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CHEMICAL TRACKING AND MARINE ENVIRONMENTAL IMPACT OF OCEAN DISPOSAL OF CALCIUM CARBONATE RESIDUE IN THE NORTH YELLOW SEA

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The objective of this study is **to** determine the sedimentation rate and dispersion area of calcium carbonate residue dumped at sea and the impact to marine environment of dumping by a laboratory simulation experiment; chemical tracking in the field with the help of acoustic and optical tracking; and a comparative study of baseline conditions and marine environmental impact after dumping. Turbidity, pH and phosphate are selected as the chemical tracers to be monitored.

Results show that in the dumping area of 15 square miles with water depth of 50 m, if 217 t calcium carbonate residue is dumped (spot dumping) in the presence of a pycnocline with a current velocity of 60 cm/s (close to the maximum) the maximum dispersion distance of the calcium carbonate residue plume front is less than 2100 m; the dispersion area is less than 0.56 km²; and the maximum dispersion time is about *60* min when the turbidity and pH in the whole dispersion area return to background level. Therefore, the ocean disposal of calcium carbonate residue is feasible.

KEY WORDS: Calcium carbonate waste, ocean disposal, environmental impact

INTRODUCTION

Papers published on research on ocean dumping have greatly increased since early 1970s. Most studies were done by scientists from the conttacting parties of the London Dumping Convention. About **470** papers on ocean dumping were published in the last decade (1981-1990). Duedall and others edited a series of monographs under the title of 'Wastes in the Ocean' (Duedall, et *al.,* 1983; Kester, *et* al., 1983; Park, *et af.,* 1983; Duedall, *et af.,* 1985; Kester, et al., 1985; Ketchum, *et al.,* 1985). Much research was done on ocean dumping of dredged material (Chave, 1983; Dayal, 1983; Bohem, 1983), fly ash (Crecellus, 1985; Rose, 1985; Norton, 1985; van der Sloot, 1985; van der Sloot, 1985b; Merifield, 1985), and sewage sludge (McKnight, 1985; Oakley, 1985; Duedall, 1977). Until now, research on ocean dumping of calcium carbonate residue has rarely been reported in the literature.

The Dalian Chemical Company produces *800* thousand tons of sodium carbonate annually with 300 thousand tons of calcium carbonate residue waste. How to dispose of the residue waste is one of the key problems in keeping a continuous production of sodium carbonate. The preliminary comparative assessment of land-based and ocean disposal shows that the ocean disposal of calcium carbonate residue waste is less harmful to the environment and more economic. But further research is needed to provide more information to ensure that ocean disposal of calcium carbonate

residue is feasible before a dumping permit can be issued. This paper presents some results of a project entitled 'Study on ocean disposal of calcium carbonate residue'.

0 bjectives

There are two main objectives of this study:

- 1. to identify the possible impact of ocean dumping of calcium carbonate residue to the aquatic environment.
- 2. to determine the rate of sedimentation and the area of dispersion of calcium carbonate residue dumped at sea by the quantitative study of temporal and spatial distribution of chemical tracers, i.e. turbidity, pH and phosphate.

Experimental design

The following three types of experiment were designed to answer the questions above:

- 1. laboratory simulation using the calcium carbonate residue from the Dalian Chemical Company and sea water taken from the dumping site in order to simulate the possible impact of dumping and to select the best chemical tracer to be monitored;
- 2. field chemical tracking to determine the vertical and geographical distribution of the chemical tracers using chemical, acoustic and optical techniques;
- 3. comparative field study of baseline conditions before dumping and of environmental impact after dumping to verify the results from the laboratory simulation and to understand the real impact of dumping of calcium carbonate residue.

LABORATORY SIMULATION

Chemical composition of the residue waste

The main components of the residue waste are carbonates, hydroxides and sulphates of calcium, magnesium and sodium. The weight percentage of calcium carbonate in dry residue is 70-95%. The chemical composition of the residue waste is very similar to that of marine carbonate sediment. Therefore, it is considered to be nontoxic. The diameter of all particles of $CaCO₃$ residue is less than 80 μ m and of total particles 96% is less than $50 \,\mu\text{m}$, $70\% < 20 \,\mu\text{m}$, $30\% < 10 \,\mu\text{m}$, $20\% < 8 \,\mu\text{m}$ and $7.5\% < 5 \,\mu\text{m}$.

Experimental methods

Selection of *impact parameters*

According to the chemical composition and characteristics of the residue waste, turbidity, **pH,** phosphate and harmful heavy metals are selected as possible impact indicators.

Materials

The residue waste is from the Dalian Chemical Company and sea water is taken from outside of Dalian Bay.

Analysis

pH: pH meter Model HSD with detection limit of 0.01 pH unit. Turbidity: Turbidimeter Model GDS-3 with detection limit of 0.1 mg **1.'.**

Phosphate: Phosphomolybdate complex method; Spectrophotometer Model LGC2-1 with detection limit of 0.01μ mol 1^{-1} .

Acid soluble and water soluble heavy metals: ASV for Cu, Pb, and Zn and cold vapour AAS for Hg.

RESULTS AND DISCUSSION

Impact on heavy metals

The acid soluble and water soluble heavy metal concentrations in the residue waste from Dalian Chemical Company are shown in Table 1.

Chemical analyses show that the heavy metal content of the residue waste is very low. There will be no harmful impact to marine environment caused by heavy metals from ocean dumping of calcium carbonate residue.

Impact on pH

The pH values in mixtures of the residue and sea water with different ratios and different times after mixing are shown in Table 2. The calcium carbonate is alkaline with high pH value (~ 12) and the pH of sea water increases after residue-sea water mixing. The pH of the mixture varies with different residue/sea water ratios. The pH of 1/50 (residuekea water) mixture and 1/1OOO mixture is 9.63 and 8.42, respectively, after holding for 24 hr. Even when the sea water/residue ratio is as high as 10000/1 the pH of the mixture is still 0.4 unit higher (8.42) than the pH of sea water (8.01). This suggests that pH can be used as a good tracer of the residue for field tracking and assessing environmental impact.

Impact on Turbidity

Similarly, Table 3 shows the measured turbidity of mixtures in different residue/sea water (filtered) ratios. **When** the ratio increases from 1/100 to 1/160000 the corresponding turbidity decreases **from** 194.4 to 0.8 mg **1-'.** This indicates that ocean

Table 2 Change of pH in a mixture of CaCO₃ residue and sea water in different ratios **Table 2** Change of **pH in** a mixture of CaCO, residue and sea water **in** different ratios

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disposal of calcium carbonate residue will have a significant impact on the turbidity of sea water and turbidity can also be used as a good tracer.

$CaCO3$ residue/sea water (g cm ⁻³)	1/100	1/200	1/300	1/400	1/500	1/600	1/800
Turbidity $(mgl-1)$	194.4	131.2.	91.2	83.2	76.2	68.1	55.6
$CaCO3$ residue/sea water (g cm ⁻³)	1/1000	1/2000	1/3000	1/4000	1/6000	1/8000	
Turbidity $(mg l^{-1})$	48.6	29.6	24.6	13.8	10.8	7.4	
$CaCO3$ residue/sea water $(g \text{ cm}^{-3})$ Turbidity (mg Γ^1)	5.5	4.5	4.0	2.3	ብ ዓ	1/10000 1/16000 1/20000 1/40000 1/80000 1/160000 0.8	

Table 3 Change of turbidity in the mixture of CaCO₃ residue and sea water with different ratios

Impact of Phosphate

From the chemical composition and characteristics of $CaCO₃$ residue, one of the anticipated impacts to aquatic environment the decrease of phosphate concentration in sea water. The laboratory experiment was carried out by mixing the $CaCO₃$ residue with sea water which was spiked with different amounts of phosphate standard solution. After stirring and standing for 2hrs, the sample solutions were filtered and measured for remaining phosphate.

The rate of precipitation and adsorption of phosphate by the residue decreases with the decreasing of residue/phosphate spiked sea water ratio. In case of residue/ sea water ratio of 1/3000, the rate of precipitation and adsorption of phosphate is still about 20% of the total phosphate spiked. This is in good agreement with what we anticipated and the impact of phosphate decreasing in the water column of the dump area might be an important problem in the dumping practice of $CaCO₃$ residue.

CHEMICAL TRACKING OF CALCIUM CARBONATE RESIDUE IN OCEAN DUMPING PRACTICE

Experimental Design

Dumping area and selection of *tracers*

The dumping site for calcium carbonate residue is located about 30 miles southeast of Dalian Bay. It is about 15 square miles $(122^{\circ} 10' 00''E - 122^{\circ} 13' 45''E; 38^{\circ} 32' 00''N$ -38° 37"00"N) and was approved as a test dumping area for calcium carbonate residue by the State Oceanic Administration of China in 1985 (Figure 1).

Turbidity, pH and phosphate were selected as tracers of chemical tracking as well as for assessment of environmental impact based on the chemical composition and characteristics of residue waste and the results of laboratory simulation.

Two Research Vessel Simultaneous Investigation

Single vessel investigation does not provide information on both vertical and horizontal distributions of chemical tracers. A two research vessel simultaneous investigation was then designed. One stationary vessel (300t) is used for monitoring at the dumpsite to collect samples continuously at different depths analysing samples

on board until tracers cannot be detected. Then the vessel moved to the residue plume front indicated by a drifting drogue as a second station, again until no chemical signal can be detected in the whole dumping area. Another research vessel (800t) is used to track the pycnocline residue plume front indicated by a drifting drogue thrown into the centre of the residue plume just after the residue was dumped. The presence of residue particles above the pycnocline was monitored by acoustic tracking and chemical sampling simultaneously. Acoustic monitoring and chemical sampling were conducted continuously from the time of dumping until the signal **of** the pycnocline plume front cannot be detected. The tracking was designed to be carried out under various field conditions of flood and ebb current, close to maximum pycnocline current and close to minimum pycnocline current etc.

Tracking by Using Underwater Fluorometer

To obtain more reliable data, an underwater fluorometer was used for optical tracking of the pycnocline. Thus, three independent sets of data from acoustic, optical and chemical analyses can be matched and compared.

Baseline Conditions Prior to Dumping

The vertical distribution of T, S, σ and the depth of pycnocline, as well as the pycnocline current direction and velocity, must be determined before dumping. Also the vertical distribution of turbidity, pH and phosphate at the dumpsite and the background level of these chemical tracers at the pycnocline must be recorded.

Estimate of Deposition of Fine Particles of *Calcium Carbonate Residue in the Dispersion Area by Sediment Trap Technique*

Mooring stations with sediment traps were designed to capture the fine residue particles at selected depths in the water column both above and below the pycnocline in order to estimate the amount of deposition of residue in the dispersion area (Figure **2).** The technical design is as follows:

- **1.** to obtain the current vector diagram (Figure **3)** at different depths at the centre of the dumping area throughout five days by continuous measurement of current **(21-25** June 1989) before dumping;
- **2.** to predict the drift route of the pycnocline residue by tracking the pycnocline drifting drogue in the relatively stable tidal current direction selected from the current vector diagram (Figure 4). The tracking of the drift route was done by using two radars from two research vessels;
- **3.** the mooring system with sediment traps was set up one day after the predicting the drifting of the pycnocline; and the sediment traps were set up one day before residue was dumped;
- 4. calcium carbonate residue must be dumped at definite position and at a definite time determined by the tidal current regulation.

Sampling and Analysis

All the water samplers and bottles were soaked in 1:1 HCl, rinsed in deionized water, and sealed in plastic bags before use. Turbidity, pH and phosphate were all analyzed on board.

Figure 2 Depth of sediment traps at sites M_1 and M_2

Figure 3 Current vector diagrams

Figure **4** Drifting route of **the** pycnocline drogue

Positioning

A Model 542 microwave positioning system manufactured by ORE Company (US) was used and associated with Model Racal-Decca 970BJ navigation radar **(UK).** The precision of the positioning is ± 1 m.

RESULTS **AND** DISCUSSION

Stationary Monitoring at the Dumpsite

The variation of the vertical distribution **of** turbidity and **pH** with time after dumping at the dumpsite are shown in Figure *5.*

Figure *5* Variation of the vertical distribution **of** turbidity and pH with time after dumping at dumpsite

The maximum concentration **of** turbidity in the surface sea water *5* min after dumping was $41 \text{ mg } l^{-1}$. Twenty minutes later the turbidity in the surface sea water had almost entirely returned to the background level. The time to return to background turbidity level in the whole water column at the dumpsite was about 35 min. In the case of **pH,** because of the buffer capacity of sea water, the maximum **pH** in surface sea water was **8.48,** only **-0.3** unit higher than the **pH** of sea water before dumping. The pH of sea water in the whole water column at the dumpsite almost returned to the background level *25* min after **dumping.**

Table **4** shows the time necessary for the chemical tracers to return to background level in the whole water column at the dumpsite from data collected over three cruises.

Cruise	Dry Weight dumped (tons)	Pycnocline current velocity (cm/s)	Time for tracers to return to background (min)		
			Turbidity	рH	
6–24–1989	181	58	40	60	
6–26–1989	217	60	30	30	
8-27-1989		45	30	30	

Table 4 Time for the concentration of chemical tracers to return to background level in the whole water column at the dumpsite

Tracking of calcium carbonate residue at the pycnocline

The variation of turbidity and pH with time and dispersion distance at the pycnocline are shown in Figure 6 and Figure 7 respectively.

The variation of turbidity and pH with time and distance is very similar for all the cruises. For example, in August 23, 1989 cruise maximum turbidity and highest pH in surface sea water was recorded soon after dumping but both suddenly drop down to turbidity 2 mg I^{-1} and pH 8.220 min after the dumping and almost entirely returned to the background levels at 40-60 min after dumping.

From the data obtained from August 23, 1989 cruise, the following regression equations can be obtained:

$$
\log T = -0.8373 \log (t+1) + 1.246
$$

\n
$$
r = -0.976, n = 14
$$

\nwhere T = Turbidity, 0.5 > T < 12.9 and t = time, t > 0

$$
\log pH = -0.0061 \log (t+1) + 0.9228
$$

\n
$$
r = -0.944, n = 14
$$

\nwhere 8.18 < pH > 8.39, and t > 0

and

$$
pH = 0.0177T + 8.1835
$$

\n
$$
r = 0.947, n = 14
$$

\nwhere 0.45 > T < 12.9, and 8.18 > pH < 8.39

Equation (1) and (2) indicate that the level of turbidity and pH at the centre of the residue plume decreases exponentially with time. It follows the point source diffusion model: $C = at^{-b}$. Equation (3) explains why both turbidity and pH are good chemical tracers of residue and calcium carbonate show good correlation.

Table *5* summarizes the maximum dispersion distance, area and the time for the chemical signals to return to background levels under various pycnocline current velocities from the results of three cruises.

Results from Table *5* indicate that in the dumping area of about 15 square miles with water depth of 50 m, if 217 tons calcium carbonate residue is dumped (spot dumping) in the presence of a pycnocline with the current velocity of 60 cm/s (close to the maximum), the maximum dispersion distance of the residue plume front **is** less than 2100 m; the dispersion area is less than 0.56 km^2 ; and the maximum dispersion

 $(\Delta \times \mathrm{Rm})$ TURBIDITY

Figure 7 Variation of pH with time and dispersion distance

time is about 60 min when the turbidity and pH signals in the whole dispersion area return to the background level.

Variation of vertical distribution of chemical tracers with time and diffusion distance

The variation of vertical distribution of turbidity and pH with time after dumping and the diffusion distance from the results of August 28, 1989 cruise are shown in Figure 8.

Figure 8 Variation of the vertical distribution of turbidity and pH with time and dispersion distance

It is clear that there is a very high turbidity $(\sim 260 \text{ mg l}^{-1})$ in the surface sea water at dumpsite T_1 soon after dumping. Another peak of turbidity (\sim 35 mg $I⁻¹$) appears at the depth of 15m, possibly caused by the pycnocline. The turbidity almost entirely returns to background level in the whole water column at station T_2 , 700m distant from T_1 , 30min after dumping. In the case of pH , the vertical distribution pattern and the variation of pH with time and diffusion distance are very similar to that **of** turbidity. In addition, another important point is that the impact depth of both turbidity and pH is limited to 30m. Below this there is little difference in the chemical signals measured before and after dumping. This is in good agreement with results from the stationary monitoring at the dumpsite (Figure *5).*

Optical tracking

The results of optical tracking from July 3,1989 cruise are shown in Figures 9,10,11, and Table **6.** The amount of calcium carbonate residue dumped is217tons (dry basis) and the depth of the pycnocline is 20m. The underwater fluorometer was at a depth of 14m. Dumping started at 17:06hr and the optical signal was recorded from 17:OShr. The tracking experiment lasted for 54 minutes until the optical signal returned to the background level, 1.3-1.7 mg **I-'** at 18:02hr.

Table 6 Variation of the peak concentration in the drifting plume front with time after dumping

Time after dumping (min) 2				
Peak concentration (mg/l) 300				

Figure 10 shows that the concentration (c) of the residue particles at the centre of the plume decreases with time exponentially with the following equation:

$$
\log C = 3.26 - 1.77 \log (t+1)
$$

$$
r = -0.99, n = 7
$$
 (4)

This is in good agreement with Figures 6, 7, and equations **(l),** (2). The drifting route and dispersion area of the residue found by optical tracking are shown in Figure 11. The maximum drift distance and dispersion area obtained by optical tracking are similar to those from chemical tracking (Table *5).*

Estimate of amount of *calcium carbonate residue deposited in the dispersion area*

The technical design has already been described above. The preliminary results reported here are obtained from the July 1, 1989 cruise. The maximum drift distance is 775 m and the dispersion area is $245,000$ m². The two mooring systems with six sediment traps are just within the dispersion area, indicating that the technical design is correct. The data in Table 7 show the amount deposited from two surface traps and two middle traps. The total amount of the fine particles of calcium carbonate residue deposited in the whole dispersion area could be estimated using this technique.

Figure 9 Real time record of **underwater optical measurement**

Figure 10 Peak concentration of residue front vs. time

Figure 11 Drifting route and dispersion area of CaCO₃ residue

FIELD INVESTIGATION OF THE IMPACT OF DUMPING OF CALCIUM CARBONATE RESIDUE ON THE AQUATIC ENVIRONMENT

Design of the field investigation

The impact on the marine environment by ocean dumping of calcium carbonate residue was conducted by field investigation **of** baseline conditions before dumping compared with conditions after dumping. Three types of baseline study were carried out as follows:

- 1. five cruises in the region 12 times larger than the dumping area (180 square miles) in May 1986, July 1986, November 1986, February 1987 and May 1987;
- 2. one day cruise on June 21,1989 before dumping of the calcium carbonate residue was started;

3. sea water sampling prior to dumping on each occasion (June 24, 26; July 1, 3; August 22,23; and August 27,28,1989).

Three types of environmental impact study was carried out as described below:

- 1. time for the chemical signals to return to background level in the whole dumping area **for** each dumping occasion;
- 2. one day cruise on July 4,1989 was conducted on the day following dumping (July 3,1989);
- 3. one day cruise on September 4, 1989, one week following dumping (August 28, 1989).

RESULTS AND DISCUSSION

Table 8 Results **of** field investigations (June-July, 1989)

Table 9 Results of field investigations (August-September, 1989)

Parameter	Before/After Cruise Dumping	Date		Surface (0m) Mean Range		Depth(10m) Mean Range		Bottom (48m) Mean Range
pH	Baseline	$8 - 27 - 89$	8.19	$8.18 - 8.20$	8.07	$7.98 - 8.10$	8.03	$7.91 - 8.08$
	Impact	$9 - 04 - 89$	8.16	$8.15 - 8.18$	8.11	$8.02 - 8.18$	8.06	$8.03 - 8.20$
PO ₄	Baseline	$8 - 27 - 89$	0.04	$0.03 - 0.06$	0.41	$0.40 - 0.43$	0.64	$0.63 - 0.65$
$(\mu \text{mol} \, \text{l}^{-1})$	Impact	$9 - 04 - 89$	0.03	$< 0.01 - 0.04$	0.42	$< 0.40 - 0.44$	0.60	$< 0.52 - 0.64$

Table 8 and Table 9 show the results of the baseline survey. For July 4,1989 (Table 8) mean values of turbidity, pH and phosphate are very close to background values before dumping at all water depths one day after dumping (July 3,1989). Comparing the data from September 4 (Table 9) with those from June 21 (Table 8) and August 27, 1989 (Table 9), we found that mean values of pH and phosphate on September 4, 1989 (one week after dumping) are very close to the background mean values measured on August 27 at all water depths but rather higher than those obtained from the June 21, 1989. Thus, the impact of dumping of calcium carbonate residue is a relatively short-term effect and the time to return to background levels is from one hour (turbidity and pH) to several hours (phosphate). Table 8 shows that there is enough time for residue-sea water mixing and dilution within one day and more than sufficient time in a week. The small difference (mostly increase) between the data from June 21 and September 4,1989 could be attributed to biological activity in the summer season rather than to effects of dumping.

CONCLUSIONS

The following conclusions can be drawn:

- **1.** The ocean dumping of calcium carbonate residue causes an increase in turbidity and pH and decrease of phosphate concentration in sea water. When the sea water/residue ratio increases from 50/l to 10000/l, the pH of the mixture decreases from 9.63 to 8.42, which is still 0.4pH unit higher than the pH of original sea water (8.01) . When the sea water/residue ratio increases from $100/1$ to $160,000/1$, turbidity decreases from 194.4 mg l^{-1} to 0.8 mg l^{-1} (detection limit 0.1 mg l^{-1}). Both turbidity and pH are good tracers of the calcium carbonate residue and for environmental impact assessment. In the case of phosphate, with a phosphate spiked sea water/residue ratio of 3000/l, the rate of precipitation and adsorption of phosphate by calcium carbonate residue is still about 20% of the total phosphate.
- 2. The sedimentation rate, dispersion area and impact to the aquatic environment **by** calcium carbonate residue dumped at sea can be determined quantitatively by study of the temporal and spatial distribution **of** chemical tracers such as turbidity, pH and phosphate. In a dumping area **of** about 15 square miles with water depth of 50m, if 217tons **of** calcium carbonate residue is dumped (spot dumping) in the presence of a pycnocline with the current velocity of 60cm **s-l** (close to the maximum), the maximum dispersion distance of the residue plume front is less than 2100m; the dispersion area is less than 0.56km^3 ; the maximum impact depth is about 30m; and the maximum dispersion time is about 60min after which turbidity and pH in the whole dispersion area returns to background.
- 3. More reliable information can be obtained in the field by chemical tracking associated with acoustic and optical tracking. Three sets of data can be matched and compared. Results from chemical and optical tracking of the plume front of the pycnocline calcium carbonate residue show that turbidity and pH at the centre of residue plume decrease with time exponentially, following the point source diffusion model, $C = at^{-b}$. This is in good agreement with results from the laboratory simulation.
- 4. Sediment trap techniques can be used to estimate the amount of the fine particles **of** calcium carbonate residue deposited in the dispersion area from ocean dumping.
- 5. Results from comparative field investigations of baseline conditions and following dumping indicate that the impact of ocean dumping of calcium carbonate residue is restricted to a limited area with a limited time interval. The decrease of phosphate concentration in the upper water column is the only problem which needs to be considered carefully. The adverse impact of phosphate decrease will be greatly reduced if the specific surface area of calcium carbonate residue deposit is reduced by solidifying the residue using a filter press.
- 6. Results from chemical tracking and impact assessment show that in the dumping area of 15 square miles, dumping of 1000 tons of calcium carbonate residue (dry basis) per day is acceptable.

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