Transect sampling at BIC allows for collecting relict LGM ice without deep drilling, thus making it possible to develop a relatively high resolution record of $\delta^{18}O/\delta D_{ice}$ and atmospheric impurities (concentrations, sizes, source signatures) that will complement existing ice core records, and provide insights into past atmospheric conditions over the Canadian Arctic and aerosol source region characteristics. Large ice mass sampling will enable the independent ^{14}C dating of ice to establish a more robust chronological control than is feasible in deep ice core strata and also the recovery of dust for combined Hf-Sr-Nd isotopic and clay mineralogy measurements for the first time in this sector of the Arctic. Using these dust tracers, we can gain a better understanding of aerosol sources of the LIS, centennial scale changes in dust cycle, source and transport dynamics in relation to abrupt climate fluctuations (D–O type variability) over the LGM and the early Holocene and test different dust transport scenarios to LIS/GIS.
Major findings of the project will be published in leading international journals in the field (Geology, Earth and Planetary Sciences Letters, etc.), and presented on conferences (e.g. EGU General Assembly) during the last two project years. Activities related to this project will be featured on the personal homepage of GU (www.ujvari.ggki.hu) and the website of the Geochemistry and Paleoclimate Research Group, Institute for

Geological and Geochemical Research (www.geochem.hu/gp). Information on field activities and major project outcomes will be shared with the general public in Europe and Canada, including Arctic communities through the Nunavut Research Institute (NRI, Iqaluit). We will liaise with press offices of the main universities involved in this work and report our findings to national and international media, where appropriate, via these channels. Project-related information will be regularly posted on Twitter.

Risk assessment | There are some risks related to this project. First, the field work is going to be done at a remote place in the Canadian Arctic. Snow, fog and rain can occur at any time during late spring/early summer and bad weather conditions may cause some delay in sampling. Thus, we calculated with a stay of 10 days at the sampling site and an additional 10 days reserved for bad weather and traveling back and forth between the site, Iqaluit and Ottawa. Second, the COVID-19 pandemic led to restrictions on travel to Nunavut in Canada, but fully vaccinated persons may enter the territory. As all members of the fieldwork team have been fully vaccinated with vaccines approved in Canada, we anticipate that COVID-19 will not jeopardize our field work in late spring/early summer of 2023. Third, research in Nunavut is subject to various Federal and Territorial laws and regulations. The Nunavut's Scientists Act requires that anyone conducting physical and natural sciences research anywhere in Nunavut must first obtain a licence from NRI. It usually takes 6 months to issue the licences and there is a low risk of not getting them. However, considering that our proposed research has a negligible impact on the environment, we expect to obtain the permissions with no arising issues.
Scientific risks include the failure of accurate ^{14}C-dating of DOC, which may be affected by in-situ production. However, this problem has a low chance[37] and we plan to use POC in this unlikely case as an alternative for ^{14}C-dating. These risks seem to be manageable and worth taking considering the huge scientific benefits of the project.

Research infrastructure and cooperations | This project is dependent, but also capitalizes on the expertise of international collaborators from 6 countries, including Canada, Sweden, Switzerland, Austria and Hungary. A small group of scientists will take part in the expedition to BIC, including the P.I. of this application (G. Ujvari), the cooperative partner in Austria (Prof. Urs Klötzli), Prof. Denis Lacelle (University of Ottawa), who is an expert of permafrost research in the Canadian Arctic with 2 decades of Arctic field experience, and Christian Zdanowicz (Senior Lecturer, Uppsala University), a glaciologist who conducted field work at BIC in 2000 and have years of expertise in Arctic field operations. Thomas Stevens (Senior Lecturer, Uppsala University), an internationally leading expert of Quaternary paleoclimate and dust-climate interactions, will contribute to the planning of field activities and take part in data interpretations. Field activities will be supported by an PCSP and NSERC collaborative grant application, which will be submitted by Denis Lacelle in 2022.
The project has significant infrastructural requirements. First of all, on-site $\delta^{18}O_{ice}$ measurements will be performed using a portable LGR stable isotope analyser available from the University of Ottawa. These analyses are planned to be complemented by stable O/H isotope analyses using laser spectroscopy at the Institute for Geological and Geochemical Research, Hungary. To establish an independent chronology, AMS ^{14}C measurements on DOC will be done at the Paul Scherrer Institute (PSI), Switzerland and the A.E. Lalonde AMS Radiocarbon Laboratory, Canada. Such measurements are challenging, but the Laboratory for Environmental Chemistry of PSI recently pioneered ^{14}C dating of low volume ice samples[37]. The Lalonde Lab also has experience with Artic/polar ice samples having very low DOC contents[60]. Concentrations of ionic species, especially Ca^{2+} and SO_4^{2-}, and the concentration and sizes of aerosols in ice samples will be measured in the Canadian Ice Core Lab (CICL), University of Alberta, which is dedicated to the running of clean polar and glacial ice and snow samples, with instruments specifically configured for this purpose. CICL is led by Alison Criscitiello PhD, who is one of the leading experts of this field and her expertise is crucial in planning and performing ice collection for ionic species and dust measurements.
The Hf-Sr-Nd isotopic fingerprinting of dust sources will be a key part of this project, which will be complemented by clay mineralogy, another powerful dust tracer. The P.I. (G. Ujvari) together with the cooperative partner in Austria (Prof. Urs Klötzli) developed methods, including a new rock decomposition technique and column chemistry setup, for the combined Hf-Sr-Nd isotopic analyses of small size (2-10 mg) dust samples in the frame of a FWF Lise Meitner postdoc project between 2018-2020. This setup and related mass spectrometry were successfully tested with geological reference materials (GRMs) of the USGS[52] and several last glacial dust samples from the NorthGRIP ice core[32]. Samples can now routinely be prepared for isotopic measurements owing to this 2 years research project in Vienna, where the necessary clean laboratory environment and ion exchange chemistry lab is accessible. A TIMS instrument is also available in Vienna for Sr-Nd isotopic analyses, while the Hf isotope measurements are going to be done using a MC-ICPMS at the Institute for Nuclear Research, Debrecen, Hungary. Thus, the cooperation between the Austrian and Hungarian partners is necessary and a prerequisite of the successful completion of this key project part.

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