210Pb, 137Cs and 239,240Pu in East China Sea sediments: sources, pathways and budgets of sediments and radionuclides

Chih-Chieh Su, Chih-An Huh *

Institute of Earth Sciences, Academia Sinica, P.O. Box 1-55, Nankang, Taipei, Taiwan

Received 5 February 2001; accepted 12 December 2001

Abstract

Profiles of 210Pb, 137Cs and 239,240Pu measured in 83 sediment cores collected from various sedimentary regimes in the East China Sea were analyzed to elucidate the sources, routes and budgets of sediments as well as these radionuclides. Distributions of sedimentation rates and nuclide inventories reveal alongshore transport of sediments, 137Cs and 239,240Pu from the mouth of the Yangtze River toward the south, largely confined to the inner-shelf area (water depth < 70 m). Mass balance calculations suggest that the East China Sea is a sink for the particle-reactive 210Pb and 239,240Pu, with about one-sixth of their sedimentary budgets supplied via boundary scavenging. In contrast, due to lower affinity of 137Cs for particles and rapid turnover of the shelf water, the East China Sea serves as a source for 137Cs. About two-thirds of the cumulative input of 137Cs have been transported out of the East China Sea, leaving the remaining one-third stored in the bottom sediments and the overlying water column. As for the sediment budget, mass balance cannot be established due to a shortfall in sediment supply of more than 30% based on a comparison between input terms documented thus far and the sedimentation flux derived from this study. It is very likely that we have overestimated the sediment burial flux or that long-distance transport from the Yellow River’s dispersal system to the East China Sea is underestimated. Alternatively, the imbalance could be explained by the discrepancy between sediment input and output on decadal to centennial timescales. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: sedimentation; East China Sea; 210Pb; 137Cs; plutonium; Changjiang River

1. Introduction

The East China Sea (ECS) is a marginal sea over a broad continental shelf located between the largest continent (Asia) and the largest ocean (Pacific) in the world (Fig. 1). In the west and north, it receives riverine inputs from two of the largest rivers in the world, namely the Yangtze River (Changjiang) and the Yellow River (Huanghe). In the east, it is separated from the open ocean by the Kuroshio (KC), which is a western boundary current flowing northward along the edge of the continental shelf. In the middle of the ECS shelf and parallel to the Kuroshio is the Taiwan Warm Current (TWC), which flows toward the Tsushima Strait and is joined by a branch of the Kuroshio to become the Tsushima Current (TC). Through the circulation of currents and mixing of water masses, the
ECS shelf water is turned over approximately every two years (Nozaki et al., 1989; Chen and Huh, 1999), causing fairly rapid exchange of chemical materials between this marginal sea and the open ocean. Such a setting makes the ECS an interesting and important area for studying the role of marginal seas in global biogeochemistry. Therefore, it was identified as a test ground to examine the hypotheses of the Kuroshio Edge Exchange Processes (KEEP) program (Wong et al., 2000). Furthermore, it has also been realized during the course of the KEEP program that the human dimension should be included in the scope of this study as well. Because the ECS is adjacent to one of the most heavily populated region in the world currently under intense development, the impact of human activities to this marginal sea has aroused contemporaneous issues such as coastal pollution (e.g., Huh and Chen, 1999) and changes of sediment yield (e.g., Lu and Higgitt, 1998). Of special concern is the construction of the Three Gorges Dam in the upper reaches of Changjiang, which would be by far the largest dam in the world. Upon its completion and operation, scheduled for 2009, the supply of sediments to the ECS will most likely be reduced substantially. The outflow of water and various chemical materials will conceivably be changed as well. Consequently, the coastal environment may be altered irreversibly. With an uncertain future ahead, the environmental baseline data in this marginal sea remains inadequate. It is an urgent task to establish the database in a more comprehensive and systematic manner, so that the would-be changes in the future can be monitored and studied.

As a component of the KEEP program, our objective is to elucidate sedimentation processes in the ECS using a multitracer approach. For this study, we have since 1996 made five cruises and collected a large number of sediment cores throughout the ECS shelf and sometimes down the slope. The cores were analyzed for $^{210}\text{Pb}$, $^{137}\text{Cs}$ and $^{239,240}\text{Pu}$, among other radionuclides and chemical materials. This paper is built upon an earlier one (Huh and Su, 1999). Now, by integrating a substantially expanded database, we intend to delineate a more detailed picture of the sources, routes and budgets of sediments and the studied radionuclides in the ECS.

2. Materials and methods

2.1. Characteristics and provenance of sediments in the East China Sea

Based on sediment grain size together with the overlying water masses, the ECS can be divided into three regimes: the inner-shelf mud area, the outer-shelf sand area, and the slope plus Okinawa Trough mud area (Qin et al., 1987; Saito and Yang, 1994). The inner-shelf area, with water depths within ~70 m, extends southward from the Changjiang’s mouth to 26$^\circ$N along the coast of the Mainland China. The alongshore distribution of fine-grained sediments in this area manifests the dispersal of Changjiang’s plume (McKee et al., 1983). The outer-shelf area is influenced by the Taiwan Warm Current and covered primarily by relict sand and shell fragments. The slope and Okinawa Trough area is underlain by silty clay along the track of Kuroshio.

Based on distinct mineral assemblages, the ECS was also divided into three regimes by Chen et al. (1984a,b): muscovite, chlorite and calcite in the western province; orthoclase, glauconite, hornblende and epidote in the middle province; and volcanic glass, pyroxene and magnetite in the eastern province. It is informative to note that these three provinces coincide approximately with the three regimes defined by grain size and the associated water masses. Thus, it is quite obvious that the distributions of grain size, mineralogy, and other characteristics in ECS sediments reflect the combined effects of source region, topography and hydrodynamic conditions.

Although we do not have any mineralogical data from this work and our grain size data are limited, we found that the activity $^{214}\text{Pb}$ (which is available for most of the samples) correlated positively with water content in the bulk sediment but negatively with grain size. This is obviously due to the enrichment of this $^{238}\text{U}$-series nuclide in fine-grained clay minerals. Thus, we believe that
spatial and down-core distribution of this nuclide in conjunction with water content (as summarized in http://www.earth.sinica.edu.tw/~huh/keep.htm) can be used to index the variations of sediment grain size and lithological changes in the ECS.

2.2. Sampling and analytical methods

The sediment cores were collected onboard R/V Ocean Researcher-I on cruises OR-460 (August 1996), OR-493 (July 1997), OR-499 (August 1997), OR-525 (August 1998) and OR-551 (June 1999). The sampling locations are shown in Fig. 1. Near the mouth of Changjiang, where steep physical and chemical gradients are expected, the coring sites were spaced more closely. In contrast, virtually no sediment cores can be collected in the mid-shelf area under water depths of 100–200 m. This is because seafloor in that area is primarily covered by relict sand and shell fragments, which are old materials deposited in littoral zones during low sea stands in the last glacial stage. Such material is too hard to be penetrated and collected by the corer.

In order to calculate sedimentation rates and nuclide inventories more rigorously, it is necessary to recover sedimentation records of at least five decades. Thus, in addition to the collection of box cores at all sites, gravity cores were also collected in the estuarine area and at some inner-shelf sites with high sedimentation rates. Upon collection of the gravity cores or subcores from box cores, sediments were extruded vertically with a hydraulic jack and sampled at 1–2 cm intervals. The outer rim (~0.5 cm) of each sediment slab was trimmed off to minimize contamination between layers. The sectioned samples were sealed in 8-oz plastic jars and kept frozen until further process-
ing in the home laboratory. Based on weight loss after freeze-drying, the water content of the wet sediments was calculated. A correction for the amount of salt retained in the dry sediments was made based on the water content and the salinity of the bottom water. Activities and inventories of nuclides and mass accumulation rates reported in this paper are calculated on salt-free basis.

$^{210}\text{Pb}$ (via $^{210}\text{Po}$) and $^{239,240}\text{Pu}$ were determined by $\alpha$-spectrometry. $^{209}\text{Po}$ and $^{242}\text{Pu}$ obtained from ORNL were added as the yield determinants prior to total digestion of samples. The $^{209}\text{Po}$ and $^{242}\text{Pu}$ spikes have been calibrated versus NIST-certified $^{208}\text{Po}$ (SRM-4327) and $^{244}\text{Pu}$ (SRM-996), respectively. Polonium isotopes were plated onto a silver disc from the sample solution (in 1.5 N HCl, in the presence of ascorbic acid) at 80–90°C for 2 h. Isotopes of Pu were electroplated onto stainless steel discs. The counting results were corrected for the decay of $^{210}\text{Po}$ (from the time of plating to counting) and $^{210}\text{Pb}$ (from sample collection to Po plating). More detailed description of the radiochemical procedures can be found elsewhere (Huh et al., 1987, 1990, 1996).

$^{214}\text{Pb}$ (a precursor of $^{210}\text{Pb}$, used as an index of supported $^{210}\text{Pb}$) and $^{137}\text{Cs}$ were measured by $\gamma$-spectrometry based on photon energies at 351.99 keV and 661.62 keV, respectively. The counting system is equipped with a 150% efficiency (relative to $3\times3\text{ NaI}$) HPGe detector (EG and G ORTEC GEM-150230) interfaced to a digital $\gamma$-ray spectrometer (DSPec®). The detector was calibrated using NIST SRM 4353 (Rocky Flats soil) and 4350B (Columbia River sediments), and IAEA standard SD-N-1 (Irish Sea sediments). At 661.62 keV, for example, the absolute counting efficiency for our samples (in plastic jars of 8.5-cm diameter, 7-cm height) varied from 6.15% for 10-g samples to 4.66% for 100-g samples, and the peak resolution was 1.21 keV (FWHM) with a peak-to-Compton ratio higher than 90.

3. Results and discussion

Space limitation does not allow us to list the very massive nuclide activity data here. Interested readers can browse the complete data set (plotted as downcore profiles) at the web site http://www.earth.sinica.edu.tw/~huh/keep.htm. In what

![Figure 2](image-url)
follows, we choose to present some representative profiles and show salient features. The overall results on sedimentation rates and nuclide inventories are plotted in figures and contour maps to facilitate the discussion.

3.1. Spatial variation of sedimentation rate

A primary objective of this study was to derive sedimentation rates over the ECS, so that the history of deposition and burial fluxes of radionuclides and various chemical materials can be studied by us as well as other components of the KEEP program. In calculating sedimentation rates, all nuclides analyzed (namely $^{210}\text{Pb}$, $^{137}\text{Cs}$ and $^{239,240}\text{Pu}$) are employed as sediment chronometers and the results are compared. Given in Fig. 2 is an example from OR499-16, a core located at a depocenter in the estuary area. It shows that sedimentation rates calculated from the downward decrease of excess $^{210}\text{Pb}$ and the subsurface peaks of $^{239,240}\text{Pu}$ and $^{137}\text{Cs}$ (taken to be the fallout maximum circa 1963; Huh and Su, 1999) compare favorably with one another. Fig. 3 shows that, for cores collected from Changjiang’s estuary and the inner shelf areas to the south, good agreement between sedimentation rates derived from different tracers is generally observed. This is primarily due to the fact that sedimentation rates in these near shore areas are fast enough to dominate over other terms in controlling profiles of nuclides. Away from the near shore areas, the much lower sedimentation rates render post depositional processes (i.e., mixing and diffusion) important in modifying the distribution of nuclides. Consequently, sedimentation rates based on different tracers become less consistent.

Despite the varied degree of consistency between sedimentation rates derived using different methods for different sedimentary regimes in the ECS, the spatial distribution and variation can still be gleaned based on any of the nuclides (please see http://www.earth.sinica.edu.tw/~huh/keep.htm). Because the data are more complete for $^{137}\text{Cs}$, we choose to use $^{137}\text{Cs}$ profiles to show spatial trends. Shown on Fig. 4 are $^{137}\text{Cs}$ profiles at sites along five lines (A, B, C, D, E and F) in the cross-shelf direction and one line (G) alongshore in the inner shelf. Along lines D, E and F, the depth of the subsurface maximum and/or the penetration depth of $^{137}\text{Cs}$ deepen toward the shore and pinch out toward the east, indicating offshore decrease of sedimentation rates. The same trend can also be seen on line G, but the deepening is alongshore toward the mouth of Changjiang. Also following the systematic trend is the downcore inventory of $^{137}\text{Cs}$ (Fig. 6a), which decreases offshore toward the east and alongshore toward the south. All these point to the fact that Changjiang is a major source of sediments and $^{137}\text{Cs}$ in the ECS shelf.

Along lines A, B and C, the picture is complicated by localized effect of topography and water flow, as explained below. There is a belt of abnormally low sedimentation rate trending in the NW– SE direction and crossing the middle of lines A
This belt coincides with a submarine canyon, which is a drowned relict river valley off the mouth of the old Changjiang. It is important to note that, up the canyon toward the northwest flows a branch of the Taiwan Warm Current, which causes year-round upwelling off the Changjiang estuary (Chao, 1993; Jacobs et al., 2000). Because the flow of the Taiwan Warm Current and the topographically induced upwelling is much stronger than the river plume and along-shore current, it impedes offshore transport of riverine sediments across the valley (Zhang, 1999). Besides low sedimentation rates, inventories of all fallout nuclides also show minimum values along the axis of the valley (to be further discussed later).

Fig. 5 provides an overview of the spatial distribution of sedimentation rates in the indicated area. It was derived from $^{137}$Cs-based sedimentation rates taking the subsurface $^{137}$Cs maximum as the time horizon of 1963. The contour lines are plotted by the SURFER software package. Aside from the general decrease of sedimentation rates offshore from the largest sediment source in the ECS (i.e., Changjiang), the zone of minimum sedimentation mentioned above emerges as a rather pronounced feature.

3.2. Sources, pathways and budgets of nuclides

3.2.1. Distribution of $^{137}$Cs inventory

Shown in Fig. 6 are sediment inventories of $^{137}$Cs, excess $^{210}$Pb and $^{239,240}$Pu in the ECS. The distribution of $^{137}$Cs inventory (Fig. 6a) mimics the pattern of sedimentation rate (Fig. 5), indicating again that riverine input is a major source for this anthropogenic nuclide in the ECS. Considering that the area of Changjiang’s drainage basin
(1 800 000 km²; Milliman and Jin, 1985) far exceeds that of the ECS (353 000 km²; used for this study), it is quite conceivable that delayed wash-in input of 137Cs from the drainage basin may be more important than direct fallout from the atmosphere above the ECS. This point can be further argued by comparing observed sediment inventories of 137Cs with expected cumulative fallout as follows. Inventories of 137Cs in outer shelf sediments (< 1 dpm cm⁻²) are much lower than that can be expected from global fallout, which is 7.1 dpm cm⁻² as of January 1998 at this latitudinal band (Huh and Su, 1999), indicating that much of the fallout 137Cs still resides in the seawater reservoir. On the other hand, the sediment inventory of 137Cs near Changjiang’s mouth (e.g., 53 dpm cm⁻² in core OR499-16) is seven to eight times higher than expected from cumulative fallout, which is primarily due to the deposition of 137Cs associated with river-borne particles. In other words, scavenging from the water column alone cannot account for the observed 137Cs inventories. More on this will be discussed by mass balance calculations later.
3.2.2. Distribution of excess $^{210}$Pb inventory and an evaluation of riverine input

The distribution of excess $^{210}$Pb inventory in sediments is quite different from that of $^{137}$Cs in showing high values at the shelf edge and in the southern part of the inner shelf (Fig. 6b). Riverine input does not seem to be as pronounced for $^{210}$Pb as it is for $^{137}$Cs, and sediment inventories of excess $^{210}$Pb in much of the estuary area is even lower than expected from global fallout (Huh and Su, 1999). Its cause can be revealed from mass balance calculations below.

We are not aware of any published data for $^{210}$Pb in the Changjiang drainage basin or the river water, thus it is not possible to get an accurate estimate of the riverine input of this radionuclide. Although several previous studies (e.g., Benninger, 1978; Olsen et al., 1986; Baskaran and Santschi, 1993) have indicated the insufficiency of riverine input relative to the atmospheric input of $^{210}$Pb at some estuaries, it has also been shown that, for estuaries with large drainage basin to estuarine area ratios, riverine input could be an important $^{210}$Pb source (Ravichandran et al., 1995; Baskaran et al., 1997). With a drainage basin area of 1 800 000 km$^2$, which is about 50 times that of the estuary area (35 000 km$^2$), the Changjiang estuary certainly falls in the latter category. Based on this area ratio and by taking 3000 yr as the residence time of $^{210}$Pb in the drainage basin (Smith and Ellis, 1982; Dominik et al., 1987), the riverine input of $^{210}$Pb is roughly estimated using the same equation applied in previous studies (Ravichandran et al., 1995; Baskaran et al., 1997), i.e.,

$$I_d = \frac{A_d}{A_{es}} \times I_f \times I_e$$

where $A_d$ is the area of the drainage basin, $A_{es}$ is the area of the estuary, $I_f$ is the inventory of $^{210}$Pb (58 dpm cm$^{-2}$; DeMaster et al., 1986) and $I_e$ is the fraction of the inventory of $^{210}$Pb eroded each year from the watershed ($I_e = \ln 2/3000 \text{ yr} = 2.3 \times 10^{-4} \text{ yr}^{-1}$). The calculation results in a riverine flux of about 0.69 dpm cm$^{-2}$ yr$^{-1}$, corresponding to 38% of the atmospheric flux.

It should be noted that, the residence time of $^{210}$Pb in the Changjiang drainage basin is a highly uncertain factor. Although we used 3000 yr as the drainage basin residence time of $^{210}$Pb in the calculation above, values from 800 to 10 000 yr have

Fig. 6. Contour maps showing spatial distribution of sediment inventories of (a) $^{137}$Cs, (b) excess $^{210}$Pb and (c) $^{239,240}$Pu in the East China Sea.
been estimated for various drainage basins around the world (Benninger et al., 1975; Lewis, 1977; Smith and Ellis, 1982; Dominik et al., 1987; Porcelli et al., 2001). Considering that the drainage area of the Changjiang River is among the largest in the world, it may be reasonable to take 10,000 yr as the residence time and hence $7 \times 10^{-5}$ yr$^{-1}$ for $k_e$ in Eq. 1. If so, the riverine flux of $^{210}$Pb is reduced to 0.21 dpm cm$^{-2}$ yr$^{-1}$, corresponding to $\sim 12\%$ of the atmospheric flux.

3.2.3. Pu distribution and inventory in sediments

Because of the time-consuming and laborious procedures for the separation and purification of Pu, $^{239,240}$Pu analysis was performed only on selected cores. Thus, the inventory of $^{239,240}$Pu in sediments (Fig. 6c) is depicted less clearly. Although $^{239,240}$Pu has the same source as $^{137}$Cs (both come from nuclear tests), it behaves more like $^{210}$Pb in terms of affinity for particles. As with $^{137}$Cs, sediment inventory of $^{239,240}$Pu increases offshore, indicating that riverine input is an important source. Unlike $^{137}$Cs, however, is that sediment inventory of $^{239,240}$Pu in the eastern side of the ECS is close to or higher than that (0.21 dpm cm$^{-2}$; Hardy et al., 1973) expected from global fallout, indicating that $^{239,240}$Pu is more effectively scavenged from the water column by settling particles.

3.3. Sedimentary budgets of nuclides

In what follows, an attempt is made to calculate the mass balance of these nuclides. To facilitate the calculation, the area covered in this study is divided into four boxes: estuary, inner shelf, outer shelf, and slope (Fig. 7). Although the compartmentalization is a bit arbitrary in some places, it is basically based on the circulation pattern, topography and grain size distribution, as described earlier. The estuary area is bound between 30$^\circ$N and 32$^\circ$N, with its eastern border set at 60 m isobath from 30$^\circ$N to 31$^\circ$N and along the axis of the old Changjiang relict valley from 31$^\circ$N to 32$^\circ$N. Sediments in this region are mainly silt in the north and clay in the south. The inner shelf area is defined as the coastal zone at the south of the estuary, between 26$^\circ$N and 30$^\circ$N and at water depth within 70 m. This area is along the path of the Changjiang Coastal Water and is covered mainly (70% on average) by clay-sized ($<63$ $\mu$m) sediments. The outer shelf area is the rest of the continental shelf extending to the shelf break at 200-m water depth. The Taiwan Warm Current is the dominant flow in this area where most of the seafloor is underlain by sand, silty sand and shell fragments. The slope area is a relatively narrow band between the 200-m and 1500-m isobaths, stretching from 26$^\circ$N to 31$^\circ$N. It coincides with the path of the Kuroshio where the bottom sediments are clay or silty clay. The surface areas of the four boxes are 35,000, 65,000, 230,000 and 23,000 km$^2$, respectively. The corresponding mean depths estimated using the SURFER program based on NOAA's ETOPO5 bathymetry database are approximately 50 m, 50 m, 80 m and 1000 m, respectively. Thus, the seawater volumes are 1750, 3250, 18,400 and 23,000 km$^3$, respectively.

First, it is more straightforward to begin the calculation for $^{210}$Pb, because riverine input is less important for this nuclide (Huh and Su, 1999; Lin et al., 1996), which can be calculated by:

$$0.69 \text{ dpm cm}^{-2} \text{ yr}^{-1} = 0.031 \text{ yr}^{-1} \times 36,000 \text{ km}^2 = 7.7 \times 10^{15} \text{ dpm}.$$  

If the $^{210}$Pb flux of 1.8 dpm cm$^{-2}$ yr$^{-1}$ in the nearby Hangzhou City (De-Master et al., 1985) is applied to the estuary box, atmospheric input would account for $20.3 \times 10^{15}$ dpm of excess $^{210}$Pb in sediments of the estuary box at steady state. Production of $^{210}$Pb from the decay of $^{226}$Ra (taking the mean $^{226}$Ra concentration to be 0.18 dpm l$^{-1}$; Lin et al., 1996) in the water column would contribute to $0.3 \times 10^{15}$ dpm. The actual amount of excess $^{210}$Pb buried in the estuary box is estimated to be $11.6 \times 10^{15}$ dpm. To balance the budget, $\sim 59\%$ of the total excess $^{210}$Pb input must be exported from the estuary box. Indeed, summer deposition followed by winter resuspension is a perennial process in the Changjiang’s estuary (McKee et al., 1983; Liu et al., 2000), which may very well be responsible for the observed deficiency in the sedimentary budget of $^{210}$Pb over there.

Sediments removed from the estuary are transported by the alongshore current and deposited in the inner shelf area in the south (Huh and Su,
In the inner shelf box, the import of excess \(^{210}\text{Pb}\) from the estuary \((16.7 \times 10^{15} \text{ dpm})\) combined with atmospheric input \((37.7 \times 10^{15} \text{ dpm})\) and water column production \((0.4 \times 10^{15} \text{ dpm})\) contribute to 82% of the total burial in sediments \((66.9 \times 10^{15} \text{ dpm})\). The remaining 18% (or \(19.8 \times 10^{15} \text{ dpm}\)) must be from the offshore area, most likely via the transport of the Taiwan Warm Current. Performing the same calculation for the outer shelf and the slope areas in sequence, we found that lateral input from offshore, or the so-called ‘boundary scavenging’ (Bacon et al., 1976; Spencer et al., 1981), is required to balance the budget for excess \(^{210}\text{Pb}\). This is conceivably promoted by the flow of the Kuroshio along the shelf edge and its intrusion onto the shelf at some places such as the northeast of Taiwan (Hsueh et al., 1992; Tang et al., 2000) and at 29°N (Nitani, 1972). The above calculation is summarized schematically in Fig. 8. If all four sedimentation regimes are combined into a single domain, it shows that atmospheric fallout is responsible for 76% of the excess \(^{210}\text{Pb}\) budget in the ECS as a whole, with the remaining 24% derived from boundary scavenging (17%), riverine input (4%) and in situ production from \(^{226}\text{Ra}\) (3%).

In calculating the budget for \(^{239,240}\text{Pu}\) and \(^{137}\text{Cs}\), the four boxes discussed above are merged...
into one to avoid over-interpretation, which is necessary in light of the uncertainty in riverine inputs for these two anthropogenic nuclides. First, in view of the similarity in geochemical behavior between Pu and 210Pb, it may be reasonable to assume that, as with 210Pb, boundary scavenging also accounts for 17% of the sedimentary budget of 239\(^{\text{Pu}}\)240. Thus, of the total 239\(^{\text{Pu}}\)240 stored in sediments (2.5 \(\times\) \(10^{15}\) dpm), 0.4 \(\times\) \(10^{15}\) dpm is derived from boundary scavenging. Then, with total atmospheric input fixed at 0.7 \(\times\) \(10^{15}\) dpm, the riverine input of 239\(^{\text{Pu}}\)240 is estimated to be 1.4 \(\times\) \(10^{15}\) dpm, accounting for 56% of the sedimentary budget of 239\(^{\text{Pu}}\)240 in the ECS. This is summarized in Fig. 9. The distribution of 239\(^{\text{Pu}}\)240Pu inventory in the estuary area (Fig. 6c) is consistent with large riverine input. One may question why riverine input is so much more important for Pu than for 210Pb. A very possible cause is the release of Pu (as well as 137Cs) from

**Budget of \(^{210}\text{Pb}_{\text{ex}}\) in the East China Sea (10^{15}\) dpm)**

Fig. 8. Schematic diagram showing the mass balance of excess \(^{210}\text{Pb}\) in the East China Sea based on the four-box model. The unit (10^{15}) dpm for the numbers results from flux (dpm cm\(^{-2}\) yr\(^{-1}\))\(\times\)area (cm\(^2\))\(\times\)10\(^{15}\), where \(\lambda\) (\(\times\)0.031 yr\(^{-1}\)) is the decay constant of \(^{210}\text{Pb}\).

**Budget of Pu in the East China Sea (10^{15}\) dpm)**

Fig. 9. Mass balance of 239\(^{\text{Pu}}\)240Pu in the East China Sea. The unit (10^{15}) dpm for the numbers results from cumulative fallout or sedimentary inventory (dpm cm\(^{-2}\)\(\times\)area (cm\(^2\))\(\times\)10\(^{15}\).
nuclear facilities along Changjiang and its tributaries (Norris et al., 1993), whose discharges are unknown, and hence not considered in our budget calculation.

The mass balance calculation for $^{137}$Cs (shown in Fig. 10) is complicated by its lower affinity for particles and, therefore, partition between dissolved and particulate forms. Based on our previous estimate, direct precipitation from the atmosphere over the ECS would lead to a cumulative $^{137}$Cs fallout of 7.1 dpm cm$^{-2}$ (Huh and Su, 1999), thus a total standing stock of $25 \times 10^{15}$ dpm (assuming no loss from the ECS). If $^{137}$Cs bears the same riverine input to atmospheric input ratio as Pu, then riverine input of $^{137}$Cs would result in a cumulative input of $50 \times 10^{15}$ dpm, which could be an underestimate in view of the fact that $^{137}$Cs has lower affinity for particles, hence is expected to be washed off the drainage basin more easily than Pu. So, the sum of total $^{137}$Cs input (riverine plus atmospheric) would be at least $75 \times 10^{15}$ dpm, about four times that buried in sediments (i.e., $19 \times 10^{16}$ dpm). To account for the difference, we still need to estimate the standing stock of $^{137}$Cs in the seawater reservoir, which is about one-half of that buried in sediments. With the combined standing stock ($28 \times 10^{15}$ dpm) subtracted from the total input, we still have at least $47 \times 10^{15}$ dpm unaccounted for, which can only be explained by transport of $^{137}$Cs out of the ECS in dissolved and suspended forms. Export of $^{137}$Cs can be facilitated by rapid turnover of the ECS shelf water ($\sim 2\text{ yr}$ residence time; Nozaki et al., 1989; Chen and Huh, 1999). In summary, the amount of $^{137}$Cs exported out of the ECS is $\sim 2\text{--}3$ times that buried in sediments, suggesting that this marginal sea is an effective source of $^{137}$Cs to other ocean basins.

### 3.4 Sediment budget

With the updated database, a re-evaluation of the sediment budget in the ECS may be in order. Based on the spatial distribution of sedimentation rates and using the SURFER program, an annual deposition of approximately $13 \times 10^8$ tons in the covered area is calculated. This deposition flux is more than the reported discharge from Changjiang ($5 \times 10^8$ tons yr$^{-1}$; Milliman and Meade, 1983) plus the erosion from Taiwan ($2.6 \times 10^8$ tons yr$^{-1}$; Water Resources Planning Commission, 1996). As discussed earlier, much of the sediments discharged from Changjiang are deposited in the estuary and the inner shelf area to the south, which can be largely substantiated by the
distribution of sedimentation rates in these two areas. So, the problem may rest on the offshore areas. The circulation pattern of currents suggests that the Taiwan Strait in the south and the Yellow Sea in the north are the possible source regions for sediments in the vast mid-shelf and outer shelf regions. However, even if all sediments eroded from Taiwan can be transported to the ECS shelf (by the Taiwan Warm Current), we still need an annual input of $5.4 \times 10^8$ tons from the Yellow Sea to balance the budget. This additional input is about one-half of the reported annual discharge from the Yellow River (Milliman and Meade, 1983; Schubel et al., 1984). Previous studies (Zhang, 1996; Lee and Chough, 1989) suggested that only a small portion ($\sim 15\%$) of the sediment load discharged by the Yellow River is transported southward into the ECS by the Yellow Sea Coastal Current (YSCC). Could it be that long-distance transport from the Yellow River and Yellow Sea to the ECS, perhaps via reworking of sediments, is more than previously thought? From a different standpoint, it is also highly likely that sedimentation rates we obtained for the middle and outer shelf sediments may be overestimated due to sediment mixing, reworking, or improper interpolation of their spatial distribution (e.g., over the mid-shelf area covered by relict sediments where there is probably no net sedimentation).

Although it is not yet possible to establish mass balance for sediments in the ECS, we have obtained a mean sediment burial flux in the ECS shelf that is fairly comparable with the flux of sediment derived from the adjacent basins and seas. It should be pointed out that sedimentation rates deduced here are on time scales of a few decades (based on $^{137}$Cs and $^{239,240}$Pu) or up to $\sim 100$ yr (based on $^{210}$Pb). Even if the documented values of river discharge or land erosion are reasonably correct, it is unknown how variable they could be on decadal to centennial time scales in response to environmental changes, either natural or induced by human activities. In other words, insofar as the covered time is concerned, we are probably only looking at a transient state rather than a steady state condition.

4. Conclusions

Based on profiles of $^{210}$Pb, $^{137}$Cs and $^{239,240}$Pu measured in 83 sediment cores collected from various parts of the ECS, spatial distributions of sedimentation rates and nuclide inventories in this marginal sea are delineated. The results lead us to the following conclusions:

(1) The highest sedimentation rates ($>2$ g cm$^{-2}$ yr$^{-1}$) are found in the estuary of Changjiang, the largest source of sediments in the ECS. There exists a general decrease of sedimentation rates from the mouth of Changjiang alongshore toward the south and offshore toward the east. Superimposed on the overall trend is a small but pronounced belt of abnormally low sedimentation along the valley of a submarine canyon, which is the drowned relict river valley off the mouth of the old Changjiang. The strong flow up the canyon (i.e., a branch of the Taiwan Warm Current) serves as a barrier preventing Changjiang’s sediment plume from crossing the valley.

(2) Spatial distributions of sediment inventories of $^{137}$Cs and $^{239,240}$Pu in general follow the distribution of sedimentation rates, suggesting that outflow from Changjiang is a dominant source of these two anthropogenic nuclides. For $^{210}$Pb, riverine input is much less than direct precipitation from the atmosphere over the ECS.

(3) The effect of boundary scavenging for the particle-reactive $^{210}$Pb and $^{239,240}$Pu is enhanced by the transit of the Kuroshio and the Taiwan Warm Currents. This process is responsible for about one-sixth of the sedimentary inventories of these two nuclides in the ECS. By contrast, the ECS as a whole serves a source for the somewhat conservative $^{137}$Cs. Of the total $^{137}$Cs delivered into the ECS (from the drainage basin and the atmosphere), only about one-third resides in the ECS (sediments plus water column); the majority of the remainder is transported out.

(4) From the distribution of sedimentation rates, a sedimentation flux of $13 \times 10^8$ tons yr$^{-1}$ in the ECS is calculated. This is much more than the sum of Changjiang’s sediment discharge ($5 \times 10^8$ tons yr$^{-1}$) and the sediment yield of Taiwan ($2.6 \times 10^8$ tons yr$^{-1}$). It could be that sedi-
ment input versus output in the ECS is out of equilibrium on decadal time scales, or that long-distance transport from the Yellow River’s dispersal system to the ECS is more than previously thought.

Finally, in order to put what we have learned from the ECS into a global perspective, it should be informative to compare our results with corresponding data for the Amazon continental shelf. As summarized in Table 1, the Changjiang River and the Amazon River are fairly comparable in some attributes; both are among the largest rivers in the world. However, there is a contrasting difference in their drainage basin to shelf area ratios: \(~2\) for the ECS shelf and \(~78\) for the Amazon shelf. Consequently, the impact of riverine input as a source for \(^{210}\text{Pb}\) (and presumably for other fallout nuclides as well) is much greater for the latter, whereas atmospheric input is more important for the former. As for the effect of boundary scavenging (i.e., input from the open ocean), it also appears to be more pronounced at the narrow Amazon shelf than at the broad ECS shelf on per unit area basis.

### Acknowledgements

We are thankful to Y.-C. Huang, K.-J. Chang, S.-F. Lu, and S.-Y. Lee for sampling at sea and/or radiochemical analysis in the laboratory. The constructive reviews by the editor and Drs. S.A. Kuehl, M. Baskaran and M.P. Bacon greatly improved the manuscript. This work is supported by National Science Council Grants NSC 87-2611-M-001-002, 88-2611-M-001-002, and 89-2611-M-001-002.

### References


---

### Table 1

Comparisons of some attributes between: (a) the Changjiang and the Amazon Rivers and (b) the East China Sea and the Amazon continental shelves

<table>
<thead>
<tr>
<th>(a) Comparison of river length, drainage basin area, water and sediment discharge, and sediment yield</th>
<th>Changjiang River</th>
<th>Amazon River</th>
</tr>
</thead>
<tbody>
<tr>
<td>River length (km)</td>
<td>6300</td>
<td>6400</td>
</tr>
<tr>
<td>Drainage area (km²)</td>
<td>(1.8 \times 10^9)</td>
<td>(6.9 \times 10^6)</td>
</tr>
<tr>
<td>Water discharge (m³ yr⁻¹)</td>
<td>(9 \times 10^{12})</td>
<td>(6 \times 10^{12})</td>
</tr>
<tr>
<td>Sediment discharge (tons yr⁻¹)</td>
<td>(0.5 \times 10^9)</td>
<td>(1.2 \times 10^9)</td>
</tr>
<tr>
<td>Drainage basin sediment yield (tons km⁻² yr⁻¹)</td>
<td>278</td>
<td>174</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>(b) Comparison of the area and (^{210}\text{Pb}) sources in the continental shelf</th>
<th>East China Sea</th>
<th>Amazon Shelf</th>
</tr>
</thead>
<tbody>
<tr>
<td>Shelf area (km²)</td>
<td>(90 \times 10^4)</td>
<td>(8.9 \times 10^4)</td>
</tr>
<tr>
<td>Atmospheric flux (dpm cm⁻² yr⁻¹) Nearshore 1.8ᵃ</td>
<td>Nearshore 1.15ᵇ</td>
<td>0.33ᶜ</td>
</tr>
<tr>
<td>Atmospheric fallout (%)</td>
<td>76.4</td>
<td>2.3ᵈ</td>
</tr>
<tr>
<td>Riverine input (%)</td>
<td>3.9</td>
<td>31ᵈ</td>
</tr>
<tr>
<td>In situ production (%)</td>
<td>2.9</td>
<td>0.1ᵈ</td>
</tr>
<tr>
<td>Open ocean input (%)</td>
<td>16.8</td>
<td>67ᵈ</td>
</tr>
</tbody>
</table>

ᵃ DeMaster et al. (1985)
ᵇ Huh (unpublished data)
ᶜ DeMaster et al. (1986)
ᵈ Smoak et al. (1996)
Discharge of the Changjiang (Yangtze River) into the East China Sea. Cont. Shelf Res. 4, 57–76.


Saito, Y., Yang, Z., 1994. Historical change of the Huanghe (Yellow River) and its impact on the sediment budget of the


