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## Radionuclide deposition in the Rhône River Prodelta (NW Mediterranean sea) in response to the December 2003 extreme flood

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### Abstract:

The extreme Rhône River flood that peaked in early December 2003 with water discharges as high as  $11,500 \text{ m}^3 \text{ s}^{-1}$ , induced major damage in southern France and transferred large amounts of radionuclides that were associated with suspended particulate matter such as  $^7\text{Be}$ ,  $^{137}\text{Cs}$  and  $^{210}\text{Pb}$ . Specific marine locations in the close vicinity of the Rhône River Mouth were sampled three times in December 2003, February 2004 and April 2004 in order to quantify the consequences of flood events in the Rhône River Prodelta and to investigate the sedimentary depositional patterns through time. The  $^{210}\text{Pb}_{\text{xs}}$  profile analyses in sediment revealed the existence of two particulate matter inputs, one that was related to the December extreme flood and the other to a peak flood of  $4000 \text{ m}^3 \text{ s}^{-1}$  of water discharge that occurred in January 2004. The December event net deposition was assessed at  $75 \pm 19 \text{ GBq}$  of  $^{210}\text{Pb}_{\text{xs}}$  and  $27 \pm 2 \text{ GBq}$  of  $^{137}\text{Cs}$ . The net deposition in response to the January 2004 flood event, although less damaging, is of the same order of magnitude to the December 2003 deposition, i.e.  $71 \pm 21 \text{ GBq}$  of  $^{210}\text{Pb}_{\text{xs}}$  and  $16 \pm 5 \text{ GBq}$  of  $^{137}\text{Cs}$ . The sedimentary records of the December flood exhibited from the cores collected in December 2003, February 2004 and April 2004 are similar arguing for good environmental signal preservation through time but, the standard deviation of flood deposition estimate increases with time after the December flood event indicating a loss of accuracy with time.

**Keywords:** radionuclide deposition; extreme flood; Rhône River Prodelta; Gulf of Lions

## INTRODUCTION

The Rhône River is known to be the main river of the Western Mediterranean Sea and induces a large transfer of suspended particulate matter and associated radionuclides to the Gulf of Lions and the Western Mediterranean Sea when flood events occur. It is characterized by a 7.6 millions T.yr<sup>-1</sup> mean solid discharge and a 1700 m<sup>3</sup>.s<sup>-1</sup> mean water discharge (300-3000 m<sup>3</sup>.s<sup>-1</sup>; Pont et al., 2002) with a large annual variability in response to snow melting or autumn rains that could induce flood events. Such events play a major role in the annual sediment budgets (Walling et al. 1992; Picouet et al., 2001). Indeed, in Mediterranean area, sudden and large floods have a major action on both the sedimentary fluxes and the geomorphological evolution of the river bed (Serrat et al., 2001; Pont et al., 2002); for example, October 1993 flood event, which spent 24 days, delivered 10.7 million tons of suspended particulate matter to the Mediterranean Sea, the 5 days flood of January 1994 drove 6.3 millions tons (Pont, 1997) and the large November-December 2002 flood, 7.6 millions tons i.e. a 88 % of the annual particulate flux (Rolland et al., 2004a). The riverine inputs to the marine systems have been largely studied through marine sediment investigations with some relevant proxy as heavy metals (Ferrand et al., 1999; Bertolotto et al., 2005) or organic matter (Accornero et al., 2003; Miltner et al., 2005). Previous investigations on radionuclides as <sup>137</sup>Cs and <sup>210</sup>Pb allowed constraining sedimentary patterns over centennial time scale in agreement with their respective particulate affinity (Zuo et al., 1991; Zuo et al., 1997; Miralles et al., 2005). Unfortunately, Rhône River floods occur at very short time-scales, from day to week that can not be efficiently studied by geochemical tracers such as <sup>137</sup>Cs and <sup>210</sup>Pb owing to their respective half-lives. Thus, <sup>7</sup>Be (t<sub>1/2</sub>=53 d) investigations were carried

out on prodelta sediments in order to evidence the thickness of sediment deposited during the December extreme flood and the induced  $^{137}\text{Cs}$  and  $^{210}\text{Pb}_{\text{XS}}$  inventories.

This work presents a characterization of the consequences of the extreme Rhône flood event of December 2003; it deals with the  $^{210}\text{Pb}$  and  $^{137}\text{Cs}$  riverine fluxes and sediment inventories determined in the Rhône River Prodeltà area and quantifies the retention of radionuclide in prodelta deposits that can act as a secondary source of sediment and contaminants.

## SETTINGS

### *Rhône River dynamics*

The Rhône River drains a 98000 km<sup>2</sup> wide watershed along its 832 km length in France. South of Arles (50 km from the river mouth), it splits into two main channels, each with a different importance; the eastern arm, Grand Rhône, is 52 km long and encompasses 90% of the total river water flux while the western arm -Petit Rhône- is 62 km long and represents the remaining 10%. The annual average water discharge of the Rhône River is 1700 m<sup>3</sup>.s<sup>-1</sup> and the annual suspended particulate discharge varies from 1 10<sup>6</sup> T.yr<sup>-1</sup> to 26.5 10<sup>6</sup> T.yr<sup>-1</sup> (Antonelli, 2002). Taking into account the bottom load transport, 77% to 91% of the total suspended particulate matter load transit under flood conditions (Pont and Bombled, 1995; Rolland et al., 2004b).

The Rhône River is also a point source for the transfer of artificial radionuclides released by the Marcoule reprocessing plant and the weathering of the watershed contaminated by global fallout that are discharged in the river. Previous studies on radionuclide transfers revealed that 86% of the  $^{137}\text{Cs}$  is bounded on less than 450 nm particles (Eyrolle and Charmasson, 2004). In 1991,  $^{137}\text{Cs}$  delivery to the Western Mediterranean Sea was 19.6 Tbq which 40% were trapped in the prodelta area (Charmasson, 2003).

### *Sedimentation conditions at the Rhône River Mouth*

The Rhône River discharge is spread over the continental margin by a benthic nepheloid layer system (Aloisi et al., 1982; Monaco et al., 1990) and superficial plume deflected by external conditions (mainly wind forcing) as shown by numerous modeling studies (Estournel et al., 1997; Marsaleix et al., 1998; Estournel et al., 2001; Ulses et al., 2005).

The Rhône prodelta is a 30 km<sup>2</sup> area in the vicinity of the Rhône River mouth extending off the deltaic plain (Aloisi et al., 1975; Boldin et al., 1988; Durrieu de Madron et al., 2000). It is characterized by high organic carbon content (1-2%), silty mud sedimentation (Durrieu de Madron et al., 2003) and the highest sediment accumulation rates at the margin scale (Miralles et al., 2005).

### *December 2003 flood episode*

In December 2003, an exceptional flooding episode occurred in the Rhône River, with maximum water discharge reaching 11500 m<sup>3</sup>.s<sup>-1</sup> on the 3 December at Beaucaire (70 km upstream the river mouth, where the Rhône is still in one channel) – the highest ever recorded for this river. The recurrence time of such an event is nearly 100 years (Consensus Conference, 2005). The water discharge increased from 2400 m<sup>3</sup>.s<sup>-1</sup> to 11500 m<sup>3</sup>.s<sup>-1</sup> in less than 30 hours. The flood was induced by a rainy storm episode across the entire south-eastern region of France caused by the collision between cold air masses and warm humid air masses coming from the Mediterranean Sea. This event was particularly impressive because of its length - it lasted more than 48 hours, instead of the more typical 24-36 hours. In Marseilles, precipitations reached 150 mm in 24 hours at a time when monthly averages are usually in the order of 50-70 mm. The consequences of this catastrophic flood event included over 500

km<sup>2</sup> of flooded land in the Rhône River low-valley and several deaths. The city of Arles was submerged under 16 millions m<sup>3</sup> of floodwater. Here, the water discharge significantly increased from the 1<sup>st</sup> December to peak at 10 000 m<sup>3</sup>.s<sup>-1</sup> on the 3<sup>rd</sup> December followed by a steady decrease until the 20<sup>th</sup> December 2003(fig. 1).

Throughout this period, the water discharge remained in excess of the mean Rhône River water discharge (fig. 1). Suspended matter concentration ([SPM] in mg.l<sup>-1</sup>) range between 76 mg.l<sup>-1</sup> corresponding to the Rhône River off-flood event period suspended load to more than 3600 mg.l<sup>-1</sup> at its maximum (Antonelli et al., 2006). Indeed, this flood event induced the transfer of 5.4 10<sup>6</sup> T of suspended particulate matter (SPM) towards the sea during the 1<sup>st</sup> -7<sup>th</sup> December period.

Radionuclides associated to the Rhône River solid discharges delivered to the Gulf of Lions reached 99±22 Gbq of <sup>210</sup>Pb<sub>xs</sub> and 77±16 Gbq of <sup>137</sup>Cs (respectively Rolland et al., 2004b and Antonelli et al., 2006). Out of any flood period, <sup>210</sup>Pb<sub>xs</sub> activity associated to the SPM is constant at 60 Bq.kg<sup>-1</sup>. It decreased to 20 Bq.kg<sup>-1</sup> when water discharge peaked out on the 3<sup>rd</sup> December (Eyrolle et al., 2006a; Eyrolle et al., 2006b). The <sup>137</sup>Cs content remains constant at 12 Bq.kg<sup>-1</sup> all along the year without flood considerations (Antonelli et al., 2006).

## SAMPLING AND METHODS

Six sediment cores (20-40 cm length) were collected during an EUROSTRATAFORM cruise on the 16<sup>th</sup> December 2003, just after the extreme December 2003 Rhône River flood. Analyses of specific radionuclides (<sup>7</sup>Be, <sup>137</sup>Cs, <sup>210</sup>Pb) were performed in order to quantify the particulate flux to Rhône River prodelta in response to specific flow conditions. <sup>7</sup>Be (t<sub>1/2</sub>=53.2 d), a naturally occurring radionuclide resulting from the cosmic ray spallation of nitrogen and carbon in the atmosphere, was analyzed in order to clarify recent particulate deposition (up to 200 d). <sup>7</sup>Be reaches the Earth's

surface bounded on detritical particles and is delivered to marine systems with riverine inputs (Dibb and Rice, 1989; Canuel et al., 1990) depending both on the particulate composition and salinity (Bloom and Crecelius, 1983; Baskaran et al., 1997). Palinkas et al. (2005) showed that the use of  $^7\text{Be}$  to assess flood deposition has to be assessed by sedimentological variable analyses.

$^{137}\text{Cs}$  ( $t_{1/2}=30.1$  yrs) is an anthropogenically-derived radionuclide. It entered the Environment in response to atmospheric nuclear device tests from 1954 to 1962 that induce a global fallout. Others sources are accidental human releases as Chernobyl accident in April 1986 that have local influences (Anspaugh et al., 1988) and authorized releases from nuclear power plants. In freshwater,  $^{137}\text{Cs}$  has a high affinity for the clay (Rogowski et Tamura, 1970) while in seawater, the ion competition generates its releases in dissolved phase. This mechanisms are also responsible of the post depositional mobility of  $^{137}\text{Cs}$  in marine sediments (Radakovitch et al., 1999; Frignani et al., 2004). Indeed,  $^{137}\text{Cs}$  analyses are justified by its toxicity to the whole Environment.

$^{210}\text{Pb}$  ( $t_{1/2}=22.3$  yrs) is a naturally occurring radionuclide produced in the atmosphere by  $^{222}\text{Rn}$  decay which binds on submicron atmospheric particles (Gillette et al., 1972). It is deposited onto the Earth's surface by wet and dry deposition processes. Owing to its specific half-life,  $^{210}\text{Pb}$  is useful to assess centennial sedimentation rates in marine systems (Miralles et al., 2005). However, in case of river floods the  $^{210}\text{Pb}$  analysis can be used to evidence any particle depositions in sediments.

Sampling sites were chosen around the River Mouth between 20 m and about 80 m depth (fig. 3; table 1). The cores were sub-sampled in 1 cm thick layers. Each layer was dried, crushed, passed through a 2 mm sieve and conditioned in 200 ml and 20 ml geometries for gamma spectrometry investigations. An aliquot is preserved for grain-size feature characterization at LERCM, La Seyne-sur-mer (IRSN, France) on a

Coulter-Beckman LS 13 320 laser grain-sizer. This was not performed on all studied cores but focused on cores we assumed imprinted by flood.

Radionuclides activity are measured part at CEREGE (Centre Européen d'Enseignement et de Recherche des Géosciences de l'Environnement, Université P. Cézanne – CNRS, France) where gamma spectrometry is performed on 20 ml volume geometry samples using a semi-planar intrinsic Germanium detector. The detector is calibrated by counting sediment standards of known activity. The other non-destructive gamma spectrometry are undergone at IRSN Orsay with N-type hyper-purity germanium detectors in 200 ml volume containers and measured with a counting time of 20 or 40 h. Efficiency calibrations from 22.5 keV to 1.8 MeV were carried out using mixed gamma-ray sources in a solid resin-water equivalent matrix of 1.15 g.cm<sup>-3</sup> density. Activity results are corrected for true coincidence summing and self-absorption effects (Lefèvre et al, 2003). <sup>7</sup>Be and <sup>137</sup>Cs activities are obtained from the 477.7 KeV and 671 KeV photopeaks respectively. Excess <sup>210</sup>Pb (<sup>210</sup>Pb<sub>xs</sub>) activities are determined by subtracting the measured <sup>214</sup>Pb activity (352 KeV photopeak) as an indicator of supported <sup>210</sup>Pb to the total <sup>210</sup>Pb peak (46.5 KeV photopeak). The activity uncertainty (at k=2) is calculated by standard propagation of calibration source uncertainty, the statistic counting uncertainty (sample and background) and the summing and self-absorption correction uncertainties.

## RESULTS AND DISCUSSION

### *Flood deposition*

Several investigations proved <sup>7</sup>Be to be a good tracer of flood deposition in coastal zones (Sommerfield et al., 1999; Mullenbach and Nittrouer, 2000). Nevertheless, Palinkas et al. (2005) evidenced an underestimation of the deposited layer using only <sup>7</sup>Be as proxy related to the grain-size changes. In order to assess the relevance of <sup>7</sup>Be

as flood deposition proxy in this work, we determined grain-size changes in each investigated cores. The grain-size distribution (d50) permits to exhibit any changes in deposition conditions and to overview the whole flood consequence for the sediment column.

Among the six prodelta sediment cores,  $^7\text{Be}$  is not detected in BF08 and it is only found in the superficial sediment layer (1 cm depth) in BF13. Regarding the other cores,  $^7\text{Be}$  was detected at maximum depths ranging from 5 cm to 18 m indicating a large variability of sedimentary record for this area (fig. 4; table 2). The core BF05 presents a discontinuous  $^7\text{Be}$  profile down core with two peak values at 1 cm and 10 cm depth while  $^7\text{Be}$  was still detected at 18 cm depth. The profile shape could be attributed to mixing processes affecting older material deposited for more than ~200 days and recently deposited material disturbing the  $^7\text{Be}$  signal. Other explanation is that high water discharges caused re-suspension of old material stocks deposited onto the Rhône River bed removed by river floods and deposited in the prodelta area, as suggested Thomas (1997). Better understanding could be provided by comparison with  $^{234}\text{Th}$  penetration depth. Unfortunately, we did not measure the  $^{234}\text{Th}$  activity down core due to his short half life ( $t_{1/2}=24.1$  d). However, comparisons with grain size distribution allows to assess deposition condition changes down cores.

Grain-size distributions are assessed by medium diameter (d50) distribution down core (fig. 5). In core BF05, the d50 baseline is  $14\pm 4$   $\mu\text{m}$  while it peaks at 65  $\mu\text{m}$  at 7 cm depth. Nevertheless, even if this peak can be associated to high energy deposition, it can not be obviously related to the  $^7\text{Be}$  distribution. The BF05 grain size baseline is higher than other baseline that range from 6 to 9  $\mu\text{m}$  (table 3) that is synonymous of high energy conditions resulting in coarser material deposition. In core BF06 and BF07, d50 ranges from about 10  $\mu\text{m}$  to 30-40  $\mu\text{m}$  with respective baselines of  $7\pm 3$   $\mu\text{m}$  and  $9\pm 1$   $\mu\text{m}$  (table 3).



In BF06, where  $^7\text{Be}$  occurs until 11 cm depth,  $d_{50}$  increases constantly from 18 cm depth to the top with a high  $d_{50}$  value (36  $\mu\text{m}$ ) at 6 cm depth. In BF07,  $d_{50}$  peaks twice at 1 cm and 17 cm depth while  $^7\text{Be}$  only occurs until 5 cm depth. We can thus assume that the 17 cm depth  $d_{50}$  rise is caused by an old flood. The core BF09, where  $^7\text{Be}$  occurs until 9 cm depth, exhibits a low  $d_{50}$  variability, from 4  $\mu\text{m}$  to 19  $\mu\text{m}$ . The  $d_{50}$  increases from 12 cm depth to the top with higher  $d_{50}$  in the 0-7 cm depth layer and a 4 cm depth peak at 19  $\mu\text{m}$ . Indeed, this argues for a general deposition energy increase. Nevertheless, the grain size increase begins at 12 cm depth while  $^7\text{Be}$  occurs until 9 cm depth. Thus, the use of  $^7\text{Be}$  as unique tracer of flood event is doubtful as evidenced Wheatcroft et al. (2006).

In BF13 core,  $d_{50}$  distribution does not evidenced any drastic energy condition variations with a quite constant shape. This station doesn't seem to be affected by any recent flood events. Recurring investigations on benthic macro fauna at this site showed that Rhône River floods were followed by blooms of opportunistic species taking advantage of recent high organic carbon deposition (Salen-Picard et al., 2003). Nevertheless, this phenomenon was not recorded in December 2003 (C. Salen-Picard, *Pers. Comm.*), demonstrating the weak influence of the flood event at this location. Indeed, the main wind direction was SSE during the November-December period, according to the ALADIN<sup>®</sup> meteorological modeling studies (C. Estournel, C. Ulses, *Pers. Comm.*). This experiment shows a general trend of down welling along the coast, caused by elevated surface water levels, and an anticlockwise westward coastal current that pasted the Rhône River plume onto the coast (Marsaleix et al., 1998; Estournel et al., 2001).

Unfortunately, the grain size distribution assessment does not allow neither evidencing flood layer thickness nor confirming  $^7\text{Be}$  efficiency as flood tracer.

Definitively, we will not use the  $^7\text{Be}$  as flood layer proxy but as recent particulate matter deposition.

#### *$^{210}\text{Pb}_{\text{xs}}$ and $^{137}\text{Cs}$ vertical distribution and inventories*

In cores where  $^7\text{Be}$  were detected, an obvious feature is the distribution of  $^{210}\text{Pb}_{\text{xs}}$  and  $^{137}\text{Cs}$  through the sediment column, despite various emissions patterns in the environment and biogeochemical behaviors (fig. 6). While  $^{137}\text{Cs}$  post-depositional mobility has been proven in marine sediments (Radakovitch et al., 1999; Frignani et al., 2004),  $^{210}\text{Pb}$  is not labile once bound to a particle and deposited onto the seafloor. Thus, the extension of the  $^{210}\text{Pb}_{\text{xs}}$  profile all over the cores suggests a large particulate flux episode. The  $^{210}\text{Pb}_{\text{xs}}$  profiles do not exhibit clear radioactive decay trends preventing any attempt of geochronology assessment.

$^7\text{Be}$  and  $^{210}\text{Pb}_{\text{xs}}$  have similar geochemical behaviors in sediments but their respective half-lives vary from several orders of magnitude, from day to year. Despite these respective features, both radionuclide are detected at similar depths in sediments. This argues to a sediment signal forcing by grain size.

The  $^{210}\text{Pb}_{\text{xs}}$  activity distributions exhibit specific sediment layers characterized by an activity drastic decrease upwards. These decreases are assumed to be dilution phenomena in response to large particulate matter supply (Dominik et al., 1987; Miralles et al., 2004). We assume that the beginning of the decrease in surface layers is related to the onset of a flood, and that the minimum radionuclide activity value marks the maximum flood solid discharge or the flood peak; indeed, for both cores BF 05 and BF 06 the  $^{210}\text{Pb}_{\text{xs}}$  activity decreases from  $60 \text{ Bq.kg}^{-1}$  (at 18 cm and 6 cm depth respectively) to less than  $20 \text{ Bq.kg}^{-1}$  at 5 cm and 2 cm depth. For cores BF 07 and BF 09, the  $^{210}\text{Pb}_{\text{xs}}$  activity decreases from  $60 \text{ Bq.kg}^{-1}$  (at 5 cm and 2 cm depth respectively) to close to  $40 \text{ Bq.kg}^{-1}$  at the surface. Therefore,  $^{210}\text{Pb}_{\text{xs}}$  activities

measured in cores BF 05 and BF 06 are in agreement with activities associated to suspended particulate matter measured in the Rhône River at Arles (Rolland et al., *submitted*). Thus, we assume these cores strongly imprinted by flood deposition.

In cores BF 07 and BF 09, the lower  $^{210}\text{Pb}_{\text{xs}}$  activity values assumed to label the day of maximum water discharges are still higher than  $20 \text{ Bq.kg}^{-1}$ , typical of riverine SPM during high water discharge periods, that relates to a weaker dilution process and may not reflect the flood event with accuracy. However, it is difficult to accurately determine the outcome of a flood on a radionuclide profile and the induced radionuclide inventory; hence, we decided to assess the flood deposition into a large range defined by two assumptions: low or high deposition.

The low deposition assumption considers radionuclide deposition occurs from the flood start to the flood peak - i.e. low activity value, while the "high deposition scenario" assumes a deposition which begins with the flood and ends when  $^{210}\text{Pb}_{\text{xs}}$  activity reaches the values of  $^{210}\text{Pb}_{\text{xs}}$  associated to the SPM transfer in the Rhône River ( $60 \text{ Bq.kg}^{-1}$ ). When several layers of dilution phenomena exist in a sediment core, the deeper sequence layer is, the older flood event like in cores BF 06 and BF 09 (fig. 6; table 4). Indeed, the deeper dilution features, whose lower  $^{210}\text{Pb}_{\text{xs}}$  values at 13 cm and 12 cm depth respectively, are expected to correspond to the November 2002 flood. The thicknesses of the layers that correlate to dilution processes vary between 18 cm in core BF 05 to 3 cm in the BF 09 core (fig. 6; table 4). The corresponding  $^{210}\text{Pb}_{\text{xs}}$  inventories vary between  $1370 \pm 192 \text{ Bq.m}^{-2}$  and  $4625 \pm 393 \text{ Bq.m}^{-2}$ , and  $^{137}\text{Cs}$  inventories between  $462 \pm 19 \text{ Bq.m}^{-2}$  and  $1890 \pm 40 \text{ Bq.m}^{-2}$ . Nevertheless, the core BF 09 shows a  $^{210}\text{Pb}_{\text{xs}}$  shape of decreasing radionuclide activity layer from a depth of 3 cm to the sediment surface, but lacks the expected inflexion point to mark the flood peak. Thus, the flood deposition scenario cannot be determined from this core: consequently, the  $^{210}\text{Pb}_{\text{xs}}$  and  $^{137}\text{Cs}$  inventories were partial inventories that were not

taken into account in the mean flood deposition inventories estimate in this area. Taking this into account, mean  $^{210}\text{Pb}_{\text{xs}}$  and  $^{137}\text{Cs}$  inventories corresponding to the December flood event are estimated to be  $2518 \pm 628 \text{ Bq.m}^{-2}$  and  $893 \pm 62 \text{ Bq.m}^{-2}$  respectively. At the prodelta scale ( $30 \text{ km}^2$ ), these inventories correspond to the deposition of  $75 \pm 19 \text{ GBq}$  of  $^{210}\text{Pb}$  and  $27 \pm 2 \text{ GBq}$  of  $^{137}\text{Cs}$ , or, respectively, 76% of the total  $^{210}\text{Pb}_{\text{xs}}$  particulate flux and 35% of the total  $^{137}\text{Cs}$  particulate flux measured in the Rhône River (Antonelli et al., 2006). This is in agreement with the estimate of the  $^{137}\text{Cs}$  trapped in sediments of the Rhône River Prodelt area determined by Charmasson (2003), i.e. 40 % related to the prodelta sediment high clay content (e.g. Durrieu de Madron et al., 2000).

#### *Temporal variations of sediment deposition rates*

In order to assess changes in the sedimentation patterns through time, core samples were collected at site BF05 three times, in December 2003, February 2004 and April 2004, both  $^{137}\text{Cs}$  and  $^{210}\text{Pb}_{\text{xs}}$  activities were measured (fig. 7). The dilution features related to the December 2003 flood are present in each samples at various depths. In the December 2003 core the dilution feature corresponding to the flood is located between 5 to 18 cm depth. The same feature is found between 10 to 18 cm depth in the February 2004 core and between 18 to 25 cm depth in the April 2004 core. From December 2003 to February 2004, the imprint of the December 2003 flood was progressively buried by 5 cm over this two month-period for a  $2.5 \text{ cm.month}^{-1}$  sediment accumulation rate. Instead of actual deepening of the dilution feature between December 2003 and February 2004, it is probably more accurate to suggest compaction as a primary mechanism creating this feature, since the decrease of the  $^{210}\text{Pb}_{\text{xs}}$  activity is 13 cm long in the December 2003 core and 8 cm in the February 2004 core. Furthermore, a new dilution feature appears in the February 2004 core

between 2 and 6 cm depth that can be related to the increase of the Rhône River water discharges up to  $4000 \text{ m}^3 \cdot \text{s}^{-1}$  in January 2004; this event is also evident in the April 2004  $^{210}\text{Pb}_{\text{xs}}$  shape between 5 cm and 9 cm depth, deepened by 3 cm over a two month-period implying an apparent sediment accumulation rate of  $1.5 \text{ cm} \cdot \text{month}^{-1}$ , i.e. 2 orders of magnitude higher than the centennial sediment accumulation rates measured by  $^{210}\text{Pb}_{\text{xs}}$  method (Miralles et al., 2005).

#### *Time evolution of radionuclide inventories*

The estimated  $^{210}\text{Pb}_{\text{xs}}$  and  $^{137}\text{Cs}$  low and high inventories for the February 2004 and April 2004 cores were determined according to the same assumptions as the December 2003 inventories (table 5). The estimated mean  $^{210}\text{Pb}_{\text{xs}}$  inventories in sediments are similar in the December 2003, February 2004 and April 2004 cores: they range between  $4275 \pm 495 \text{ Bq} \cdot \text{m}^{-2}$  in the December 2003 core to  $2897 \pm 1488 \text{ Bq} \cdot \text{m}^{-2}$  in the April 2004 core. The mean  $^{137}\text{Cs}$  inventories are also similar through time: they range from  $1631 \pm 366 \text{ Bq} \cdot \text{m}^{-2}$  in the December 2003 core to  $1110 \pm 526 \text{ Bq} \cdot \text{m}^{-2}$  in the April 2004 core.  $^{210}\text{Pb}_{\text{xs}}$  distribution in these cores revealed a dilution feature in the first centimeters of the February 2004 and April 2004 cores. These are probably related to a water discharge increase that occurred in January 2004, with maximum value reaching  $4\,000 \text{ m}^3 \cdot \text{s}^{-1}$  in Arles (fig. 7). Induced  $^{210}\text{Pb}_{\text{xs}}$  and  $^{137}\text{Cs}$  deposition are estimates at  $2375 \pm 713 \text{ Bq} \cdot \text{m}^{-2}$  and  $537 \pm 153 \text{ Bq} \cdot \text{m}^{-2}$  in the February 2004 core, or  $71 \pm 21 \text{ GBq}$  of  $^{210}\text{Pb}_{\text{xs}}$  and  $16 \pm 5 \text{ GBq}$  of  $^{137}\text{Cs}$  at the prodelta scale, similar to the extreme December 2003 flood net deposition (table 4). In fact, maximum water discharge value was lower than the December 2003 peak but remained high for a longer time period (one day higher than  $3000 \text{ m}^3 \cdot \text{s}^{-1}$  in December 2003 and about one week in January 2004).

## CONCLUSIONS

The December 2003 Rhône River flood, which peaked on 3 December, was an extreme event characterized by water discharges that reached  $11500 \text{ m}^3\cdot\text{s}^{-1}$  in Beaucaire (70 Km upstream) and about  $10000 \text{ m}^3\cdot\text{s}^{-1}$  in Arles, where flood damages were substantial. Despite high difficulty to define the flood layer, the assumption of two various deposition scenarios allowed us to determine the range of flood particulate deposition. Radionuclide depositions related to this event are  $75\pm 19 \text{ GBq}$  of  $^{210}\text{Pb}_{\text{xs}}$  and  $27\pm 2 \text{ GBq}$  of  $^{137}\text{Cs}$ , about 76% and 35% of the annual radionuclide Rhône River budget.

The comparison between sedimentary radionuclide inventories in cores collected at different 2-month lap time (December 2003, February 2004 and April 2004) showed that the flood record ( $^{137}\text{Cs}$  and  $^{210}\text{Pb}_{\text{xs}}$  inventories) did not change with time. However, the standard deviation increased with time and reduced the accuracy of the radionuclide particulate flux estimates.

These investigations also allowed quantification of particulate flux inputs related to an increase of the Rhône River water discharges in January (up to  $4000 \text{ m}^3\cdot\text{s}^{-1}$ ), which induced a net deposition of  $^{210}\text{Pb}_{\text{xs}}$  and  $^{137}\text{Cs}$  in a same order of magnitude as the extreme December 2003 flood. We emphasize that prodelta sediment can be used as a relevant proxy of the riverine short time-scale events.

Nevertheless, it has to be taken into account that these estimates of Rhône River flood event were based on a too few number of sediment cores to hope accurately defined the sedimentary dynamics in such extreme condition at the whole prodelta scale. Improvements have still to be done on mapping of the flood layer off shore to accurately assess the response of the Continent-Ocean transitional area when Rhône River extreme events occur.

### *Acknowledgements*

This study was supported by the European project EUROSTRATAFORM (Number EVK3-CT-2002-00079) and IRSN research program. The authors thank the crews aboard the N/O Suroît (IFREMER), N/O Antedon II (INSU), and N/O Téthys II (INSU) and Serge Berné for December 2003 cruise organization. The authors thank the CNR (Compagnie Nationale du Rhône) for the Rhône River discharge data. Authors would also thank the reviewers that contributed to improve the quality of this manuscript.

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### *Figure captions*

Figure 1: Rhône River water discharges ( $\text{m}^3\cdot\text{s}^{-1}$ ) measured at Arles (50 km upstream the river mouth) during the 01/01/2001-01/03/2004 period.

Figure 2: Rhône River water discharges ( $\text{m}^3\cdot\text{s}^{-1}$ ) and associated suspended particulate matter concentration ([SPM] in  $\text{mg}\cdot\text{l}^{-1}$ ) in Arles (from Antonelli et al., 2006).

Figure 3: Sampling sites in the close vicinity of the Rhône River Mouth.

Figure 4:  $^7\text{Be}$  activity ( $\text{Bq}\cdot\text{kg}^{-1}$  dry weight) in cores collected in December 2003.

Figure 5: Medium diameter  $d_{50}$  ( $\mu\text{m}$ ) distribution in cores collected in December 2003.

Figure 6:  $^{210}\text{Pb}_{\text{xs}}$  and  $^{137}\text{Cs}$  activities ( $\text{Bq}\cdot\text{kg}^{-1}$  dry weight) distribution in sediment cores where  $^7\text{Be}$  analyses exhibited recent particulate deposition. Error bars correspond to  $\pm 2$  s.  $^{137}\text{Cs}$  activity error bars are included in the point.

Figure 7:  $^{210}\text{Pb}_{\text{xs}}$  and  $^{137}\text{Cs}$  activities ( $\text{Bq}\cdot\text{kg}^{-1}$  dry weight) at BF05 sampling site (a) in December 2003, (b) February 2004 and (c) April 2004. Error bars correspond to  $\pm 2$  s.  $^{137}\text{Cs}$  activity error bars are included in the point.

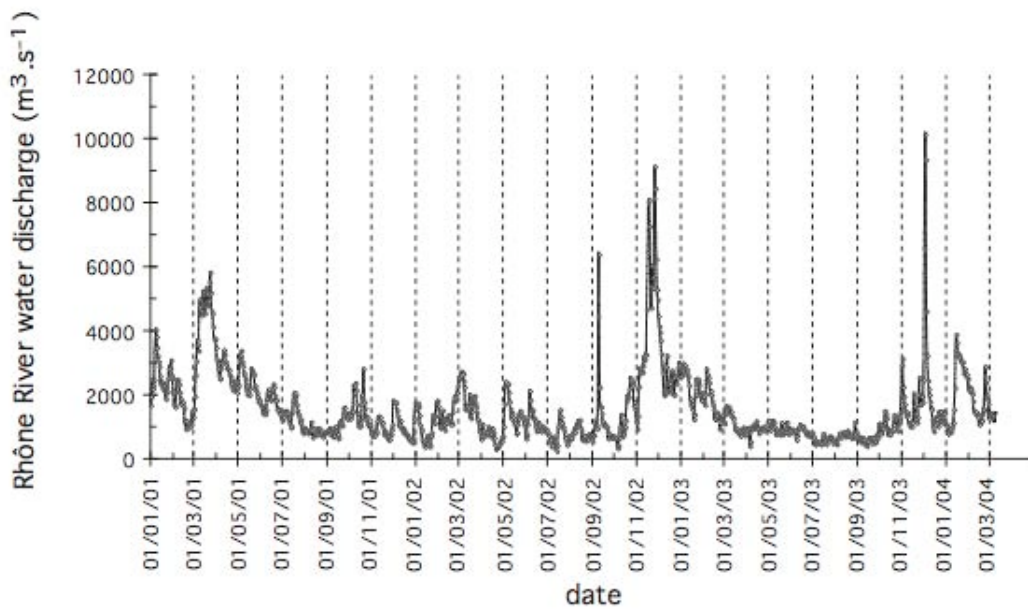


Figure 1: *Miralles et al.*

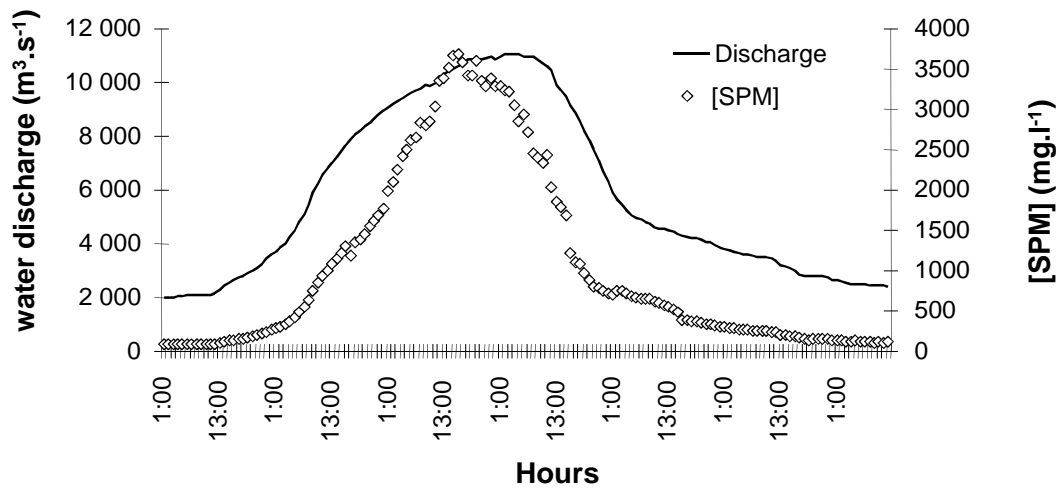


Figure 2: *Miralles et al.*

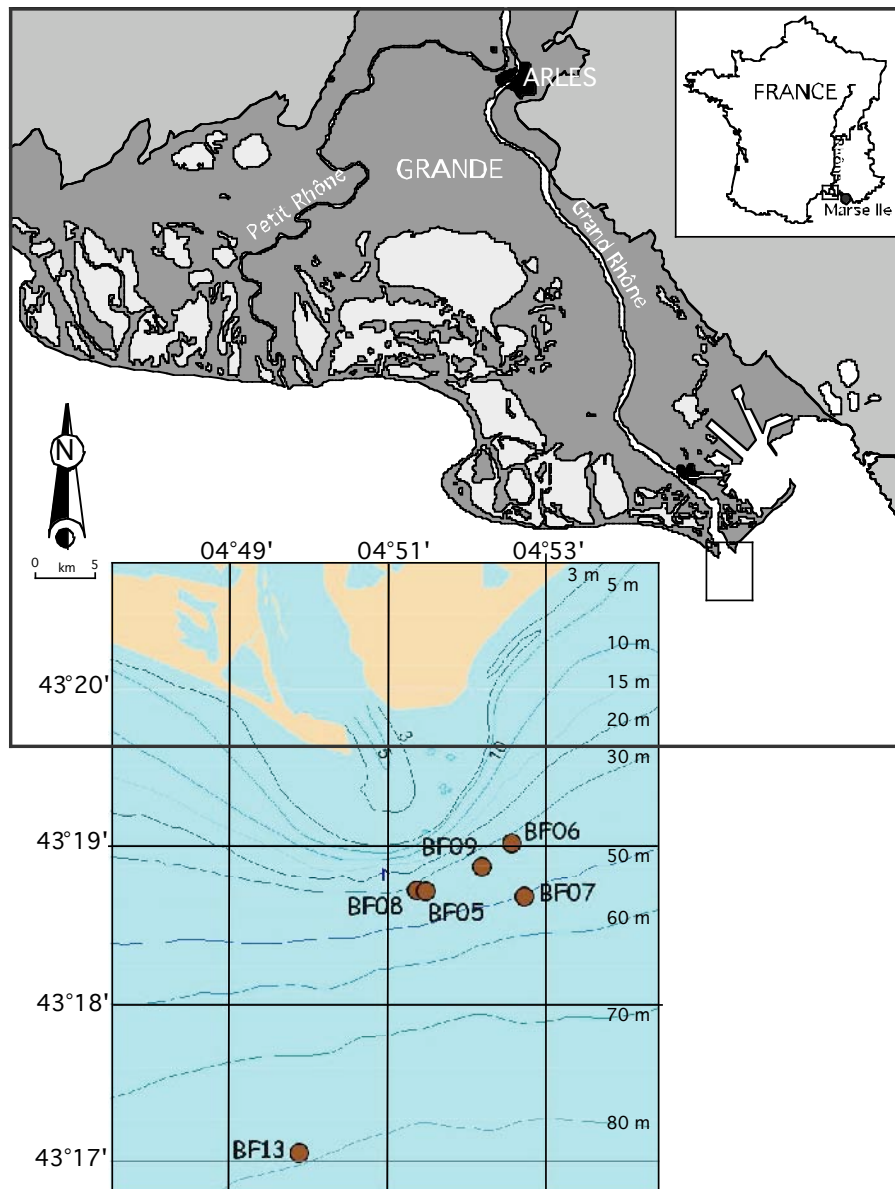


Figure 3: *Miralles et al.*

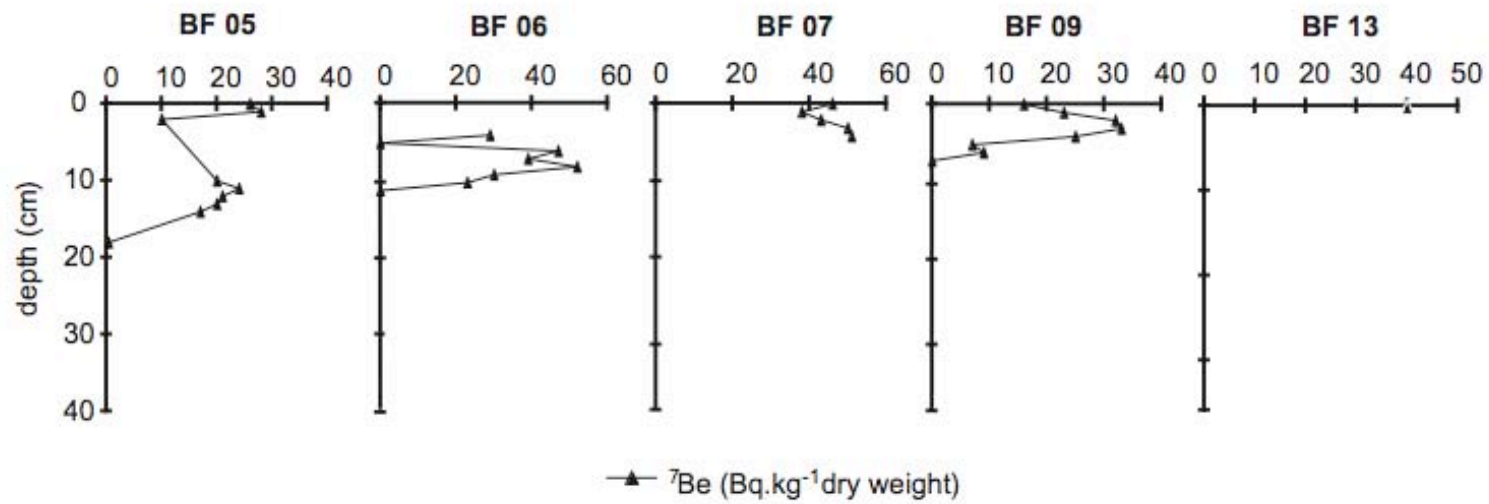


Figure 4: *Miralles et al.*



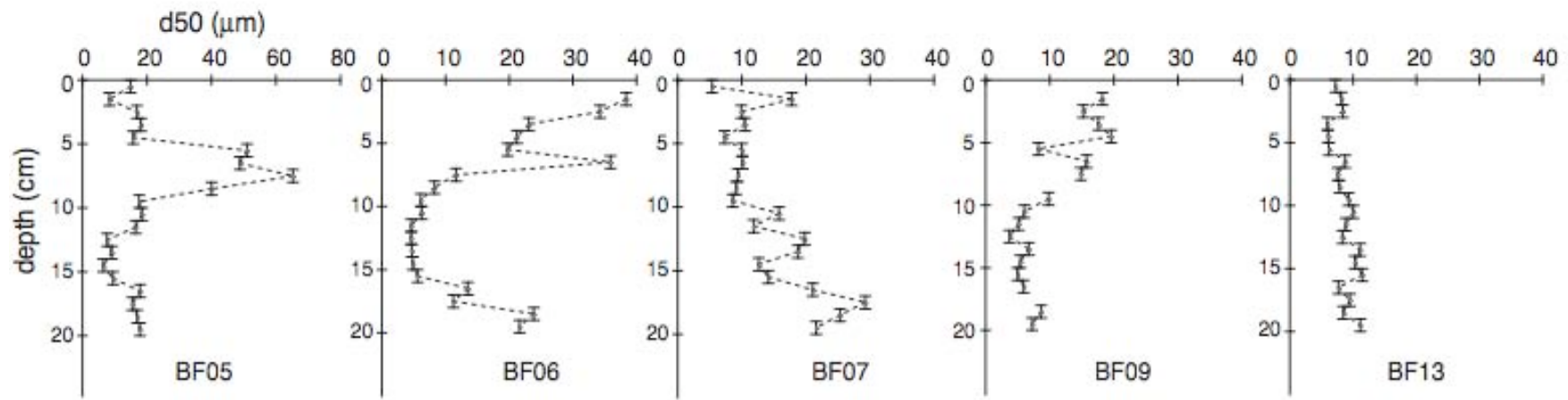


Figure 5: *Miralles et al.*

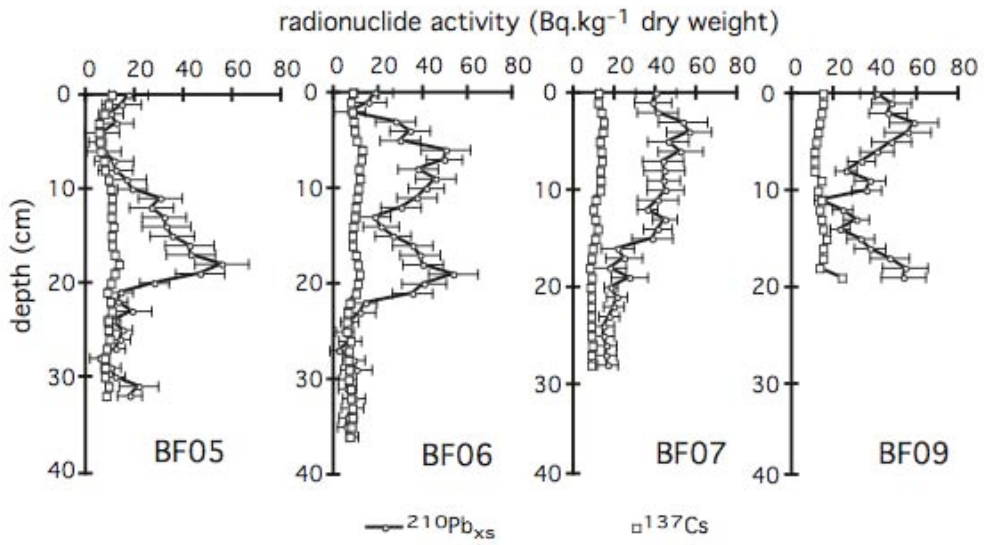


Figure 6: *Miralles et al.*

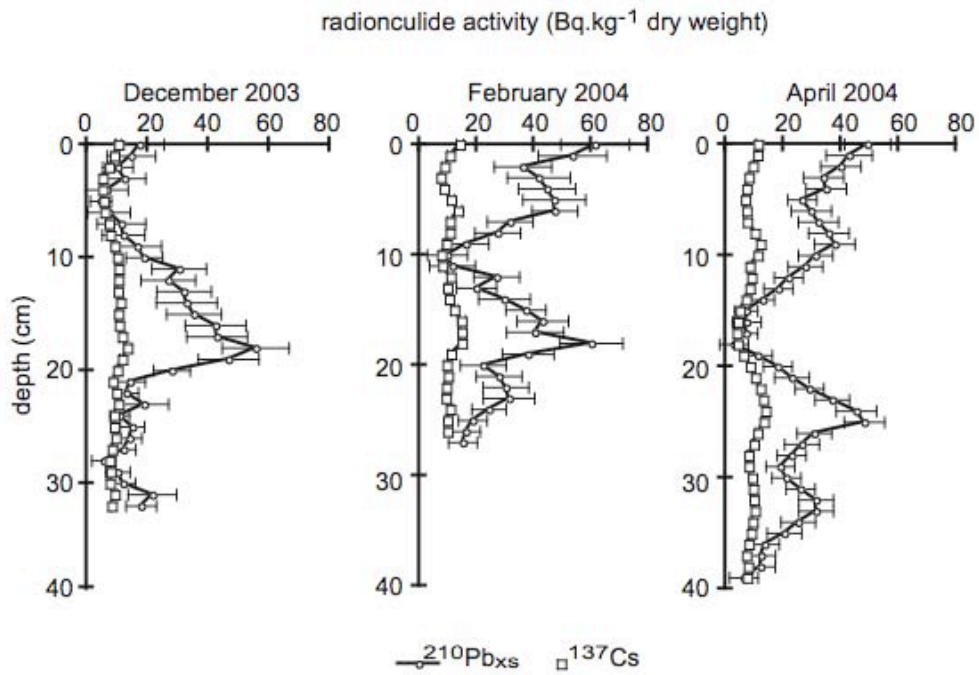


Figure 7: *Miralles et al.*

Table 1: Sampling site locations at the Rhône River Mouth (WGS 84 reference plot).

	longitude	latitude	Depth (m)	Core length (cm)
BF 05	04°51'197	43°19'696	28	33
BF 06	04°52'015	43°19'007	27	37
BF 07	04°52'118	43°18'636	45	29
BF 08	04°51'116	43°18'704	26	36
BF 09	04°51'732	43°18'855	29	39
BF 13	04°49'940	43°16'941	79	39

Table 2:  $^7\text{Be}$  ( $\text{Bq}\cdot\text{kg}^{-1}$  dry weight) limit depth of detection in sediments.

	$^7\text{Be}$ limit of detection depth (cm)	Water depth of collection (m)
BF 05	27	28
BF 06	11	27
BF 07	5	45
BF 08	Not detected	26
BF 09	9	29
BF 13	1	79

Table 3: Medium diameter (d50 expressed in  $\mu\text{m}$ ) baselines of the prodelta sediment cores.

d50 baseline ( $\mu\text{m}$ )	
BF05	$14 \pm 4$
BF06	$7 \pm 3$
BF07	$9 \pm 1$
BF09	$6 \pm 2$
BF13	$9 \pm 2$

Table 4:  $^{137}\text{Cs}$  and  $^{210}\text{Pb}_{\text{xs}}$  inventories deposited by December 2003 flood.

	sediment layer (cm)	$^{210}\text{Pb}_{\text{xs}}$ inventory ( $\text{Bq}\cdot\text{m}^{-2}$ )	$^{137}\text{Cs}$ inventory ( $\text{Bq}\cdot\text{m}^{-2}$ )
BF 05	18 to 5	Low: $3925 \pm 344$	Low: $1372 \pm 35$
	18 to surface	High: $4625 \pm 393$	High: $1890 \pm 40$
BF 06	6 to 2	Low: $1370 \pm 192$	Low: $462 \pm 19$
	6 to surface	High: $1764 \pm 232$	High: $647 \pm 19$
BF 07	4 to surface	$1712 \pm 176$	$494 \pm 19$
BF 09	3 to surface	1668	501

Table 5: Flood deposition in cores collected at site BF05 in December 2003, February 2004 and April 2004.

	December 2003	February 2004	April 2004
<sup>210</sup> Pb <sub>xs</sub> inventory	Low: 3925 ± 344	Low: 2721 ± 264	Low: 1845 ± 158
(Bq.m <sup>-2</sup> )	High: 4625 ± 393	High: 4024 ± 318	High: 3949 ± 233
Average ± sd	4275 ± 495	3373 ± 921	2897 ± 1488
<sup>137</sup> Cs inventory	Low: 1372 ± 35	Low: 1041 ± 31	Low: 738 ± 17
(Bq.m <sup>-2</sup> )	High: 1890 ± 40	High: 1523 ± 38	High: 1482 ± 24
Average ± sd	1631 ± 366	1282 ± 341	1110 ± 526

Table 6: <sup>137</sup>Cs and <sup>210</sup>Pb<sub>xs</sub> deposition at site BF05 in response to the January 2004 water discharge rise.

	February 2004	April 2004
<sup>210</sup> Pb <sub>xs</sub> inventory (Bq.m <sup>-2</sup> )	Low: 1871 ± 193	Low: 1283 ± 113
	High: 2880 ± 245	High: 3030 ± 186
Average ± sd	2375 ± 713	2156 ± 1235
<sup>137</sup> Cs inventory (Bq.m <sup>-2</sup> )	Low: 429 ± 19	Low: 335 ± 10
	High: 646 ± 24	High: 756 ± 16
Average ± sd	537 ± 153	545 ± 298