THE PAST AND THE FUTURE
OF THE \(^{210}\text{Pb} \) METHOD

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NOTE: This letter to the editor compiles and summarizes the main ideas presented and discussed in the talk given in the 2nd NKS/EKO-1 Seminar on Dating of Sediments and Determination of Sedimentation Rate, Helsinki 2-3 April 1997. Relevant publications by the authors and others are given in the list of references.

Previous research on the dating of sediments and determination of sedimentation rates, through the \(^{210}\text{Pb} \) method, did not allow full and proper exploitation of the sedimentation/accumulation dynamics in various aquatic systems. We will give some examples on existing limitations, difficulties and problems in the development of the \(^{210}\text{Pb} \) "method". Poor attention to selection/utilization of experimental facilities and narrow definition of sampling/sample specifications has induced serious consequences concerning the quality of radiometric dating. Sampling strategies and sample pretreatment are conditioned to limited number of existing facilities and do not consider the utilization of alternative tools on the one hand, and possible disturbances through various processes in aquatic systems on the other. The so-called "standard \(^{210}\text{Pb}-\)dating models" are indiscriminately used although they are based on general assumptions that are tested on few soft-water lakes in temperate regions. These facts considerably limit the developments of new radiometric approaches, including dating ones, that consider possible variations of the actual chemical, physical and sedimentological conditions influencing the concentrations and the fluxes of key radionuclides especially \(^{210}\text{Pb} \). Such unfortunate trends ought to be rectified to help proper developments and applications of the \(^{210}\text{Pb} \) method, particularly in combination with other nuclides, on a wider scale.
General introduction

The chronology of environmental and climatic changes in atmospheric and aquatic systems is essential for the understanding of dynamic processes forcing various exchanges in land-water systems. The $^{210}$Pb method can be used for the dating of recent deposits, and mass-balance studies of aquatic systems, of the past 150-175 years. This is very helpful for the evaluation of environmental and climatic impacts produced by human activities.

Previous research on the $^{210}$Pb dating of sediments and related determination of the sedimentation rates did not allow full and proper exploitation of the sedimentation dynamics in various aquatic systems. The $^{210}$Pb dating method has generally been used as a routine tool to solve problems for uncritical users. Although the complexity of aquatic systems this method has been applied to single cores with the smallest number of samples. Poor attention to selection/utilization of experimental facilities and insufficient definition of sample specification has induced serious consequences concerning the quality of the $^{210}$Pb dating. We believe that appropriate $^{210}$Pb routines ought to be based on research. Special efforts need to be devoted to improve the $^{210}$Pb dating reliability and related quality aspects. Good accuracies and agreement between alternative/complementary tools as well as extension of the upper dating limit have to be considered to enhance the quality and reliability of the $^{210}$Pb method. It is essential to use proper experimental facilities for accurate determination of $^{210}$Pb, in addition to other relevant nuclides, in different sediment layers. $^{210}$Pb could be determined by direct gamma-ray spectrometry through its weak gamma-emission at 46.5 keV (4.25%) and via alpha-particle spectrometry of its grand-daughter $^{210}$Po. $\beta$-counting of the daughter $^{210}$Bi is also a possibility in some cases where time constrains make $^{210}$Po less attractive. These alternatives have various specifications, advantages and limitations. Their combination might, therefore, be imperative in many cases.

Gamma-ray Spectrometry

No chemical separations are needed. This tool allows simultaneous determination of $^{210}$Pb, $^{226}$Ra and other relevant U/Th radionuclides for the correct application of the $^{210}$Pb method. Other artificial radionuclides like $^{137}$Cs and $^{241}$Am, that could be determined by gamma spectrometry, are extremely useful to validate/evaluate the $^{210}$Pb ages. $^7$Be is also an interesting cosmogenic ra-
dioisotope that is suitable for isotope-particle interactions in aquatic systems. Gamma-ray spectrometry has a relatively high low limit of detection (LLD) in comparison with alpha-particle spectrometry. Well-type detectors are suitable for samples with high specific activities whereas low-energy, planar and coaxial Ge detectors could be used for low specific activities where compensation by mass allows measuring bigger samples. However utilization of big samples may produce less temporal resolutions. Large sample sizes can be considered appropriate at high sedimentation rates such as in marine sediments or the like. The low energy of the $^{210}$Pb gamma-ray photons imply, in addition to geometrical sample considerations, self-absorption corrections. Self-absorption corrections are not an easy task as they ought to be based on experimental studies and for every sample/detector configuration.

**Alpha-particle Spectrometry**

The very low LLD of alpha-particle spectrometry permits the measurement of weak $^{210}$Pb activities and could be applied to small samples (0.3 - 0.5 grams or less). The application of alpha spectrometry is specially suitable at very low and moderate sedimentation rates. The application of alpha spectrometry is essential either in cases where good accuracies are required or for the older sediments where the total $^{210}$Pb is slightly above the levels of the $^{226}$Ra. Alpha spectrometry has special importance in speciation studies, that are important in rehabilitation strategies, where gamma spectrometry can not be used unless detailed chemistry is involved. Radiochemical procedures are necessary for the application of isotope dilution. Utilization of $^{209}$Po (4.832 MeV) as internal yield tracer instead of $^{208}$Po (5.114 MeV) has induced drastic simplification of the radiochemical procedure because its alpha-peak can be very well resolved from the $^{210}$Po (5.305 MeV). However, information on other radionuclides are needed through additional techniques/methods.

Gamma and alpha spectrometries are complementary techniques and ought not to be considered as competitive ones. Combination of alpha/gamma spectrometries and their alternative application to different layers is, therefore, the best approach. $\beta$-counting could be of value in some cases. In gamma spectrometry optimization of sample sizes to detector specifications is quite important for the best utilization of this tool. Chemical procedures used in alpha spectrometry ought to be conditioned to sample composition. Proper evaluation
of the supported $^{210}\text{Pb}$ through $^{226}\text{Ra}$, as indicator of the supported $^{210}\text{Pb}$ should, therefore, be critically considered.

**Sampling and sample pretreatment**

Quality and accuracy of the radiometric data are very much dependent on sampling strategies, quality and specification of the sampling sites. Traditional sampling in aquatic systems are generally reduced to the collection/measurement of single cores. Some lakes, for example, were/are heavily utilized for various independent monitoring/research programmes and high risks exist for disturbed sediments. Coordination of existing activities and proper records on sampling programmes in lakes are imperative as the credibility of lakes as indicators is extremely important for future studies especially those related to follow-up and rehabilitation issues. Spatial distribution of key radionuclides in lake bottoms is quite necessary to understand the behaviour and sedimentation dynamics, through mass-balance studies, of direct and indirect atmospheric inputs. Additional information that allow the selection/development of the most suitable $^{210}\text{Pb}$ data/models could be obtained through studies over the whole lake basin. Limited samples and amounts of material should not cause constrains on the applicability of alternative models. Lack of limnological data such as water content, organic content and grain-size in different layers may limit the possibilities to carry out various site-related, sample-related and model-related corrections/normalizations. These are examples on problems that ought to be considered via well-organized coordination between samplers, analytical teams, modelers and managers of land-water resources.

**Alternative models**

Another critical step in the $^{210}\text{Pb}$ method is the application of appropriate models as defined by site and sediment specifications rather than forcing few models to all sites and conditions of sedimentation. Two models: the C.I.C. (Constant Initial Concentration) and C.R.S. (Constant Rate of Supply) are indiscriminately used for the construction of chronologies. These models are based on general assumptions that have been tested in few soft-water lakes situated in temperate regions. These models ought not to be considered as "standard $^{210}\text{Pb}$ models" as they are not verified on global scale and additional corrections/normalizations steps are not involved in these models. Unfortu-
nately the application of these models without careful consideration of the specificity of sites/regions has considerably limited the development of technical and dating approaches on wider scales. New corrections/normalizations, that consider actual sedimentological, biological, chemical and physical conditions influencing the concentrations and fluxes of $^{210}\text{Pb}$, are expected in the future. It must be emphasized that there are no standard or universal $^{210}\text{Pb}$ dating models that could be valid for all possible cases since by definition models can only be good or bad mathematical representations of reality. Unless strong evidences exist to show the constancy of temporal and spatial fluxes of the $^{210}\text{Pb}$ the credibility of the C.R.S., in its general and simple form, could be questioned. It is, therefore, recommended that independent alternatives ought to be used to validate the reliability and accuracies of the $^{210}\text{Pb}$ dates.

In cases where the $^{210}\text{Pb}$ flux is a site-related parameter, on spatial and temporal scales, rather than a constant quantity it would be very informative to know various processes influencing it as this would help the development of new applications. Coastal marine sites, such as fjords, estuaries and semi-arid lakes with variable input levels from land could be critical sites for dating.

**Alternative and complementary dating techniques**

Alternative and independent dating tools used to validate the $^{210}\text{Pb}$ dating can be classified to radiometric, geophysical and isotopic approaches. None of these tools, if applied separately, can serve as a general tool for climatic and environmental studies as they only provide partial solutions to specific issues. $^{137}\text{Cs}$ records have been served to provide chronological information through peak values of main atmospheric inputs. $^{241}\text{Am}$, if measured with good accuracies, provides suitable alternative in cases where $^{137}\text{Cs}$ suffers from geochemical mobility as is usually the case in peatlands and lake-sites with high water content and/or low sedimentation. Other problems that complicate the utilization of $^{137}\text{Cs}$ is $^{137}\text{Cs}$ inputs from point sources. Exact information on input functions from point sources are generally lacking as in marine sites suffering from mixed inputs from different sources such as those occurring in the North Sea and the Baltic Sea. Long residence time caused by poor sedimentation of $^{137}\text{Cs}$ in water bodies, such as in the Baltic Sea, may cause further limitations in the utilization of $^{137}\text{Cs}$. $^{239+240}\text{Pu}$ is also as good alternative as $^{241}\text{Am}$, however the measurement of $^{239+240}\text{Pu}$ is limited to alpha spectrometry which is a
rather difficult task because of its low concentrations \( ^{239+240}\text{Pu}/^{137}\text{Cs} \) activity ratio in fallout is about \( 10^{-4} \). Unlike these radionuclides \(^{90}\text{Sr} \) is a very conservative environmental isotope and its utilization, that necessitates low-level beta counting, is only limited to ice-deposits. The sedimentological behaviour of \(^{90}\text{Sr} \) under different geochemical and limnological conditions is not properly studied yet.

In addition to the construction of high-resolution chronologies through radiometric tools there are potential applications to explore various settling and sedimentation dynamics of particulate matter in water-bodies. "Within-system" and "between-system" interactions in lakes, or the like, could be detailed through combined radiometric measurements using sediment traps and sediment cores. Differentiation between these interactions has very interesting applications in rehabilitation studies especially if management policies are to be based on the specific conditions of the land-water resources.

References


