The contribution of the fine sediment fraction to the Fluffy Layer Suspended Matter (FLSM)*

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KEYWORDS

Lead 210 Polonium 210 Isotopic disequilibria Pomeranian Bay Baltic Sea

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Abstract

Fluffy Layer Suspended Matter (FLSM) is a layer of fairly concentrated suspended matter resting on the sea floor. Its passage to the depositional basins in the Pomeranian Bay – Arkona Deep System of the Baltic Sea is estimated to take around six months. In the course of this migration, the properties of FLSM change as a result of ageing and the influx of fresh particles from the water column, and possibly also because of mass exchange with the uppermost sediment layers. Measurements of radioisotopes (210 Po, 210 Pb, 137 Cs) have demonstrated that in shallow water this topmost layer of sediments, from 8 cm to 3 cm in thickness, is subject to mixing. This creates redox profiles favourable to biota and bioturbation.

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Basing on $^{210}\text{Pb}/^{210}\text{Po}$ disequilibria and the ^{210}Po excess, it was estimated that under steady state conditions from 1.5 to 2.2% of fine fraction (FSF) in the mixed layer of sediments is freshly imported from FLSM. This implies replacement of FSF from the sediments and its incorporation into FLSM. On the assumption that the surface density of FLSM is 10 mg cm⁻², FSF freshly exported from sediments actually comprises up to 15% of FLSM. Therefore, the properties of FLSM are strongly influenced by the processes taking place in the sediments, although FLSM by definition is independent of sediments.

1. Introduction

Material transport across the boundary between the ecosystems of land and sea is important as regards a number of problems ranging from the exposure of biota to pollutants, to elemental biogeochemical cycles. An important aspect in this respect is the formation and transport of suspended matter in the coastal environment. The dispersal of chemical species in the water column is determined by a variety of processes such as coastal circulation, particle absorption and adsorption, vertical and horizontal scavenging. The contribution of the processes to particulate matter formation varies seasonally, since the climate modifies both hydrological factors and biological activity. Numerous studies have been carried out to establish the importance of these factors (Alldredge & Gotschalk 1990, Mayer-Reil 1994, Bale & Morris 1998, Gustafsson et al. 1998, Jago & Jones 1998, Jones et al. 1998, Ransom et al. 1998, Laima et al. 1999, Lund-Hansen et al. 1999, Mayer 1999, Leipe et al. 2000, Löffler et al. 2000, Emeis et al. 2002, Martino et al. 2002).

An important feature of the phenomenon is suspended matter formation and transport in the near-bottom water layer. As presented in Fig. 1, particulate matter there is formed from suspended matter entering the coastal environment with river run-off, produced by abrasion of the shore and shallow sea bottom, primary production, atmospheric deposition, autogenic mineral formation, and anthropogenic discharges. En route from the high-energy, shallow, coastal environment to the low-energy, deep, depositional basins the matter is subject to later resuspension, migration driven by near-bottom currents, and sedimentation (Emeis et al. 2002). Wind-induced currents (Lund-Hansen et al. 1997) and aggregation of suspended matter particles (Thomsen & Ritzrau 1996) are thought to be major factors influencing near-bottom transport. However, in periods of calm weather suspended-matter sedimentation leads to the formation of a high-density fluffy suspended matter (FLSM) layer resting on the sea bottom. Once the FLSM is at rest, both physical and biological processes contribute to the incorporation of the suspended matter into the sediments. The same factors cause fine particles to be released from the sediment



Fig. 1. Sketch diagram showing sources and migration of the fluffy layer suspended matter (FLSM) in the coastal environment (A – atmospheric deposition; Rr – river run-off; Ab – abrasion; Pp – primary production; T – resuspension, transport, aggregation, and sedimentation; M – mass exchange between FLSM and the uppermost sediment layer)

and, at least temporarily, incorporated into the suspended matter layer. Being a part of FLSM, these particles share its fate, that is, resuspension, transport, aggregation and sedimentation in the new environment.

This paper presents the results of a study of the processes of mass exchange between the sediment and the near-bottom suspended matter. It was carried out in the Odra (Oder) River mouth – Arkona Deep system in the Baltic Sea. Surface sediments, FLSM, and nepheloid layer suspended matter were collected for the measurement and comparison of properties. Isotopic disequilibrium between ²¹⁰Pb and its granddaughter ²¹⁰Po was used to indicate the extent of the rapid mass exchange process, while ¹³⁷Cs activity profiles were used to validate assumptions concerning past mixing events of the surface sediments. The study was performed in order to quantify the exchange of mass between the uppermost layers of sediments and the overlying suspended matter.

2. The study area

The Pomeranian Bay – Arkona Deep system extends from the Odra (Oder) River estuary to the Arkona Basin (Fig. 2). Three sampling stations



Fig. 2. The study area (sampling stations: OT – Odas Tonne, TW – Tromper Wiek, AB – Arkona Basin)

 Table 1. Geographical positions, water depths, and average near-bottom water quality at the sampling stations

Station	Latitude (N)	Lonitude (E)	Water depth [m]	$\begin{array}{c} \text{Oxygen} \\ [\text{ml } l^{-1}] \end{array}$	Salinity [PSU]
OT	$54^{\circ}05'$	$14^{\circ}10'$	16	5.8	7.5
TW	$54^{\circ}36'$	$13^{\circ}46'$	26	5.9	9.1
AB	$54^{\circ}56'$	$13^{\circ}50'$	47	2.6	16.4

(Fig. 2) – Odas Tonne (OT), Tromper Wiek (TW), and Arkona Basin (AB) were established there, with the water depth increasing from 16 m at OT to 47 m at AB. The geographical co-ordinates and characteristics of the sampling stations are given in Table 1.

The hydrography of this area is influenced by the Odra with its annual water run-off of about 17 km³, and by the weather conditions, which influence the water circulation. Sediments in the Pomeranian Bay are temporarily covered by a thin fluffy layer of suspended matter (FLSM). On a weight basis the quantity of FLSM is estimated to range from 50 g m⁻² to 150 g m⁻² (Leipe et al. 2000). The layer is most often from 1 cm to 3 cm thick. The sediments are composed of a well sorted fine sand (0.1 to 0.25 mm) grading to muddy sand below 20 m water depth and to mud in the depositional basin of the Arkona Deep.

The wind is the main factor governing the currents in the Pomeranian Bay (Löffler et al. 2000). Density-driven currents and tides are of minor importance. Easterly winds dominate between February and May. At this time surface water moves to the north while the near-bottom water layer is transported to the east. Westerly winds are dominant between June and September, while westerly to south-westerly winds generally occur between October and January. During these periods the surface water is transported eastwards along the coast. This surface current is compensated by a near-bottom current, modified by the bottom topography, that flows towards the Arkona Deep (Siegel et al. 1999). The sampling stations are situated along a transect following the main transport route of the near-bottom water layer and the suspended matter it carries (Leipe et al. 2000). Although the grain size at the sediment surface was different at the sampling stations (Lund-Hansen et al. 1999), the critical shear stress at all stations was surprisingly uniform, corresponding to flow velocities of about 4 cm s^{-1} . This low value is due to the presence of an organic-rich layer resting on top of the actual sediment surface - FLSM. A flow velocity of 4.5 cm s⁻¹ is needed to transport the fluffy material (Leipe et al. 2000). The Arkona Deep acts as a final deposition area for the material, with an average mass accumulation rate from 1.0 to 1.8 kg(dry mass) m^{-2} year⁻¹ (Widrowski & Pempkowiak 1986, Emeis et al. 2000).

3. Sources and composition of FLSM

Altogether, the Odra discharges about 425 000 t year⁻¹ of suspended solids. The material is rich in organic matter (15.3% particulate organic carbon) and heavy metals (Pb – 200 μ g g⁻¹, Zn – 1700 μ g g⁻¹, Cu – 120 μ g g⁻¹, Cd – 9 μ g g⁻¹, Hg – 2.5 μ g g⁻¹) (Emeis et al. 2002). In comparison to pelagic suspended matter (PSM), FLSM is rich in hydrated three-layer and mixed-layer silicates such as illite and smectite (Leipe et al. 2000). This is due to their ability to enhance aggregation and sedimentation of suspended matter particles (Leipe et al. 2000). The organic matter content in FLSM is much smaller than in the nepheloid layer suspended matter (NLSM), indicating that organic matter may be undergoing mineralisation during sedimentation. This conclusion is supported by the fact that the organic matter content at the deepest station (AB – 47 m) is less than at the shallowest one (OT – 16 m). Fe-oxihydroxides are present in FLSM: the content of these oxides decreases from about 10% at OT to 5% at TW and 3% at AB. Mn-oxihydroxides are present only at OT (2%) (Leipe et al. 2000). This is most likely due to the anoxic conditions in the uppermost sediments, causing reduction of Fe(III) to Fe(II) and of Mn(IV) to Mn(II). The reduced species migrate to the overlying water, where they are oxidised and precipitated (Pempkowiak et al. 2000).

4. Experimental

A rosette sampler connected to a CTD-profiler was used for collecting water samples 5 m above the sea bottom (samples designated as PSM). A bottom-water sampler was used to collect samples (NLSM) 0.4 m above the bottom (Thomsen et al. 1994). The fluffy layer covering the sediment surface was collected by scuba divers, or by a remotely operated vehicle (ROV). The material (FLSM) was siphoned from the sediment surface to an on-board pump and transferred into 100 l containers.

The particulate matter was separated from the water samples by filtration through pre-weighed glass-fibre filters and analysed according to the following procedures. Particulate organic carbon (POC) and particulate organic nitrogen (PON) were measured in a Fission Instruments CHN Analyser. The activity of ²¹⁰Po was determined according to the method described in Pempkowiak (1991). In brief, the method consists of the following steps: 0.250 g d.m. of each sample is wet-digested (HNO₃:HClO₄:HF = 1:1:3), excess acids are evaporated, the dry residue is redissolved in 0.1 mol dm^{-3} HNO₃, and ²¹⁰Po is spontaneously deposited on silver discs. The activity of the deposited ²¹⁰Po is measured in a 1024 Multichannel Analyser (Polon) coupled with a Si surface barrier detector (Canberra). In the present study, the yield measured by ²⁰⁹Po (Amersham) addition prior to digestion ranged from 84.3% to 106.1% ($92.4 \pm 4.2\%$). Repeated measurements of ²¹⁰Po in the samples after 12 months of in-growth following the initial extraction of ²¹⁰Po were used for the ²¹⁰Pb activity measurements. Excessive activity of ²¹⁰Po was obtained subtracting 'repeated' ²¹⁰Po activity from the 'initial' ²¹⁰Po activity.

Sediment accumulation rates at station AB were determined using the constant rate of supply model (Robbins 1978, Pempkowiak 1991).

 137 Cs was determined by γ -spectrometry with a HPGe detector, with an energy resolution of 1.8 keV for 60 Co (1332 keV), and a relative efficiency of 30%.

5. Results and discussion

The results for samples collected at station OT are presented in Table 2, for TW in Table 3 and for AB in Table 4.

SAMPLE*	$\mathrm{C}_{\mathrm{org}}$	δ $^{13}\mathrm{C}$	δ $^{15}\mathrm{N}$	^{137}Cs	²¹⁰ Pb	$^{210}\mathrm{Po}_{\mathrm{ex}}$	Concentration
	[%]	[‰]	[‰]	$[\mathrm{mBq}~\mathrm{g}^{-1}]$	$[\mathrm{mBq}\;\mathrm{g}^{-1}]$	$[\rm mBq~g^{-1}]$	$[\mathrm{mg}~\mathrm{dm}^{-3}]$
PSM	22.6		9.6	nd^{**}	nd	nd	2.1
NLSM	14.1		9.1	nd	62.8	32.8	3.5
FLSM	8.7	-25.0	9.9	89.7	61.5	26.5	10×10^3
s $0-2\mathrm{cm}$	7.2		9.8	56.2	38.7	1.4	
s $2-4\mathrm{cm}$	7.3		nd	57.4	39.4	2.3	
s $4-6\mathrm{cm}$	6.9		nd	53.5	36.1	bdl^{***}	
s $6-8\mathrm{cm}$	7.0		nd	54.6	38.3	bdl	
s $8-10\mathrm{cm}$	6.1		nd	2.7	30.4	bdl	
s 10–12 cm	6.4		nd	bdl	31.5	bdl	

Table 2. Characteristics of the samples collected at station OT

* PSM – pelagic (5 m above bottom) suspended matter,

NLSM – nepheloid layer (40 cm above bottom) suspended matter,

FLSM – fluffy layer (0–3 cm above bottom) suspended matter,

S – fine fraction (< 0.67 mm) retrieved from 2 cm thick slices of a sediment core,

 ** not determined,

 *** below detection limit.

Table 3. Characteristics of the samples collected at station TW

SAMPLE*	$\mathbf{C}_{\mathrm{org}}$	$\delta \ ^{13}\mathrm{C}$	δ $^{15}\mathrm{N}$	^{137}Cs	$^{210}\mathrm{Pb}$	$^{210}\mathrm{Po}_\mathrm{ex}$	Concentration
	[%]	[‰]	[‰]	$[\mathrm{mBq}~\mathrm{g}^{-1}]$	$[\mathrm{mBq}\;\mathrm{g}^{-1}]$	$[\rm mBq~g^{-1}]$	$[\mathrm{mg}~\mathrm{dm}^{-3}]$
PSM	28.6		6.1	nd^{**}	nd	nd	1.9
NLSM	7.2		6.4	nd	nd	nd	5.9
FLSM	6.1	-24.0	7.0	94.6	114.7	28.6	10×10^3
s $0-2\mathrm{cm}$	6.6		6.8	85.4	104.2	1.2	
s $2-4\mathrm{cm}$	6.2		nd	76.2	104.1	1.3	
s $4-6\mathrm{cm}$	5.6		nd	75.8	102.9	0.9	
s $6-8\mathrm{cm}$	5.7		nd	1.7	32.3	bdl***	
s $8-10\mathrm{cm}$	5.4		nd	bdl	33.5	bdl	
s $1012\mathrm{cm}$	5.5		nd	bdl	31.6	bdl	

Symbols and explanations as in Table 2.

SAMPLE*	$\mathrm{C}_{\mathrm{org}}$	$\delta \ ^{13}\mathrm{C}$	δ $^{15}\mathrm{N}$	^{137}Cs	$^{210}\mathrm{Pb}$	$^{210}\mathrm{Po}_{\mathrm{ex}}$	Concentration
	[%]	[‰]	[‰]	$[\mathrm{mBq}~\mathrm{g}^{-1}]$	$[\mathrm{mBq}\;\mathrm{g}^{-1}]$	$[\mathrm{mBq}\;\mathrm{g}^{-1}]$	$[\mathrm{mg}~\mathrm{dm}^{-3}]$
PSM	7.6		4.8	nd^{**}	nd	nd	3.4
NLSM	5.1		5.7	nd	nd	nd	11.2
FLSM	6.4	-22.1	4.8	107.5	137.4	6.2	2.5×10^4
s $0-2\mathrm{cm}$	6.4		5.0	100.1	211.5	bdl***	
s $2-4\mathrm{cm}$	6.0		nd	94.3	130.9	bdl	
s $4-6\mathrm{cm}$	5.8		nd	64.2	97.0	bdl	
s $6-8\mathrm{cm}$	5.4		nd	1.8	69.4	bdl	
s $8-10\mathrm{cm}$	5.3		nd	1.3	61.3	bdl	
s 10–12 cm	5.6		nd	bdl	55.2	bdl	
s 12–14 cm	nd		nd	bdl	52.1		
s 16–18 cm	nd		nd	bdl	51.3		
s 18–20 cm	nd		nd	nd	51.9		

Table 4. Characteristics of the samples collected at station AB

Symbols and explanations as in Table 2.

It follows from the data that particulate matter is progressively more abundant in the water column towards the sea floor (Fig. 3a). There is also an increase in particulate matter in the nepheloid layer in the transect from OT to AB. The latter phenomenon is caused by frequent resuspension events in the high-energy shallow environment of OT as compared to the environment of both the deeper stations, favouring a more uniform distribution of particulate matter at the shallow stations. The same applies to the steeper concentration gradient of particulate matter at AB as compared to OT. The slightly larger concentration of particulate matter in the surface water (PSM) at OT is caused by the spring algal bloom there. The high proportion of organic matter in PSM at OT corroborates this conclusion. There is a decrease in δ^{13} C towards station AB, indicating the increasing contribution of autochthonous organic matter to FLSM. This is caused by both mineralisation of allochthonous organic matter and its dilution in the marine environment as a result of primary production. Both vertical and horizontal gradients of δ^{15} N substantiate this conclusion (Fig. 3b).

Vertical profiles of the measured radionuclides are presented in Figs. 4a, 4b, and 4c respectively for stations OT, TW, and AB.

The 210 Pb activity profile at OT exhibits a rapid activity shift 8 cm below the sediment-water interface. Below this depth only supported 210 Pb

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Fig. 3. Concentrations of particulate matter (a) and $\delta^{15}N$ (b) in the water column (PSM – 5 m above sea floor; NLSM – 0.4 m above sea floor; FLSM – 0–3 cm above sea floor, and the uppermost (0–2 cm) sediment layer at the sampling stations OT, TW, AB)

occurs in the sediment column. The sharp increase in 210 Pb activity in the 6–8 cm sediment layer is due to the presence of supported 210 Pb. The constant activity profile in sediments above 8 cm indicates that the activity shift was caused by a rapid mixing event. Both bioturbation and mechanical mixing due to wave action may explain this shift since the area abounds in macrofauna (Warzocha 1994), whereas storms causing resuspension of the bottom sediments occur on average once in 12 years (Emeis et al. 2002). The 137 Cs profile provides confirmation of such an explanation: it shows constant activities down to the 6–8 cm layer. A slight increase in 210 Pb activity in the 0–4 cm layer of sediments may have been caused by recent rapid mixing.

The mixing process at the FLSM/sediment interface could be followed using 234 Th (Tanaka & Tsunogai 1983, Tanaka et al. 1983, Gustafsson et al.



Fig. 4. Activity profiles of 137 Cs, 210 Pb and 210 Po_{ex} in the NLSM, FLSM, and sediments at (a) station OT, (b) station TW, and (c) station AB

1998). In this study 210 Po/ 210 Pb isotopic disequilibrium was used because of the simplicity and precision of the measurements. In the southern Baltic this disequilibrium was first reported by Skwarzec (1995).

In this study, the presence of ²¹⁰Po_{excess} was found in NLSM. It is an example of the polonium and lead isotopic disequilibrium frequently found in the marine environment (Poet et al. 1972, Bacon et al. 1976, Nozaki & Tsunogai 1976, Schell 1977, Heyraud & Cherry 1983, Skwarzec 1995). The disequilibrium is apparently caused by preferential accumulation of polonium by biota (Tanaka & Tsunogai 1983, Tanaka et al. 1983, McDonald et al. 1986, Skwarzec & Bojanowski 1988, Skwarzec & Falkowski 1988, Skwarzec 1995). The excess of ²¹⁰Po in the uppermost sediment laver (Tables 2–4) is caused by the rapid entry of fine particles from the FLSM to the uppermost sediments. The mechanism leading to the ²¹⁰Po and ²¹⁰Pb activity profiles may be as follows: scavenging of ²¹⁰Po from the water column (Schell 1977, Heyraud & Cherry 1983) and accumulation by macrobenthos (McDonald et al. 1986, Skwarzec 1995) leads to isotopic disequilibrium with ²¹⁰Pb in FLSM (Skwarzec 1995). FLSM is utilised by benthic organisms, thus part of it is incorporated into the sediments. The process must be a rapid one, since the ${}^{210}Pb/{}^{210}Po$ ratios in FLSM are similar to those in the uppermost sediment layer.

An isotopic disequilibrium between ²¹⁰Po and ²¹⁰Pb in FLSM was also recorded in the FLSM at station TW. It was caused by the same phenomenon as at OT. The ²¹⁰Pb activity in the uppermost, 6 cm thick, layer of the sediment column is greater than that at OT because of the higher proportion of the fine fraction in the sediments there (Leipe et al. 2000). The unsupported ²¹⁰Pb reaches as deep into the sediments as the excess ²¹⁰Po. This indicates that the uppermost layer has been subjected to rapid recent mixing, most likely as a result of bioturbation. There is no difference between the depths where ¹³⁷Cs, the unsupported ²¹⁰Pb profile, and the excess 210Po profile occur, an indication that no processes other than bioturbation leading to surface mixing have taken place. This inference is further corroborated by the slight increase of C_{org} in the uppermost 6 cm thick layer of sediments. Below 6 cm down to the sediments the activity of ²¹⁰Pb is steady. It is regarded as supported ²¹⁰Pb profile, since no ¹³⁷Cs is present there.

The sediment core at site AB represents the muddy sediments typical of the Arkona Deep. Reducing conditions ($E_h = -150 \text{ mV}$) exist in the Arkona Deep sediments. The exponential decrease in ²¹⁰Pb activity to 51 mBq g⁻¹ at a depth of 20 cm below the sediment-water interface indicates steady sedimentation and no surface mixing. The lack of isotopic ²¹⁰Pb/²¹⁰Po disequilibrium indicates that no rapid recent mixing has occurred. Bioturbation has not taken place because of the reducing conditions in the sediment column, while the considerable depth prevents mixing due to wave action. The sedimentation rate calculated from the ²¹⁰Pb profile is 0.91 ± 0.02 mm year⁻¹. The rate is similar to the earlier findings in the area (Pempkowiak 1991). The depth of actual ¹³⁷Cs activity (4–6 cm) is the same as the one calculated using the sedimentation rate derived from the radiolead profile (5 cm). This validates the calculated sedimentation rate. The ¹³⁷Cs maximum, often found in undisturbed sediments (McCall & Sörensen 1984, Wong & May 1984) is absent owing to the delayed river run-off maximum (Müller et al. 1980, Widrowski & Pempkowiak 1986).

6. Mass balance

The isotopic disequilibrium found in FLSM and in the uppermost layer of sediments combined with the concentration of particulate matter in FLSM and the proportion of fine sediment fraction (FSF) are used to calculate mass exchange between sediments and water. It is based on the assumption that the exchange is instantaneous once the FLSM is formed on the sandy bottom of the study area. The contribution of FLSM to FSF is calculated from the following equation:

$$C = \frac{{}^{210}\text{Po}_{\text{excess}}(\text{FLSM}) \times L_{\text{(FLSM)}}}{\Sigma {}^{210}\text{Po}_{\text{excess}}(\text{FSF}) \times D \times F \times d} \ 100 \ [\%], \tag{1}$$

where ²¹⁰Po_{excess} is the excess activity of ²¹⁰Po in particulate matter (FLSM), or sediments (FSF); $L_{(FLSM)}$ is the surface load of FLSM; D is depth of ²¹⁰Po_{excess}; F is the proportion of the fine sediment fraction; C is the contribution of FLSM to FSF; d is the density of sediments.

The contribution of FSF to FLSM (S) is

$$S = C \times \frac{L_{(FSF)}}{L_{(FLSM)}} = \frac{D \times F \times d}{L_{(FSF)}} \quad [\%],$$
(2)

where $L_{(FSF)}$ is the cumulative surface load of FSF.

Taking the surface FLSM load and values of D from Tables 3 and 4, and assuming F = 0.006 (OT) or F = 0.025 (TW) and d = 2.5 g cm⁻³, the respective contributions of FLSM to FSF under steady state conditions are 4.5% and 1.7% at stations OT and TW, while the FSF contributions to FLSM are 12.5% at OT and 3.6% at TW.

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