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Natural radionuclides as tracers of the dispersal of dredge spoil dumped at sea

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Abstract

Monitoring large (underwater) surfaces, with rapidly varying composition, requires a sampling density which exceeds the capabilities of standard techniques. These techniques involve sample collection and a number of treatments and measurements in the laboratory; both steps are laborious, tedious and costly. This paper presents an *in situ* method in which a detector system is trailed over the surface and measures continuously the gamma rays emitted by the natural radionuclides in the sediment. Since each sediment component has its own characteristic set of activity–concentration values (radiometric fingerprint), the composition of the sediment can be deduced quantitatively. This paper shows the application of this technique for monitoring the dispersal of dredge spoil from Rotterdam harbour, dumped in the North Sea. In addition to a qualitative picture of dredge spoil dispersal, a mass–balance equation has been used to quantitatively assess the dredge spoil transport with time.

Keywords: Natural radioactivity; Gamma-radiation; Synoptical measurements; Sea-floor sediment characterisation; Sediment transport; Dredge-spoil monitoring.

1. Introduction

Like many major ports of the world, Rotterdam harbour is almost continuously dredged to keep it accessible. The dredge spoil is, taken to sea and is, for economic reasons, dumped at a location closest to the harbour with a presumed minimal return

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flow of the dumped material to the harbour. Based on such considerations, a location, named LOSWAL-NOORD with a water depth of between 10 and 15 m about 15 km from the harbour entrance was used until July 1996. However, recent hydrodynamic modelling indicated that this location was not optimal since a considerable portion of the dumped material was calculated to return to the harbour mouth. Until then, no synoptic measuring techniques were available to monitor quantitatively the dispersal of the dumped dredge spoil. Based on similar calculations, a new dumping site, LOSWAL-NOORDWEST, was designated about 15 km NW of LOSWAL-NOORD (Fig. 1).

For LOSWAL-NOORDWEST, the hydrodynamic modelling predicted that 30% of the dredged material would be transported in a north-easterly direction, away from the harbour along the Dutch coast, and the remaining 70% would stay at the dumping site. If this prediction is correct, the higher transport costs are compensated

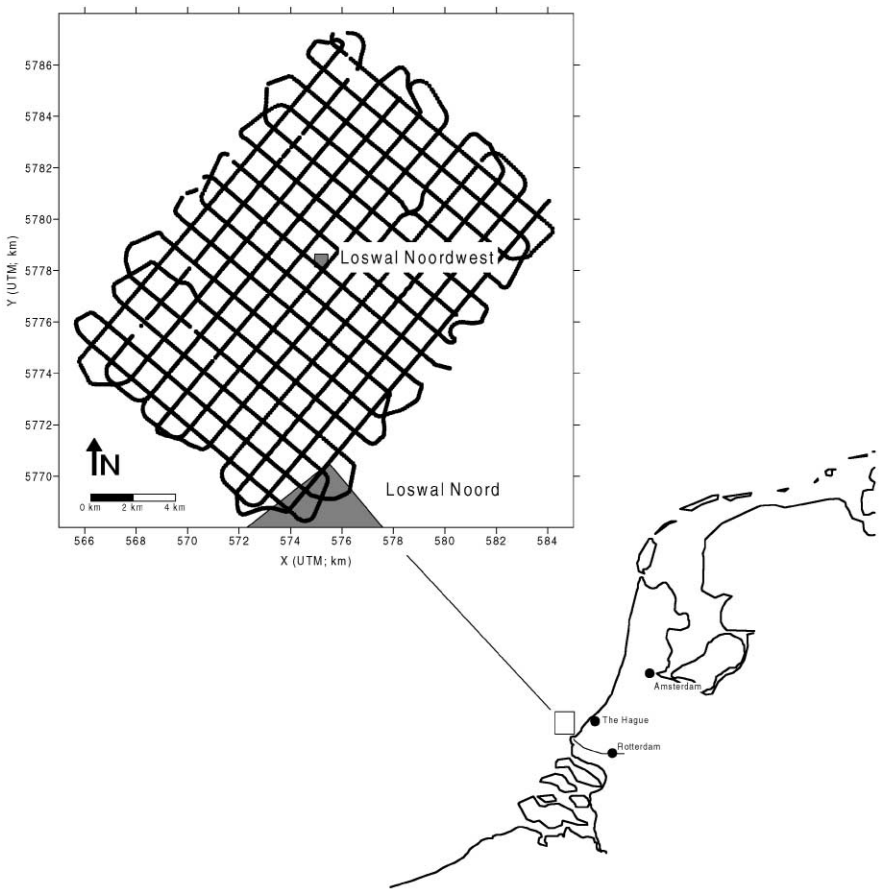

Fig. 1. Map of the Dutch Coast showing the surveyed area as an inset with the survey lines near the present dumpsite (Loswal-Noordwest) and the NW corner of the former dumpsite (Loswal-Noord).

by the reduction in dredging costs. As the harbour of Rotterdam is a very dynamic area, where changes in the coastal morphology due to man-made activities are continuous, monitoring the dredging effort is not a reliable method to establish whether the change in dumping strategy leads to the expected decrease in return flow. Consequently, an alternative method was applied for monitoring dredge-spoil dispersal.

This method makes use of the multi-element detector system for underwater sediment activity (MEDUSA) detection system. The design of this system is strongly influenced by the experience of the British Geological Survey with their Eel-system (Miller, Roberts, Symons, Merrill, & Wormald, 1977; Jones, 1994). Improvements in the design and in the analysis of the data resulted in a sensitivity of higher order of magnitude (de Meijer, 1998; Hendriks, Limburg, & de Meijer, 2001). For natural sandy sediments, a measuring time of the order of 10 s is sufficient to measure typical concentrations of 300 Bq/kg ^{40}K, 5–10 Bq/kg ^{232}Th and 5–10 Bq/kg ^{238}U with a 25% uncertainty per sampling point. In addition to the measuring device, a method has been developed to convert activity concentrations to sediment composition by means of a fingerprint technique (de Meijer et al., 1997; de Meijer, 1998). With this technique, radionuclide distribution maps can be converted to sediment distribution maps.

Based on a first demonstration of the technique (1994), The Ministry of Public Transport and Water Management, National Institute for Coastal and Marine Management (RWS/RIKZ) commissioned us to conduct a feasibility study to investigate the possibilities of quantitatively monitoring the dispersal of dredge spoil dumped at LOSWAL-NOORDWEST.

2. Objectives

The main objective of the project was to investigate the feasibility of synoptically mapping the dispersal of dredge spoil released into the sea by radiometric means. This objective was achieved by:

1. Mapping in detail a $10 \times 15\,\mathrm{km}^2$ area using a 1 km grid of survey lines and determining the activity concentrations of γ-ray-emitting radionuclides along these lines.
2. Collecting samples after the surveys from areas of interest, followed by geochemical and radiometric analysis of these samples and the derivation of radiometric fingerprints for mud (mobile part of the dredged material) and sand (original sea floor).
3. Interpolating the radionuclide data along the lines to produce radionuclide maps and converting these maps to sand/mud maps, using the established radiometric fingerprints.
4. Establishing a mass balance to assess the efficacy of the dumping practices.

3. Methods

3.1. Survey

The distributions of the natural radionuclides ^{40}K, ^{232}Th and ^{238}U in the sea-floor sediment were determined for an area of $10 \times 15\,km^2$ around LOSWAL-NOORD-WEST, roughly 10 km west of the Hague (Fig. 1). The distribution was measured using the detector system MEDUSA (de Meijer, 1998), equipped with a 5 cm ϕ (diameter) and 15 cm long cylindrical BGO γ-ray scintillator; details found in de Meijer (1998) and Hendriks et al. (2001).

The MEDUSA system is towed behind a vessel over the sea floor. A water-pressure gauge and a microphone are continuously monitored, along with the γ-ray signal, to ensure that the MEDUSA system remains on the sea floor (to avoid uncontrollable absorption of γ-rays by a water layer of variable thickness). These sensors are mounted in the watertight casing of the detector system and on its electronics board, respectively. The detector system is towed using an armoured coaxial cable that also transmits electrical power to the probe and signals from the detector electronics to the logging-PC onboard the ship. This PC also logs the position of the ship (DGPS) and the amount of cable deployed. The PC logs the γ-ray spectra and the information of the MEDUSA and on-board sensors every 10 s.

The equipment was installed onboard the Rijkswaterstaat vessel MS MITRA. The ship towed the detector system along a rectangular grid with an interline distance of 1 km. The speed of the vessel was kept at about $2\,m\,s^{-1}$. Higher speed resulted in the loss of contact of the detector with the sea floor. At this speed and at water depths of 20–30 m, the cable length required is approximately 10 times the water depth.

In total, three surveys were carried out: t_0 in June 1996, just prior to the beginning of the dumping; t_1 in November 1996 and t_2 in October 1997. In all the surveys, the same basic grid of lines was towed, with the exception of the t_2 survey in which the line system was reduced to 500 m in the immediate vicinity of the dumpsite to provide a more detailed picture.

Samples were taken with a Van Veen grab in the t_0 and t_1 surveys and with a box corer in the t_2 survey. After a preliminary analysis of the data, the locations for sampling were chosen such that

- the expected activity concentrations in the samples covered the range observed during the survey, and
- the sampling covered both the dump location and the rest of the survey area.

The samples of the t_2 survey focussed on a relatively small area to supplement the data obtained in the t_0 and t_1 surveys. Fig. 2 indicates the sample locations during the t_2 survey on a background of the measured $^{232}Th + ^{238}U$ activity concentrations. In addition, samples of the dredge spoil were collected from the transport vessels.

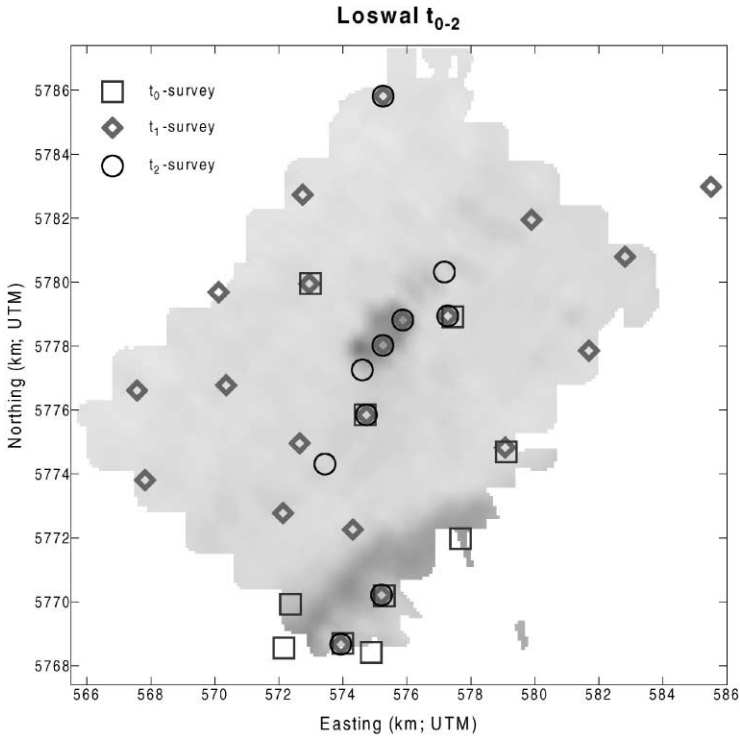

Fig. 2. Sample locations in the t_0-t_2 surveys, presented on a background of the distribution of the Th + U activity concentrations of the t_2 survey.

The total survey, including sampling, was carried out in each case for 3–4 days (working 24 h per day), depending on the weather conditions.

3.2. Laboratory work

The methodology to relate the radiometric characteristics to sediment properties has been developed partly during this investigation. One of the consequences is that the sample treatment was changed during the course of the study. At first, in the t_0 and t_1-surveys, the characteristics of the relevant sediment fractions were obtained by wet sieving (0.063, 2.0 mm), drying and analysing the sample fractions by γ-ray spectrometry. For studying the grain-size dependence in more detail and investigating the presence of another independent sediment fraction, one sample was further split to 0.016 mm grain size, employing the settling times for different grain sizes (Stokes). In the t_2-survey, sediment samples were analysed by γ-ray spectrometry and a number of standard geochemical/geophysical techniques.

3.2.1. Geochemical/geophysical techniques

These analyses have been carried out by NITG-TNO.[1] In this paper we restrict ourselves to the grain-size determination. Complete analysis also comprised the determination of the total carbon and sulphur contents and the analysis of a range of inorganic elements by XRF.

Grain-size distributions were measured with a Malvern χ-mastersizer. This technique is based on the correlation between the diffraction angle of a laser beam and the grain size of a suspended particle. This method has the advantage of producing detailed information on the grain-size distribution of the samples. The grain-size distribution determined by this technique tends to shift the diameter of the smallest grains to larger values compared to traditional grain-size determinations like the pipette method (Konert & Vandenberghe, 1997).

3.2.2. Gamma-ray spectrometry with HPGe

^{238}U and ^{232}Th are indirectly analysed using a few daughter radionuclides that emit γ-rays with sufficient intensity in the energy range of 0.1–2.6 MeV, essentially ^{214}Pb and ^{214}Bi for ^{238}U and ^{228}Ac and ^{208}Tl and ^{212}Pb for ^{232}Th. Depending on the size of the sample, samples were put either in Marinelli beakers or in small polystyrene boxes to achieve an optimal geometry and placed on top of a HPGe detector, mounted inside 10 cm lead shielding. Escape of ^{222}Rn is prevented by sealing and storing the samples for a period of three weeks prior to measurement. The efficiency of the detector, corrected for the sample matrix, is regularly checked with standard solutions and by (inter)national intercomparisons.

In this investigation, activity concentrations of ^{40}K, ^{238}U, ^{232}Th were determined. The reported values of the series are the weighted averages for the activities of each γ-ray transition under the assumption of secular equilibrium between the various decay products. Values will be presented together with uncertainties in which the goodness-of-fit is included.

3.3. MEDUSA data processing

The processing of the data obtained with MEDUSA has been carried out in two steps: data quality checks and a procedure called radiometric sediment characterisation. The first step contains

- Removal of "off-bottoms": events in which the contact between detector and sea floor was lost, were removed from the data set. These events are recognised by a sudden drop in the activity measured by the γ-ray detector and in the friction-sound level (measured with the microphone in the probe) and a sudden change in water depth, not supported by the echo sounder of the survey vessel.
- The X and Y co-ordinates of the ship (DGPS) are transformed to X and Y co-ordinates of the detector, based on the amount of cable released and the course of the ship.

[1] Netherlands Institute of Applied Geoscience TNO — *National Geological Survey*.

- Water-depth measurements have been converted to bathymetry using the time-evolution of a bottom water-height sensor, located in the vicinity of the survey area, to correct for the swell and tidal effects.

Radiometric sediment characterisation is a method to characterise sediments according to their radionuclide activity concentrations. It uses the intrinsic properties of the sediment, determined in a laboratory calibration, and measurements of the (natural) radionuclide concentrations in situ with MEDUSA. The laboratory calibration is based on a limited number of samples.

Fig. 3 illustrates the principle of radiometric sediment characterisation. The boxes at the bottom indicate the measuring instruments in the detector system and the data obtained from the ship's positioning system. These data are combined to form one record.

The radiometric data are analysed as a total count rate (TC), counting all γ-ray events with an apparent energy above a certain threshold ($\sim$10 keV), and as the activity concentrations of ^{40}K and the decay series of ^{232}Th and ^{238}U. For the latter, the recorded γ-ray spectra are analysed in a full-spectrum analysis method using standard spectra (de Meijer et al., 1997; Hendriks et al., 2000). Standard spectra reflect the response of the detector in a particular geometry for an activity concentration of 1 Bq/kg for a specific nuclide. For the present survey, standard spectra were measured in a water-filled tank, placed on concrete calibration pads at the British Geological Survey, Keyworth, UK. The goodness of fit (χ^2), of the spectra, obtained by comparing the standard spectra to the actually measured spectrum, is used as a quality check on the data processing. The results of this stage in the processing are the maps of TC, and K, Th and U activity concentrations.

To convert the activity concentration distributions to sediment distributions, the radiometric fingerprinting method was used (de Meijer et al., 1997; de Meijer, 1998). In this method, the sediment is divided into a number of groups, corresponding to geochemical or geophysical properties, e.g. grain size. For each group, the activity

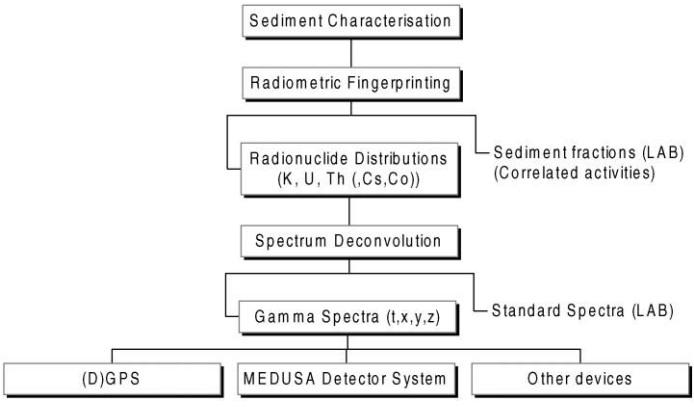

Fig. 3. Schematic presentation of the method of radiometric sediment characterisation.

concentrations of K, Th and U were determined. In the t_0 and t_1 surveys, the groups were defined as sieved fractions of the sediment samples, taken at the end of the survey. The fingerprint was obtained from the radiometric measurements of these fractions. In the t_2 survey, the full grain-size distribution was used and the total activity of the sample was written as the sum of the (unknown) activity concentration of each radionuclide in each fraction (see Appendix A). We refer to the latter procedure as "fraction analysis". The radiometric fingerprints were subsequently used to "translate" the activity–concentration distributions to sediment–composition distributions.

The line data were interpolated to a regular grid using Punctual Kriging techniques (see e.g. Burgess & Webster, 1980). The method calculates weighted averages based on nearby sampled data points. The weight function used is based on a parametrisation of the variance in the data (see Cressie, 1991). The Kriging was carried out in Surfer 6 (Golden Software Inc).

More details on the above procedures are given in a set of data and synthesis reports that are available from the authors (Venema, Koomans, & de Meijer, 1997a; Venema, Koomans, Stapel, de Meijwe, & Groen, 1997b; Venema, Koomans, Stapel, de Meijer, & Okker, 1997c; Venema, Koomans, Stapel, Zwanenburg-Nederlof, & de Meijer, 1997d; Venema, Manso, Koomans, Limburg, & de Meijer, 1998; Venema, Limburg, & de Meijer, 1999a; Venema et al., 1999b).

3.4. Mass balance

A mass–balance equation was used to assess the quantitative aspects of the dispersal. For this purpose, the area is subdivided in sectors and in each sector integrated values of the radionuclide concentrations are calculated. These area-integrated values are subsequently converted to mud concentrations using the fingerprints of sand and mud, and the fact that the relative masses sum to unity.

An exception is made for the dumping area itself. Since the thickness of the material exceeds the depth "seen" by the detector system, the amount of material remaining had to be estimated. It was assumed that the dumpsite had a homogeneous composition. The estimation is based on the difference in mud content of the dredge-spoil samples and in the top layer of the dumpsite, measured with MEDUSA. According to this procedure, 30% of the dumped material remains at the dumpsite. This assumption was supported by the results of bathymetric measurements.

4. Results and discussion

4.1. Radionuclide distributions

The interpolated maps of total activity, summed $^{232}Th + ^{238}U$ (see next section) concentrations and the ^{40}K activity concentration for the three surveys are presented in Fig. 4. The former dumpsite (LOSWAL-NOORD) is clearly seen as an area with

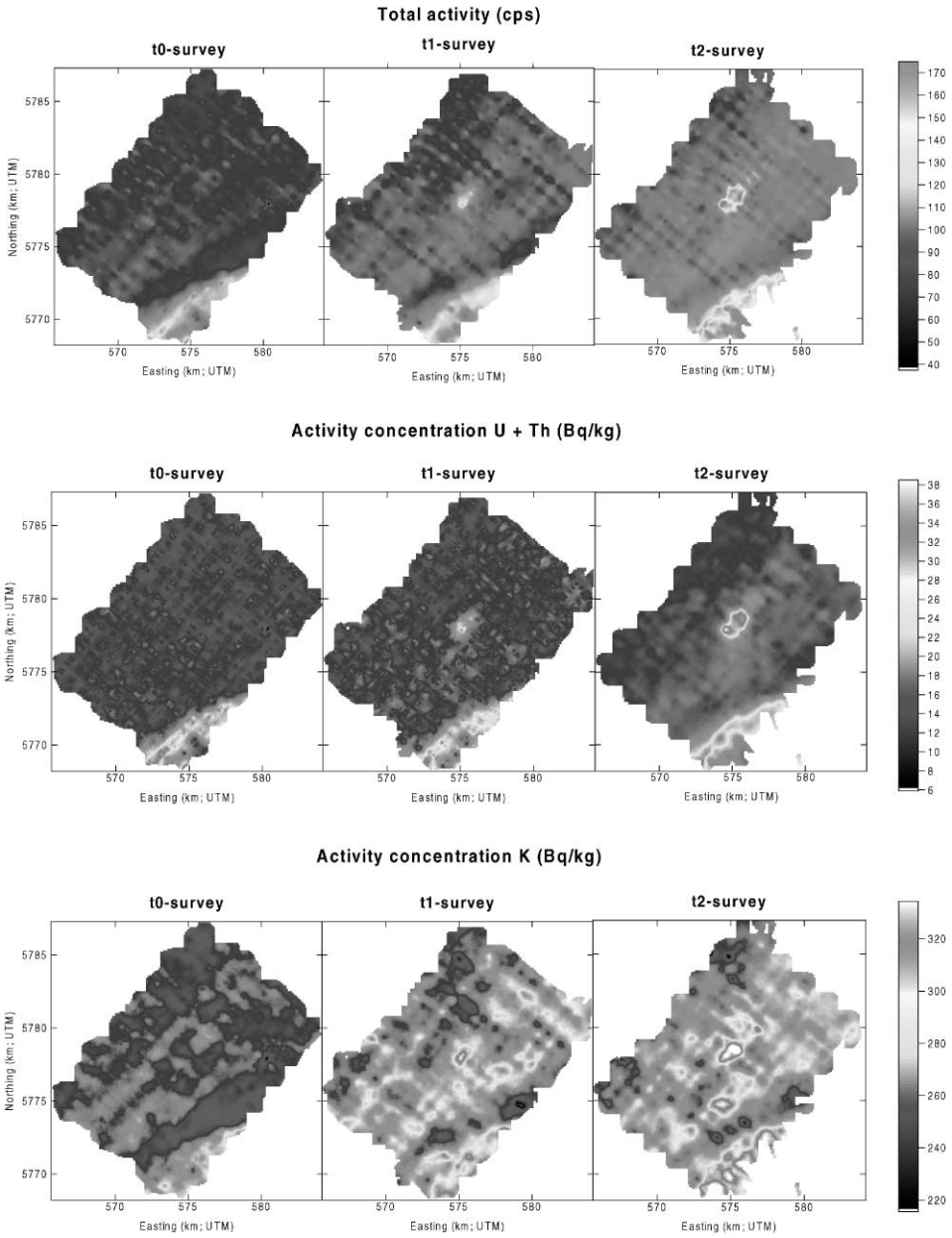

Fig. 4. Radiometric sea floor maps for the t_0, t_1 and t_2 surveys.

enhanced concentrations (lower-right corner). In the t_0 survey, there are no other areas with enhanced Th + U concentrations, whereas, the K activity concentrations still show some patchy enhancements. A low-activity area at the NW edge of LOSWAL-NOORD is especially noticeable in the K maps. This area at 20 m water depth is probably caused by selective sediment transport, removing a finer and more active sediment fraction, by currents that were enhanced by the presence of the dumpsite.

In the t_1 and t_2 surveys, the effect of the dumping is clearly visible. In the combined Th + U maps, the dumpsite stands out clearly and increases in intensity with time. There is also some evidence for dispersal in a north-easterly direction, most clearly seen in the total activity figures. It is worth noting that the activity concentrations in the U + Th map vary by a factor of 6. In the K activity distribution map, the variation is only by a factor of 1.5. Due to the smaller variation in signal, the colour scheme enhances the spread of K over the total area. Also, for K the activity concentrations increase between surveys. In addition to dispersal to the NE, some movement in a southerly direction is observed, especially in total activity.

4.2. Radiometric sediment characterisation

Table 1 lists the results of the laboratory sample analysis of seafloor sediments for the t_0 and t_1 surveys. The initial task was to distinguish between the mud ($d < 63\,\mu m$) and the sand ($63\,\mu m < d < 2\,mm$). In the t_0 and t_1 surveys, 30 samples were sieved and analysed by γ-ray spectrometry. In view of the different patterns for Th + U and K activity concentrations in the surveys, one mud sample was analysed for grain-size distribution to investigate a possible further separation of mud into "clay" and "silt". Fig. 5 presents this grain-size distribution. As can be seen from Fig. 5, at $d \sim 10\,\mu m$ there is an extra component. Therefore, this sample was split, by settling, into a component with $d < 16\,\mu m$ (clay) and another component with $16 < d < 63\,\mu m$ (silt). This type of splitting is quite time-consuming and not too accurate; for that reason only one sample was split. Each component was measured on the HPGe γ-ray detector for its radionuclide content.

The results in Table 1 indicate the relatively large difference in radiometric fingerprints between (the original) sand and the (dumped) mud on the sea floor: a factor of two in K, a factor of 5–6 in Th and U. The ratio between the activity

Table 1
Activity concentrations of dry sediment (with 1σ uncertainties) of sediment size-fractions from the LOSWAL-NOORDWEST area, determined from samples from the t_0 and t_1 surveys

Grain-size fraction	^{40}K (Bq/kg)	^{232}Th (Bq/kg)	^{238}U (Bq/kg)
Clay ($d < 16\,\mu m$)	540 (30)	32.4 (1.1)	31.2 (1.2)
Silt ($16 < d < 63\,\mu m$)	340 (20)	28.5 (1.0)	29.7 (0.9)
Mud ($d < 63\,\mu m$)	440 (30)	29.7 (1.0)	33.5 (1.1)
Sand ($63\,\mu m < d < 2\,mm$)	213 (13)	5.8 (0.2)	5.2 (0.2)

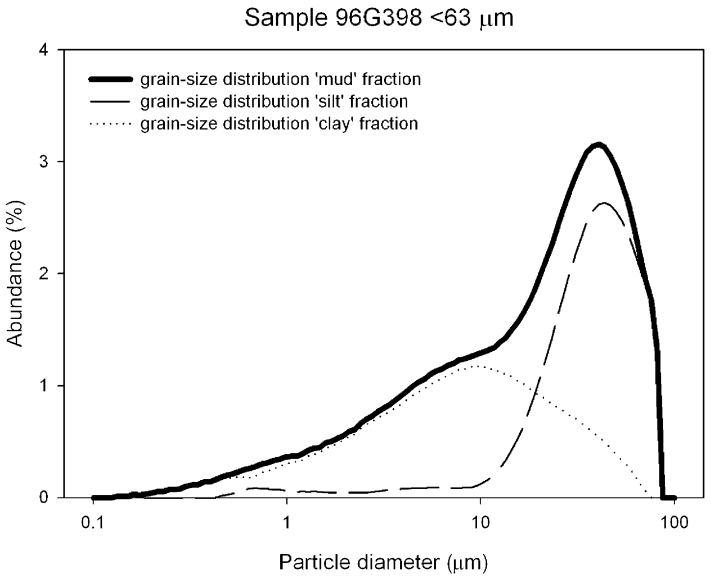

Fig. 5. Grain-size distribution of the mud fraction for a selected sediment sample.

concentrations of Th and U is approximately unity for all sediment components. Hence, a separate treatment of Th and U does not provide extra information and, as shown above, the sum of the activity concentrations has been used in the analysis. The difference between clay and silt is mainly in the K activity concentration. These fingerprints qualitatively explain that the K spread over a much larger area (see Fig. 4) is caused by the very mobile clay fraction of the sediment.

Because of the presence of carbonates and organic matter, the sediment grains stick together to form aggregates. These aggregates prevent a clear separation into the two sediment fractions. The carbonates and organic material cannot be removed before the radiometric measurement to be able to interpret the radiometric data in situ. In particular, the sand signature suffered from this admixture of aggregates. Based on the experiences of the t_0 and t_1 surveys, the strategy for obtaining the fingerprints was changed in the analysis of the t_2 survey, from splitting to the method of "fraction analysis" (see Appendix A). Moreover, prior to the grain-size measurements, the samples were chemically treated with HCl and H_2O_2 to remove carbonates and organic carbon. In principle, the grain-size distribution can be influenced by this procedure, but this bias cannot be prevented. On the basis of the 22 samples collected in the t_2 survey (12 samples from dredge vessels and 10 samples from the sea floor, for locations see Fig. 2), it was attempted to obtain a fingerprint for three fractions. This first attempt failed and could indicate that the sample form the t_1 survey used for the further splitting was not representative. The fraction analysis indicates only two significantly different components in the sediment: mud ($d < 63$ μm) and sand ($d > 63$ μm). Moreover, based on the goodness-of-fit, we find a

Table 2
Activity concentrations of dry sediment (with 1σ uncertainties) of sediment size-fractions from the LOSWAL-NOORDWEST area, determined from the 10 seafloor samples collected in the t_2 survey

Grain-size fraction	^{40}K (Bq/kg)	^{232}Th (Bq/kg)	^{238}U (Bq/kg)
Mud ($d < 63\,\mu m$)	520 (50)	34 (4)	29 (3)
Sand ($63\,\mu m < d < 2\,mm$)	254 (6)	5.9 (0.4)	6.5 (0.3)

difference in fingerprints between the samples from the dredging vessels and from the seabed. In retrospect, this could be expected because the dumpsite contains the remainder of the dumped material and the material washed out has been transported. The dredged material and the remainder on the dumpsite have different hydrodynamic properties and these differences are apparently reflected in the radiometric fingerprints.

Within the framework of the project, these subtle differences could not be further investigated and the fingerprints for the mud and sand fractions were taken as the weighted average of the values obtained for the ten seafloor samples only. The results, presented in Table 2, indicate slightly different activity concentrations than the values listed in Table 1. This change in activity is due partly to the chemical treatment prior to the grain-size distribution measurements (the mud and sand fractions no longer contain carbonates and organic matter) and partly to the different methodology in determining the radiometric fingerprints. As stated earlier, the radiometric analysis occurs on untreated samples. In addition, the fact that the radiometric fingerprints of the dredged material changed with time cannot be excluded. Samples, originating from different sections of the harbour, taken from dredge spoil in the transport vessels, showed marked differences in radiometric properties.

4.3. Mud distribution

The fingerprints of Table 2 have been used to convert the radionuclide distribution map to a mud content map for the three surveys. The result is presented in Fig. 6. From the scale one notices that the mud content of the sea-floor sediment ranges between 0 and 36%. The high mud concentrations are found at the dumpsites LOSWAL-NOORD and LOSWAL-NOORDWEST. In the remaining part of the area, one notices an increase with time and a spread of material over the total area and likely beyond its boundaries. Moreover, the patterns on LOSWAL-NOORD change with time, indicating that changes occurred in the bed of the former dumpsite.

4.4. Mass–balance equation

To quantify the dispersal of the dredge spoil, an attempt has been made to make a mass–balance equation and use the information obtained from the surveys. We have

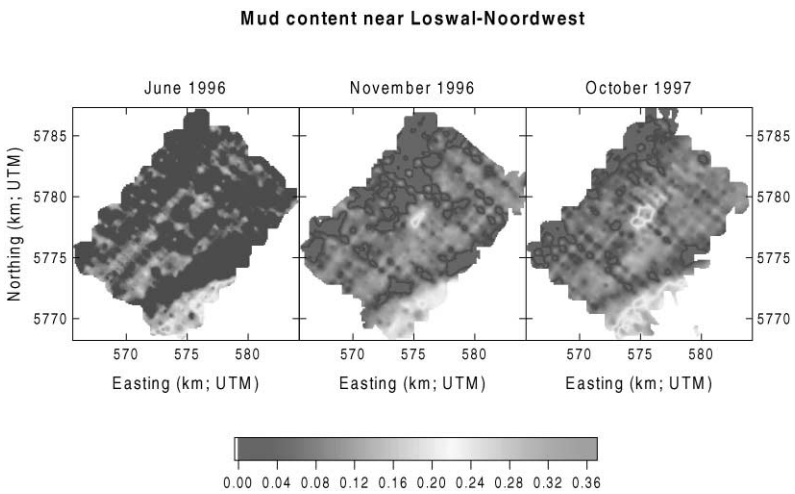

Fig. 6. Mud content of the sea floor for the t_0, t_1 and t_2 surveys.

divided the survey area into a number of regions and calculated for each region the masses of the dredge spoil from the change in the average activity concentrations of K and Th + U. The number of data points per region is high and the statistical uncertainties in the averages are negligible compared to the systematic uncertainties that enter the estimate. These systematic uncertainties encompass:

1. Accuracy of the MEDUSA measurements.
2. Accuracy of the radiometric fingerprints.
3. Uncertainty introduced in heterogeneity in the sediment bed.
4. Accuracy of the interpolation; and
5. Uncertainties about the porosity of the sediment on the dumpsite and in the upper layer of the sea floor.

It is difficult to make the systematic uncertainties quantitative, but we will discuss the various sources of these uncertainties.

1. The calibration of the MEDUSA detector system is carried out on homogeneous calibration pads with finite dimensions and with a certain density. For sandy sediment, comparisons between the activity concentrations in samples and their neighbouring MEDUSA measurements show good agreement. However, in very muddy sediments ($>70\%$ mud), we observe a systematic underestimation of the concentrations by MEDUSA, likely related to the lower density of the material and consequently absorption by extra water. This may lead to an underestimation of the mud content, but such high mud concentrations are not encountered in these surveys.

2. The radiometric fingerprints have been based on fraction analysis of a total of 10 samples. The results were consistent with the results obtained from 30 samples in the t_0 and t_1 surveys by splitting. As in all field studies, the sample selection biases the

result. In the present study, the main bias is caused by (1) the uncertainty in the extent to which the composition of the dredge spoil remains constant during the dumping period and (2) to what extent the fingerprints are equal for the mobile sediment fraction and the material residing at the dumpsite. As mentioned above, we observed marked differences in the radiometric properties of samples taken from different locations in the harbour. What and how large these systematic uncertainties are is uncertain. However, it should be realised that these uncertainties are not typical for the radiometric method; they reflect differences in sediment type. Typical values encountered in our analysis show differences of less than 10%.

3. The calibration is carried out for a homogeneous matrix. On the sea floor, it is likely that the bed is heterogeneous. In this situation, the mud layer in which no severe storms were recorded, will probably lie on top of the sandy sea floor. The intensity of the γ-radiation absorbed will therefore be less than in the case of a homogeneous bed or of sand layers covering the mud. The attenuation coefficient is about 8 cm and layers buried deeper than three to four times this value will not be observed. Depending on the situation, there can either be an overestimate or an underestimate of the actual mass of mud. Again, this is a general sampling problem; taking and analysing samples has a similar systematic bias. In extreme cases, assuming no mixing and large mud layers, the uncertainty can reach 30%.

4. The interpolation uncertainty is a general issue for observations of spatially varying quantities. Relative to standard sampling techniques, our number of data points ($\sim$20,000) is incredibly high. Moreover, our two perpendicular grid systems reduce the effect of the unknown and locally varying semi-variances. We believe that this uncertainty is not the dominant one. Nevertheless we have indications that a denser grid ($\sim$0.5 km spacing) would reduce the variance in the data and hence will lead to even better quality data. Assumed accuracy, even with this sampling density, is between 10 and 20%.

5. Porosity and effective density are strongly related. Our values for the density of the material have been estimated from the values obtained from box cores taken at the dumpsite. A value of 1.5 kg/l dry weight was adopted in the present calculations. For mass balance calculations, this is a very crucial parameter: how much material resides at the dumpsite? Values between 1.1 and 1.5 kg/l are in use.

Table 3 presents the average activity concentrations of ^{40}K and ^{232}Th + ^{238}U in the sub-regions of the surveyed area at the time of the three surveys together with the surface area and the number of data points in each region. The values in the table show the increase in time, even at the area of the former dumpsite LOSWAL-NOORD.

Using the fingerprints listed in Table 2, the values in Table 3 have been converted to changes in mud volumes. The results are presented in Table 4. In this table, the values for mud at the dumpsite have been set at 30% of the dumped material. The results in Table 4 indicate that, after five months of dumping activity (t_1), almost all sediment released is accounted for either at the dumpsite or at the surface of the surveyed area.

Fig. 7 presents the dispersal data as percentages of the total amount of material dumped up to the particular survey. The dominant transport directions (20–25%)

Table 3
Average activity concentrations of dry sediment (Bq/kg) in sub-regions of the surveyed area, for the three surveys conducted. In addition, the area (km^2) of each sub-region and the number of data-points are given. Uncertainties are not specified. The as yet unknown systematic uncertainties completely dominate the total uncertainty

		East	North	West	South	Loswal-NW	Loswal-N
K-40	t_0	261	260	268	262	274	291
	t_1	293	275	286	292	308	316
	t_2	295	286	287	294	316	321
U-238	t_0	7.2	7.1	7.5	7.7	7.2	12.8
	t_1	7.8	7.5	7.9	8.2	10.1	12.4
	t_2	9.1	8.7	8.7	9.6	14.7	15.0
Th-232	t_0	6.3	6.2	6.5	6.6	6.8	9.6
	t_1	6.9	6.3	6.6	7.2	8.4	10.0
	t_2	7.6	7.0	7.2	7.7	10.2	10.6
Area		29.98	29.095	34.135	25.165	6.505	9.77
#data-points		4300	4200	4900	3600	940	1400

Table 4
Net accumulation of mud in various locations at t_1 and t_2

Area	After t_1		After t_2	
	(m^3)	(%)	(m^3)	(%)
Totally dumped	1.2×10^6	100	4.5×10^6	100
Remaining at dumpsite	3.6×10^5	30	1.4×10^6	30
East	2.7×10^5	23	6.8×10^5	15
North	1.2×10^5	10	4.9×10^5	11
West	1.4×10^5	12	4.6×10^5	10
South	2.3×10^5	19	5.9×10^5	13
LOSWAL-NOORD	3.8×10^4	3	2.4×10^5	5

are to the east (parallel to the Dutch coast) and to the south (direction of the harbour mouth). The dispersal to the western and northern sectors is of the order of 10%. Even at the former dumpsite LOSWAL-NOORD, an increase of mud concentration is observed.

In October 1997, about 15 months after the beginning of dumping (t_2), the total volume of dumped material had increased by almost a factor of 4. As can be seen from Table 4 and Fig. 7, the amount of material remaining at the dumpsite and in the survey area no longer accounts for the amount of dumped material: about 15% of the material is missing in the mass–balance. This missing material has probably left the area. Although less marked than at t_1, the easterly and southerly directions of transport are largest and again new material is present at the former dumpsite.

If the missing material is equally divided between the easterly and southerly transport directions, one obtains a distribution pattern for t_2 which is, within the

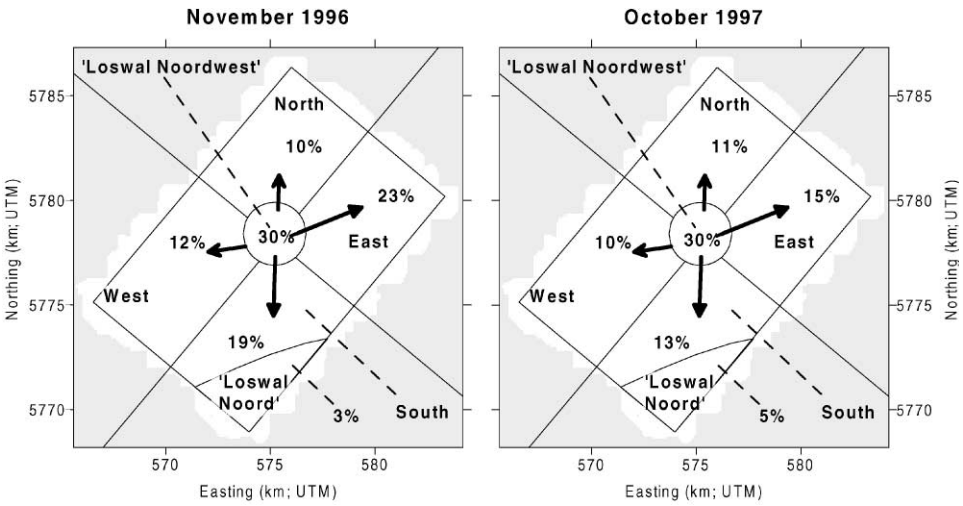

Fig. 7. The mass balance of mud changes in the surveyed area; the numbers indicate the percentage of dumped mud observed in the different areas.

uncertainties, the same as the pattern at t_1. This assumption is consistent with a distribution pattern in time that is more or less constant. In the north and west sectors the dispersed material is still present inside the sectors, whereas in the east and south directions the dispersal has carried dredge spoil beyond the boundaries of the surveyed area. The information on southerly movement means that it is likely that, in the course of 1997, the dispersed dredge spoil reached the harbour mouth. It is interesting to note that, according to the Port Authorities, an increase in dredging was required from 1998 onwards.

5. Conclusions and outlook

This project started in 1996 as a feasibility study of monitoring the dispersal of dredge spoil from Rotterdam harbour by radiometric means. It can be concluded after three surveys that the information obtained from the activity distributions of the natural radionuclides in the upper layer of the sea-floor sediment shows considerable changes since the start of dumping. These changes can be measured by the MEDUSA seabed detector system, containing a highly sensitive gamma-ray spectrometer. Towing the detector at a speed of 2 m/s allows integration times of 10 s and thereby a spatial resolution of about 20 m.

The method proved to be very sensitive to small changes in sediment composition and allows the quantitative monitoring of net transport of dredge spoil. Measurements and analyses are conducted in a very short time (about 1 week each), once the sediment characteristics have been established. The density of information (20 m along each survey line) is much higher than

could be obtained from standard sampling techniques and subsequent laboratory analysis.

This work shows that quantitative information can be obtained on the net transport flux of the components in the dredge spoil using radiometric techniques. As far as we are aware, this is the first time that such a quantitative assessment of dredge spoil release at sea has been made. This was possible because of the high sensitivity of the MEDUSA detector system and the translation by radiometric fingerprinting of the sediments. The accuracy of the assessment at present is mainly determined by (1) the quality of the classification of the sediment components and the differences in their radiometric fingerprints, (2) the density of the grid system and the uncertainties introduced by interpolation, and (3) the limiting assumptions on e.g. homogeneity and density of the seabed material. In the fingerprinting, we observed changes of less than 10% between the different surveys. Such changes have approximately a 10% influence on the mass transport. Presently, the uncertainty of the spatial distribution (both in vertical and horizontal directions) of the dredge spoil is the dominant uncertainty.

The drawback to the sensitivity of this method is that many of our surprising observations cannot be field-checked. It requires sampling with an unrealistically high densities to obtain sufficient data to observe these small changes in sediment composition.

To improve the quality of the sediment characterisation, the geochemical, geophysical and radiometric properties have to be compared in detail. The uncertainties of the interpolation can be reduced by towing in a finer grid and adjusting the grid to the observed variances. Since no information was available prior to the first survey, the present grid system was chosen and maintained during the follow up measurements. In the t_2 survey, the release site was mapped at a denser (0.5 km) grid. The effects of inhomogeneities and variations in density have recently been observed (Venema et al., 2000). We anticipate to reduce the uncertainties by obtaining information from Monte-Carlo simulations and comparisons between seabed and laboratory measurements.

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Appendix A. Fraction analysis

The fraction analysis is based on the principle that the total activity concentration of each sample (or each location) is the sum of the activity concentrations of each fraction. In mathematics

$$C_i = \sum_j D_{ij} f_j, \tag{A.1}$$

where C_i is the measured activity concentration of nuclide (or series) i, f_j the size of sediment fraction j and D_{ij} the activity concentration of nuclide (or series) i in sediment fraction j. In our case the nuclides (series) are ^{40}K, ^{238}U, and ^{232}Th.

When C_i is measured for each sample and D_{ij} is known for each fraction, f_j can be calculated from these equations. As the system might be over-determined, a least-squares procedure should be used to determine the best possible solution, minimising

$$\sum_i w_i \left(C_i - \sum_j D_{ij} f_j \right)^2. \tag{A.2}$$

The weighting factor w_i is used to incorporate the different weights for the different components and activity concentrations in this sum. The experimental uncertainty and the uncertainty in the calibration values D_{ij} determine this weighting factor in the following way:

$$w_i = \frac{1}{\sigma_{c_i}^2 + \sum_j \sigma_{D_{ij}}^2 f_j^2}, \tag{A.3}$$

where the σ^2's are the variances of the values and the f_j 's again the sizes of the fraction. Due to the dependence of the weights on the fraction sizes, the least squares procedure is not linear.

It is important to realise that there is a constraint on these fraction sizes, i.e. the total sum of all fractions equals one. This can be implemented in the procedure by minimising

$$\chi^2 = \sum_i w_i \left(C_i - \sum_j^{n_f - 1} D_{ij} f_i - D_{im} \left(1 - \sum_j^{n_f - 1} f_i \right) \right)^2, \tag{A.4}$$

where the number of adjustable fractions is reduced by 1. The expression for the weighting factors is not changed by this constraint.

The uncertainties in the calculated fraction sizes are determined by calculating the (co)-variance of the "fit" parameters (the fraction sizes), using the Hessian matrix

$$H_{ij} = \frac{\partial}{\partial f_i} \frac{\partial}{\partial f_j} \chi^2. \tag{A.5}$$

The covariance matrix (**C**) is the inverse of the Hessian matrix and the uncertainty in the fraction size is the square root of the diagonal elements. Of course, the Hessian matrix is defined only for the independent parameters. Therefore, the uncertainty of the last fraction should be determined from the covariance matrix of the other

fractions according to

$$\sigma^2_{f_{\text{last}}} = \mathbf{F}^{\text{t}}\mathbf{C}\mathbf{F}. \tag{A.6}$$

where **F** is the vector with fraction sizes except for the last fraction and **C** is the covariance matrix.

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