Assessment of historic flood plain sedimentation rates along the River Meuse in The Netherlands using ¹³⁷Cs dating with PHAROS

MARJOLEIN VAN WIJNGAARDEN

Ministry of Transport, Public Works and Water Management, Institute for Inland Water Management and Waste Water Treatment (RIZA), PO Box 52, 3300 AB Dordrecht, The Netherlands e-mail: m.vwijngaarden@riza.rws.minvenw.nl

CATHERINE RIGOLLET & ROB J. DE MEIJER

Nuclear Geophysics Division, Kernfysisch Versneller Instituut, University of Groningen, Zernikelaan 25, 9747 AA Groningen, The Netherlands

Abstract Deposition of fine sediments during high river discharges is an important factor in flood plain evolution. To assess the effect of river restoration works and climate change on sediment dynamics, a solid understanding of the present and historic sedimentation rates is required. Overbank sedimentation is a process which varies both in time and space, therefore accurate data for sedimentation rates at various locations within a flood plain are required. Episodic ¹³⁷Cs deposits offer a way to document rates of overbank deposition over the past 50 years. A laboratory-based device, PHAROS, was designed to help date sediment cores by non intrusively measuring the intensity of the gamma rays emitted from the decay of ¹³⁷Cs in 2 cm increments. From a flood plain along the River Meuse (The Netherlands) 23 sediment cores of 1 m length were obtained for PHAROS analysis. From the observed ¹³⁷Cs peak depths sedimentation rates were estimated to vary from 0.4 to 2.0 ± 0.2 cm year⁻¹. The PHAROS derived ¹³⁷Cs activity concentrations were calibrated with a regular technique (HPGe). In addition lead isotopes were used to validate the ¹³⁷Cs-derived time records.

Key words dating; caesium-137; lead-206/lead-207 ratio; tracers; overbank deposition; flood plains

INTRODUCTION

Over the past decade concern about the morphological and ecological condition of the Dutch rivers Rhine and Meuse has increased. To fulfil both nature conservation and safety objectives, a variety of landscaping measures have been proposed to improve the discharge capacity of the high-water floodway. Most types of measures will increase both the discharge and the suspended sediment loads on the flood plain, eventually leading to an increase in sedimentation rates. Moreover, climate change is expected to enhance sedimentation as a result of increases in both the magnitude and frequency of occurrence of high discharge events. To present reliable estimates for flood plain sedimentation rates in the future, a thorough understanding of past and present flood plain sedimentation is required.

Overbank deposition of fine sediments occurs during periods of high discharges. The amount of flood plain sedimentation within one discharge event depends on local flood plain topography (Asselman, 1998; Middelkoop, 1997), vegetation cover (Walling *et al.*, 1992) and sediment availability (Asselman, 1998). The mean annual sedimentation, considered over decades, is also influenced by flood frequency, since large rare events can influence the long-term average. In The Netherlands, mean annual rates of recent overbank deposition are within the range of 0–20 mm year⁻¹. Moreover, it has been illustrated that in view of irregularities of the deposition process in time and space special attention should be paid to the spatial variability in sedimentation rates on a flood plain scale (Leenaers, 1991; Middelkoop, 1997; Asselman, 1998).

One approach to investigate flood plain sedimentation rates involves the use of radioactive tracers for dating sediments, which can provide estimates of long-term average sedimentation rates. Using the activity–concentration profile of ¹³⁷Cs or ²¹⁰Pb, sedimentation rates over the past 50–100 years can be established (Sugai *et al.*, 1994; Walling & He, 1998; Radakovitch *et al.*, 1999). Where ²¹⁰Pb is a naturally occurring radionuclide, ¹³⁷Cs is a fallout radionuclide brought into the atmosphere by atomic bomb experiments between the 1950s and 1960s; the first significantly high levels of ¹³⁷Cs were detected in 1954, while a peak intensity was reached in 1963. Since the early 1980s ¹³⁷Cs levels have been very low, although the 1986 Chernobyl accident is responsible for a second fallout peak in parts of Europe, including The Netherlands. By linking peak intensities in the ¹³⁷Cs-profile to the corresponding years of fallout the average annual sedimentation rates can be estimated. To apply the ¹³⁷Cs dating technique, traditionally sediment samples from cores are analysed in the laboratory using hyper-pure germanium detectors (HPGe), which is time consuming and costly.

Given the applicability to a wide range of environments the use of 137 Cs was pursued in the light of recent advances in the sensitivity of gamma-ray detector systems (De Meijer, 1998; Hendriks *et al.*, 2001). Based on this technology, PHAROS (<u>Pluri-detector</u>, <u>High-resolution</u>, <u>Analyser of Radiometric properties Of Soil cores</u>) was designed to help date sediment cores. The 137 Cs-activity concentration profile is derived by measuring and analysing gamma-ray spectra for each 2 cm of the core. With PHAROS, the hoisting of the core and data storage of the gamma-ray spectra is an automated process (Rigollet & de Meijer, in press).

In this paper the feasibility of ¹³⁷Cs-based dating with PHAROS is discussed, based on cores obtained from a flood plain situated along the southern part of the Meuse in The Netherlands. PHAROS-derived ¹³⁷Cs-activity concentrations were validated by a traditional (HPGe) analysis. Lead-isotope ratios were used to check the ¹³⁷Cs-derived time records.

FIELD STUDY

Overbank deposition was studied at a flood plain situated along the southern part of the Meuse in The Netherlands: the Border Meuse (Fig. 1(a)), literally the border between The Netherlands and Belgium. The Meuse is The Netherlands' second largest river, originating in the northeast of France from where it travels 935 km, losing 409 m of elevation, before finally flowing into the North Sea nearby Rotterdam (Fig. 1(a)). The



Fig. 1 (a) The River Meuse in The Netherlands. (b) Sampling locations at the flood plain of Itteren and Borgharen.

Meuse is a rainfed river, with a mean discharge of 230 m³ s⁻¹. Peak flows occur during the winter period. The recorded maximum discharge is 3052 m³ s⁻¹ (Middelkoop & Van Haselen, 1999). In Fig. 1(b) the study site around the villages of Itteren and Borgharen as situated in The Netherlands is presented. Inundation commences when the discharge exceeds 1300 m³ s⁻¹, with inundation times in the low-lying areas near the river of the order of several days/year.

At the coring locations shown in Fig. 1(b), 10-cm-diameter sediment cores were collected. A mobile drilling system was used, which samples cores in 1-m sections by mechanically drilling a steel core lining into the ground while the sediment is captured in an inner tube. Visual examination of the cores revealed that the sediment within the core had remained undisturbed. In June 1999 duplicate cores were sampled at locations 1160 and 1161. Cores of 2.0 m (1160) and 2.40 m (1161) length were used to calibrate PHAROS against the standard technique (HPGe) and to assess the accuracy of ¹³⁷Cs dating in flood plain sediments. In July 2000, 21 cores of 1 m length were taken for PHAROS analysis to obtain more information on the spatial variation in sedimentation rates.

At the time of sampling in 1999, PHAROS was still under construction so one of each of the duplicate cores of 1160 and 1161 was stored for one year before PHAROS analysis, while the remaining cores (also one for both 1160 and 1161) were opened, visually inspected and sliced in 3-cm samples. From each core 25 samples were selected for ¹³⁷ Cs (HPGe) and ²⁰⁶Pb/²⁰⁷Pb ratio analysis. The ²⁰⁶Pb/²⁰⁷Pb ratio can be used to differentiate between different sources of natural and anthropogenic lead. The introduction of leaded petrol in the early 1950s resulted in a significant decrease in the lead isotope ratios (Fachetti, 1989; Walraven *et al.*, 1997) and provides a clear time marker. A selection of the 21 cores sampled in July 2000 were opened and samples were taken for HPGe analysis to validate the PHAROS results; in total 125 samples were taken from 10 cores.

ANALYTICAL TECHNIQUES

Before analysis all samples were dried at 60°C, homogenized and stored in glass containers.

Lead isotopic analysis

For lead isotopic analysis 100–250 mg of sample was treated with 4 ml 16M HNO₃, 2 ml 29M HF and 2 ml 12M HClO₄. Samples were heated to 240°C under 110 bars for 55 min in a closed microwave system and cooled afterwards. In the final solutions, made up to 50 ml with 4.5% HNO₃, ²⁰⁴Pb, ²⁰⁶Pb, ²⁰⁷Pb and ²⁰⁸Pb were measured with a VG Plasmaquad PQ 2 ICP-MS. To obtain maximum precision the samples were analysed 10 times; the resulting isotopic ratios have an accuracy better than 0.5%.

Four distinct stages can be differentiated (Walraven et al., 1997):

- (a) the pre-industrial period before 1860, with a ²⁰⁶Pb/ ²⁰⁷Pb ratio between 1.175 and 1.190;
- (b) the start of the early industrialization in the first half of the twentieth century with a ratio between 1.170 and 1.185;
- (c) the introduction of leaded petrol in the early 1950s with a strongly decreasing ratio towards 1.140;
- (d) the present situation with a 206 Pb/ 207 Pb-ratio between 1.160 and 1.165.

Although local variations may occur, the sequence as described should in general be present in Dutch soils. Therefore these stages will be closely investigated in the study cores to provide a time frame to compare with the ¹³⁷Cs results.

Radiometric analysis

The radiometric composition of sediments was carried out with a high-purity germanium detector (HPGe). The activity concentration of ¹³⁷Cs is directly determined from the intensity of its 662 keV gamma ray; the activity concentrations of ²³⁸U and ²³²Th follow from the weighted average of the intensities of several gamma-rays emitted by their daughter radionuclei. Samples were put in sealed polystyrene boxes, kept for three weeks to ensure secular equilibrium between the daughter nuclei, and subsequently placed on top of a HPGe (high-purity germanium) detector. The HPGe detector is an EG&G Ortec p-type germanium detector, with crystal dimensions: $\Phi = 60.4 \text{ mm}$, h = 78.3 mm. The efficiency of the detector is calibrated with an aqueous solution of manmade radionuclides and is corrected for the density and volume of the sample.

RADIOMETRIC DATING USING PHAROS

Figure 2 shows PHAROS: a 3-m-high steel frame with three platforms to keep detectors and shielding. A sediment core is transported downwards by a stepper motor coupled to a 2.5-m-long spindle. In this investigation two bismuth germanate oxide (BGO) detectors were used for the radiometric measurement. They are situated in the





Fig. 2 The PHAROS framework (*left-hand* side) and the detectors in a lead shielding (*right-hand* side).

upper part of the frame, shielded from background radiation by 10-cm-thick lead blocks (see insert in Fig. 2). The detectors are positioned on opposite sides of the core, offset 2 cm in height with respect to each other. Scanning a 1 m core in 2 cm increments took 48 h. To determine the activity concentrations of individual radio nuclides from gamma ray spectra obtained by PHAROS, the background activity was investigated and the system was calibrated using cores of standard geometry and known density and activity. PHAROS derives the ⁴⁰K, ²³²Th, ²³⁸U and ¹³⁷Cs activity concentrations from the measured gamma-ray spectra using spectrum deconvolution (De Meijer, 1998; Hendriks *et al.*, 2001). Refer to Rigollet & de Meijer (in press) for a detailed description of PHAROS.

Sedimentation rates were determined for the periods 1963-1986 and 1986-2000 for those cores with two distinct Cs peaks in the profile. The rates were derived from the accumulated sediment by the time markers corresponding to the top (date of collection) and the position of the Cs peaks. Moreover, the sedimentation rate for the period 1954-2000 was estimated by the initial increase of 137 Cs from a background value of zero.

SEDIMENTATION RATES ON THE ITTEREN AND BORGHAREN FLOOD PLAIN

Reliability of ¹³⁷Cs dating with PHAROS

Visual inspection of cores 1160 and 1161 revealed homogeneous very fine, silt-rich sediment throughout both cores without noticeable layering. A gravel bed was reached

at the end of both cores. The ¹³⁷Cs profiles of both the PHAROS and the HPGe for core 1160 are presented in Fig. 3, in which an excellent agreement between PHAROS and the HPGe for the upper peak ($19 \pm 2 \text{ cm}$ depth) can be observed. A fit to the PHAROS data, based on a Lorenz approximation of the curve, is presented to indicate the peaks more clearly; the two peaks were fitted separately. A similar agreement between PHAROS and HPGe data was obtained for the other cores. The presence of the second peak at $60 \pm 2 \text{ cm}$ in the 1160 core only came to light when the PHAROS results became available, proving that the initial estimate for the HPGe sampling-depth up to only 50 cm had clearly been inadequate. The PHAROS profile also presents the initial increase in ¹³⁷Cs levels (1954) to be located at $80 \pm 10 \text{ cm}$. Sedimentation rates were found to be $1.7 \pm 0.1 \text{ cm year}^{-1}$ for 1954–2000. Based on these results it was concluded that PHAROS is a comparable method to HPGe detection for ¹³⁷Cs activity concentrations.



Fig. 3 Depth distribution of 137 Cs in core 1160; both PHAROS and HPGe results are presented. The time scale on the second *y*-axis is based on a peak analysis of the PHAROS profile.

The ${}^{206}\text{Pb}/{}^{207}\text{Pb}$ results for both cores are presented in Fig. 4, with text labels to indicate the several stages of consistent lead isotopic ratios. Although sedimentation rates are clearly different, the ${}^{206}\text{Pb}/{}^{207}\text{Pb}$ ratios of both cores show a similar trend. In Fig. 5 the results for both the ${}^{206}\text{Pb}/{}^{207}\text{Pb}$ and ${}^{137}\text{Cs}$ for core 1160 are compared. For core 1160 the introduction of leaded petrol occurs at 75 ± 5 cm and corresponds to an average sedimentation rate for the period 1953–2000 of 1.6 ± 0.1 cm year⁻¹. This value is within the range of the value for the same period from the ${}^{137}\text{Cs}$ profile (1.7 ± 0.2 cm year⁻¹). For core 1161 the introduction of leaded petrol shows up at 45 ± 5 cm, which corresponds to a sedimentation rate of 0.9 ± 0.1 cm, which is also in the range of the set of 0.8 ± 0.1 cm year⁻¹ from the ${}^{137}\text{Cs}$ profile. From the detailed analysis of these



Fig. 4 Distribution of 206/207 stable lead isotopes in cores 1160 and 1161. The separate time periods of constant isotopic composition are indicated along the dotted line, which indicates the trend in time.



Fig. 5 Depth profile of both the 137 Cs activity (PHAROS data) and the stable lead isotope composition in core 1160.

two cores it can be concluded that the ¹³⁷Cs dating technique using PHAROS provides a time scale and values of sedimentation rates in agreement with the ²⁰⁶Pb/ ²⁰⁷Pb-ratio technique.

Spatial variation in historic flood plain sedimentation rates

From the ¹³⁷Cs profiles obtained from the 21 cores taken in both August 1999 and July 2000, nine cores with two distinct peaks were selected. Additionally seven of the remaining 14 cores did not show these two peaks, but showed very clearly the 1954 increase in ¹³⁷Cs. In Table 1 the estimated sedimentation rates for the periods 1954–2000, 1963–1986 and 1986–2000 are summarized. Generally the various time markers could be observed with an accuracy of several centimetres, which corresponds to an average accuracy in sedimentation rates in the order of 0.1–0.2 cm year⁻¹. Sedimentation rates range from 0.4 to 2.0 cm year⁻¹, which are fairly high for Dutch flood plains, but are realistic figures nonetheless (Asselman, 1998; Middelkoop, 1997). However, no consistent pattern could be observed in the evolution of the sedimentation rates with time, which was an incentive for further analysis.

Factors which control overbank deposition such as inundation frequency and sediment availability were hence included in the assessment of the sedimentation rates. In a first step the height a.m.s.l. for each location was linked to the corresponding Meuse discharge stage using well-established Q-H (discharge to height) relationships. The inundation frequency for each sampling location was then derived from a combination of this information with the frequency of occurrence of these discharges stages. Finally, the sediment availability for each location was estimated by combining data from a long-term daily record of suspended matter concentrations measured at

Core number	Sedimentation rate 1986–2000 (cm year ⁻¹)	Sedimentation rate 1963– 1986 (cm year ⁻¹)	Sedimentation rate 1954–2000 (cm year ⁻¹)
1160	1.4	1.7	1.7
1161	1.1	0.4	0.8
1259			1.1
1260	1.1	0.5	1.1
1263	1.3	1.7	1.5
1265	0.7	0.7	0.7
1266	1.7	1.2	1.4
1267	1.3	1.0	1.2
1268			1.0
1269	1.4	1.1	1.1
1270	2.0	0.9	1.4
1271			1.4
1272			0.7
1273	0.9	0.4	0.8
1276			0.7
1277			0.9

Table 1 Sedimentation rates derived from the 137 Cs profiles. The uncertainty in the values is estimated to be of the order of 0.1–0.2 cm year⁻¹.

Borgharen with the discharge at the moment of inundation. However, no relationship could be observed between either inundation frequency or sediment availability and the estimated sedimentation rates. A possible explanation for the lack of such a correlation may be found in the fact that local hydraulic factors significantly influence sediment deposition and should be taken into account when assessing the spatial variability in sediment deposition. In order to make accurate predictions of flood plain deposition it is essential that future research should study the processes during transport and deposition in more detail.

CONCLUSIONS

From this study it can be concluded that:

- PHAROS offers a reliable method for the estimation of ¹³⁷Cs activity concentrations and thus sedimentation rates.
- The use of ¹³⁷Cs is a reliable method to date flood plain sediments; sedimentation rates based on ¹³⁷Cs and the ²⁰⁶Pb/ ²⁰⁷Pb ratio have identical values, within the limits of determination.
- The data illustrate that the spatial variability in sedimentation rates over a flood plain is high and that relatively simple correlations with inundation or sediment supply are insufficient to fully understand the process of sediment deposition.

REFERENCES

Asselman, N. E. M. (1998) Suspended sediment transport in the River Rhine. The impact of climate change on erosion, transport and deposition. PhD Thesis, Utrecht University, Utrecht, The Netherlands.

Fachetti, S. (1989) Lead in petrol. The isotopic lead experiment. Acc. Chem. Res. 22, 370-374.

Hendriks, P. H. G. M., Limburg, J. & De Meijer, R. J. (2001) Full spectrum analysis of natural gamma ray spectra. *J. Environ. Radioactiv.*. **53**, 365–380.

Leenaers, H. (1991) De Geul: Nederlands grootste bron van zware metalen voor de Maas (in Dutch). Milieu 1, 12–19.

De Meijer, R. J. (1998) Heavy minerals: from "Edelstein to Einstein". J. Geochem. Explor. 63, 81-103.

- Middelkoop, H. (1997) Embanked floodplains in The Netherlands. Geomorphological evolution over various time scales. PhD Thesis, Utrecht University, The Netherlands.
- Middelkoop, H. & Van Haselen, C. O. G. (1999) *Twice a River: Rhine and Meuse in The Netherlands*. RIZA Report no. 99003, Arnhem, The Netherlands.
- Radakovitch, O., Charmasson, S., Arnaud, M. & Bouisset, P. (1999) ²¹⁰Pb and caesium accumulation in the Rhone Delta sediments. *Estuar., Coast. Shelf Sci.* **48**, 77–92.
- Rigollet, C., & de Meijer, R. J. (in press) Pluri-detector, High-resolution Analyser of Radiometric properties of Soils. Submitted to Nucl. Instrum. Methods A.

Sugai, S. F., Alperin, M. J. & Reeburgh, W. S. (1994) Episodic deposition and ¹³⁷Cs immobility in Skan Bay sediments: a ten year ²¹⁰Pb and ¹³⁷Cs time series. *Mar. Geol.* **116**, 351–372.

- Walling, D. E. & He, Q. (1998) Use of the fallout of ¹³⁷Cs in investigations of overbank sediment deposition on river floodplains. *Catena* 29, 263–282.
- Walling, D. E., Quine, T. A. & He, Q. (1992) Investigating contemporary rates of floodplain sedimentation. Lowland floodplain rivers. In: *Lowland Floodplain Rivers: Geomorphological Perspectives* (ed. by P. A. Carling & G. E. Petts), 165–184. John Wiley, Chichester, UK.
- Walraven, N., Van Os, B. J. H., Klaver, G. Th., Baker, J. H. & Vriend, S. P. (1997) Trace element concentrations and stable lead isotopes in soils as tracer of lead pollution in Graft-De Rijp, The Netherlands. J. Geochem. Explor. 59, 47–58.