Geochemical Determination of the pp-Neutrino Flux with ^{205}Tl - LOREX: A Progress Report

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Motivation and Goals of LOREX

The central goal of the LOREX *LOR*andite *EX*periment (1) is the determination of the long-time average (over ~ 4 MY) of the solar neutrino flux Φ_ν with the neutrino-capture reaction (2):

$$^{205}\mathrm{Tl} + \nu_e \rightarrow {}^{205}\mathrm{Pb} + e^- \qquad \ldots (1)$$

As was pointed out originally by Freedman (2), the thallium-bearing mineral lorandite, $TlAsS_2$, from the mine of Allchar, Macedonia. The average flux Φ over the exposure time (age of lorandite since its mineralization) follows from the common activation equation, where σ is the solar neutrino capture cross section and λ the decay constant of ^{205}Pb:

$$\Phi_\nu = N^{-1}\,(T - B)\,(\sigma\varepsilon)^{-1}\lambda\,[1 - \exp(-\lambda a)]^{-1} \qquad \ldots (2)$$

T – totalnumber of ^{205}Pb atoms ; B – background number of ^{205}Pb atoms [^{205}Tl (μp, n) ^{205}Pb] ; λ – decay constant of ^{205}Pb; **ε** – overall detection efficiency; σ – neutrino capture cross section

This renders finally the mean solar neutrino flux, i.e. *the mean luminosity of the sun during the last 4.3 million years*, the geological age of lorandite.

The neutrino-capture reaction: $^{205}\mathrm{Tl} + \nu_e\ (E_{\nu e} \geq 50\ \mathrm{keV}) \rightarrow {}^{205}\mathrm{Pb}^* + e \quad \ldots (3)$ exploits the *by far* ***lowest threshold of $E_{\nu e} \geq 50$ keV*** for (solar) neutrinos.

The central problem of LOREX *is the quantitative determination of ^{205}Pb* atoms in lorandite. Before entering the final phase of the experiment, three fproblems must be reliably addressed:

1. Background, erosion and paleo-depth: The background of ^{205}Pb atoms produced by cosmic radiation and by natural radioactivity must be determined quantitatively. In this context the knowledge of the erosion rate of the overburden rock during the existence of lorandite is of utmost importance.

2. Neutrino capture probability into the 2.3 keV state of ^{205}Pb: The ratio ^{205}Pb/^{205}Tl provides *only the product* of solar neutrino flux and neutrino capture probability into the different nuclear states of ^{205}Pb. However, the capture of neutrinos should populate predominantly the first excited state at E* = 2.3 keV. Hence, to get the neutrino flux itself, one has to determine the capture probability into this low-lying state of ^{205}Pb.

3. Extraction, separation and detection of ^{205}Pb trace concentration: *How can the expected ultra-low abundance of ^{205}Pb be reliably measured.*

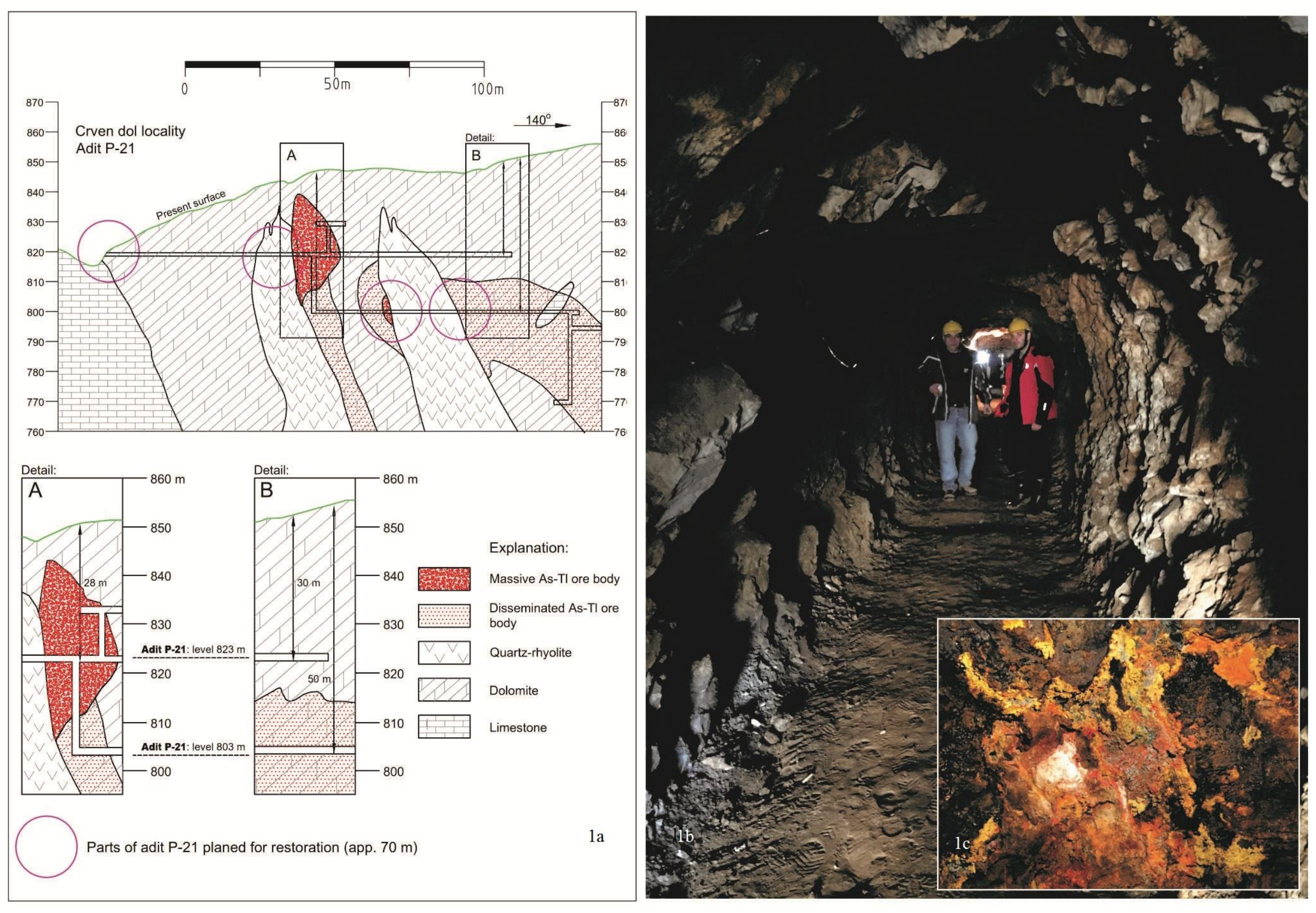

Fig. 1a, **1b and 1c:**1a Geological cross-section of ore body Crven Dol, 1b photograph of corridor in the ore body Crven Dol and 1c lorandite mineralization

A – Recent advances in research on LOREX project

1. Excavation of 10 t of lorandite from ore body Crven Dol (Figure 1a, 1b and 1c)
   a) Lorandite separation: crushing, hand-picking and cleaning of lorandite crystals; obtaining a 98 % pure lorandite.
   b) Quality control of the separated lorandite grains using SEM-EDX and ICP-MS methods.
2. Determination of erosion rate and paleo-depth for ore body Crven Dol (4)
   a) Erosion rate $\mathbf{E_{min}}$= 75 m/Ma (^{3}He, ^{21}Ne), $\mathbf{E_{max}}$= 387 m/Ma (^{26}Al, ^{36}Cl)
   b) Paleo-depth of lorandite from the orebody Crven Dol $\mathbf{d_{min}}$ = 1390 mwe, $\mathbf{d_{max}}$ = 2330 mwe for 4.3 Ma.
3. Determination of contributions ^{205}Pb i.e. underground reaction (3)
   a) Fast muons cosmic rays (see Table 1).
   b) Natural radioactivity (ppm concentration) of U = 0.102 ± 0.010, Th = 0.096 ± 0.075, Bi = 0.008±0.002, Hg = 231±92.
   c) Lead concentration in lorandite **0.311±9.4 ppm**

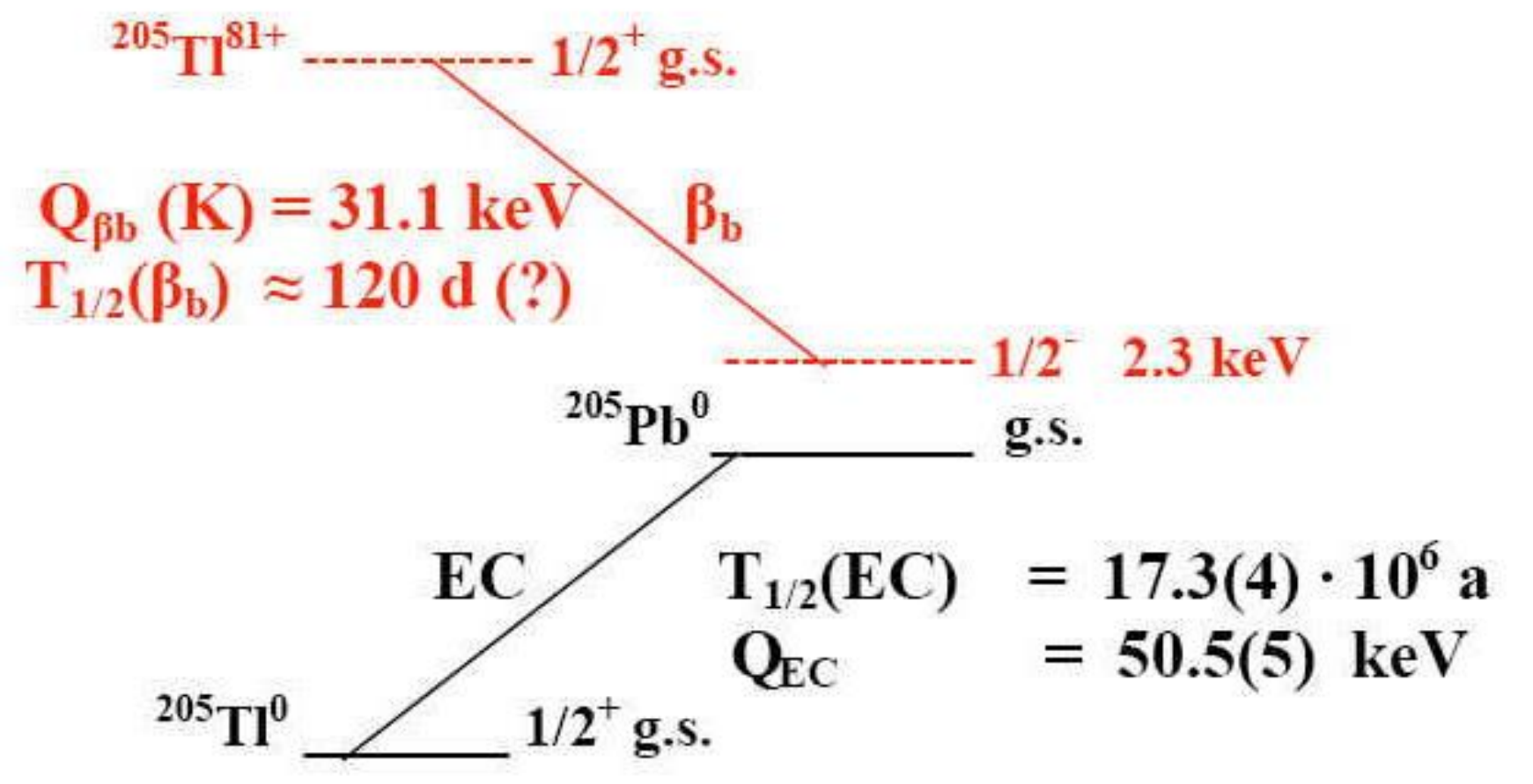

Fig.2. Decay scheme of neutral ^{205}Pb atoms (black) and of bare ^{205}Tl^{81+} ions (red). Whereas neutral ^{205}Pb atoms decay by unique first-forbidden orbital electron capture (EC) from the L and higher electron shells to stable neutral ^{205}Tl atoms with a half-life of 17.3 million years and a Q value of 50.5 keV, bare ^{205}Tl^{81+} (or H-like ^{205}Tl^{80+}) ions can decay to almost 100% by β_b decay to the first excited state of ^{205}Pb-205^{81+} at E*=2.3 keV, where the generated electron will be captured into the K shell (5).

B - Key issues for further research:

1. Neutrino capture probability into the 2.3 keV state of ^{205}Pb:
The ratio ^{205}Pb/^{205}Tl provides *only the product* of solar neutrino flux and neutrino capture probability into the different nuclear states of ^{205}Pb. However, the capture of neutrinos should populate predominantly the first excited state at E* = 2.3 keV. Hence, to get the neutrino flux itself, one has to determine the capture probability into this low-lying state of ^{205}Pb.
2. Extraction and detection of ^{205}Pb trace concentration:
How can the expected ultra-low abundance of ^{205}Pb be reliably measured.

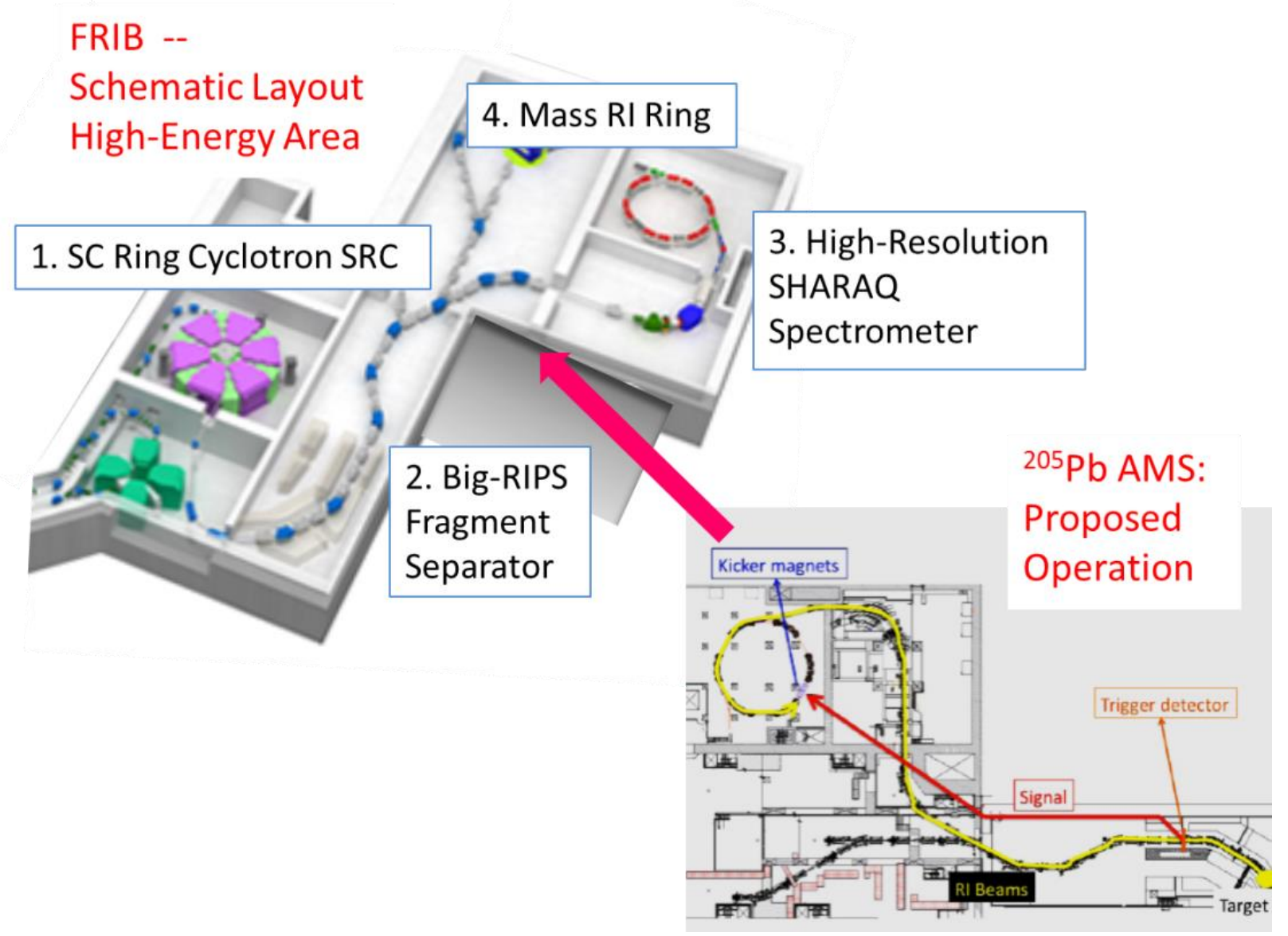

Fig.3. Big-RIPS Separator at RIBF,Nishina Center, RIKEN, Tokyo

1. Determination of the neutrino capture probability into the 2.3 keV state of ^{205}Pb

The difficult measurement of the decay probability of the bare ^{205}**Tl** nucleus to the first excited state of^{205}**Pb**, by the exotic process of **bound-state** beta decay, has been **approved at the Experimental Storage Ring of GSI.** This decay probability **provides the nuclear matrix element for the dominant pp-neutrino capture cross-section which would thus become known** with **sufficient accuracy.**

Table 1. Cosmic ray contribution N (^{205}Pb) $_{\text{fast muons}}$ and N (^{205}Pb) $_{\text{total}}$ (Ref.3)

Depth of location m	**Erosion rate m/Ma**	**Paleo-depth *d* of location (mwe)**	**N (^{205}Pb)$_{\text{fast muons}}$x10^3 (1 kg of lorandite)**	**N (^{205}Pb)$_{\text{total}}$x10^3 (1 kg of lorandite)**	**N (^{205}Pb)$_\nu$ %**	**N (^{205}Pb)$_{\text{fast muons}}$ %**
28	75*	1390	8.1	10.3	21	79
28	387**	2330	1.7	3.9	56	44

*) Min erosion rate (^{3}He, ^{21}Ne) **) Max erosion rate (^{26}Al, ^{36}Cl)

2. Extraction and detection of ultra-low amounts of ^{205}Pb in lorandite

The final steps of LOREX will be the prospection and separation of lorandite from the Allchar mine (Fig. 1), the extraction of thallium and lead (the mean concentration of lead in lorandite amounts to 1.5 ppm) and the quantitative determination of the ratio ^{205}Pb / ^{205}Tl sc. ^{205}Pb / Pb.
After the last step of chemical separation, a lead matrix will be obtained, where the ^{205}Pb/Pb ratio is expected to range from 10^{-14} to $5 \cdot 10^{-13}$. Supposing the value of *146 SNU* for the solar neutrino capture rate, the geological age *a* since the Tl-mineralization is $a = 4.3 \cdot 10^6$ y, the decay probability λ for the electron-capture decay of ^{205}Pb back to ^{205}Tl as $\lambda = 4.68 \cdot 10^{-7}\ y^{-1}$ and a molar mass M of lorandite as M = 343 g / Mol, one gets for the expected time-integrated *number of solar **pp**-neutrino induced ^{205}Pb atoms* the value of:

$$22(7)\ \text{atoms of}\ {}^{205}\mathrm{Pb/g\ lorandite} \qquad \ldots (4)$$

Chemical separation of Pb from Tl in the lorandite sample is expected to produce a ratio of ^{205}Pb/^{205}Tl of about 10^{-13}. The key challenges are therefore Pb isotope separation of the order of 10^{-14} and ^{205}Pb/^{205}Tl isobar separation of 10^{-13}.

The approaches being investigated include:
- Conventional accelerator mass spectrometry (AMS) which provides for the required isotope separation; isobar separation on the basis of characteristic energy loss measurements with particle detectors alone cannot achieve the required level (6). However, combining a gas-filled magnetic separator as a first stage, leading to partial spatial separation of the ion of interest and the interfering isobars, and an advanced energy-loss measurement based on a high-quality passive absorber and high-resolution time-of flight for the second stage, appears a possible option.
- Isobar separation in a high-energy storage ring by full stripping is the most attractive approach; except that it will most likely lead to reduced efficiency compared to conventional AMS
- High-energy accelerator mass spectrometry (AMS) at the RIBF facility at RIKEN, Japan. Increased efficiency might be gained by the novel ion-mass ring at the RIKEN Nishina Center, where upstream identification signals of ions (and ^{205}Pb candidates) are forwarded to a kicker at the entrance of the mass ring proving injection on the central orbit and thus little loss of intensity.

RIBF provides up to 345MeV per nucleon ion-beam energy. The facility provides, first, isotopic and elemental selection via the ion-optical electro-magnetic stages of the accelerator; second, particle separation via Big-RIPS fragment separator augmented by energy and energy-loss measurements in-beam. Finally, full stripping, with more than 50% efficiency at 345MeV, generates charge states 82$^+$/81$^+$ for ^{205}Pb/^{205}Tl. Injection into the newly built mass-ring at RIBF provides for final ^{205}Pb^{82+} identification.

The original suggestion for beam tuning and control was to spike a lead sample with low-concentration (~1%) ^{205}Tl. However, stripping and energy-loss processes in the beam transport generate ^{205}Pb nuclei via (p, n)-type charge exchange reaction. Alternatively, a guide-beam of a lighter chemical element will also be the whole approach then involves 4 steps: i) Establishing beam tuning and control for a trace beam with the 1% ^{205}Tl sample; ii) using the guide beam to confirm ^{205}Tl beam control, for the 1% and a ~10^{-10} Tl sample; iii) extension to a calibration sample with known ^{205}Pb concentration at the 10^{-13} level; iv) measurement of the 205Pb neutrino sample. Test experiments to verify the various aspects of the proposed approach at the RIBF are under development.

Conclusion: Taking into account the present-day state-of-the-art of all the techniques needed to solve the four perennial problems of LOREX, we conclude that it is realistic to expect the first result for the solar pp neutrino flux averaged over the last 4.3 million years in the foreseeable future. This number will have most probably still an error margin in the order of 30% or larger, at the 68% CL. We expect, however, that this accuracy could be improved with time, and that it might reach finally a level below ≤ 30% (3).

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