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► **To cite this version:**

Sabine Schmidt, Jean-Louis Reyss. Radium as internal tracer of Mediterranean Outflow Water. Journal of Geophysical Research. Oceans, 1996, 101 (C2), pp.3589-3596. 10.1029/95JC03308. hal-03545638

**HAL Id: hal-03545638**

**<https://hal.science/hal-03545638>**

Submitted on 27 Jan 2022

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## Radium as internal tracer of Mediterranean Outflow Water

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**Abstract.** Radium-226 and  $^{228}\text{Ra}$  data are presented for the Western Mediterranean Sea and the NE Atlantic. In the NW Mediterranean Sea, the near-shore surface waters of the Ligurian Current contain much higher  $^{228}\text{Ra}$  concentrations than surface waters collected in the open Ligurian seawater. The gradient of  $^{228}\text{Ra}$  along a Villefranche - Calvi transect has been used to calculate values of the horizontal eddy diffusion coefficient perpendicular to the flow direction of the Ligurian Current of about  $10^6 \text{ cm}^2 \text{ s}^{-1}$ . The observed decay of  $^{228}\text{Ra}$ , as the Mediterranean Deep Water flows toward the Gibraltar Straits, is in good agreement with the assumption of a rapid turnover of the Mediterranean basin. Concentration profiles of  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  were measured in the Mediterranean Outflow Water (MOW) off the Gibraltar Straits in the NE Atlantic basin at two stations,  $36^\circ 30' \text{N} - 15^\circ 35' \text{W}$  and  $36^\circ 27' \text{N} - 10^\circ 35' \text{W}$ . These sites showed the hydrographic characteristics of Meddies (MOW which propagates as eddies in the NE Atlantic) at intermediate depth. At the center of the Meddies, around 1000 m depth, the activity of the short half-life  $^{228}\text{Ra}$  increases up to  $0.9 \times 10^{-2}$  disintegrations per minute per liter, higher than usually found in North Atlantic Deep Water. This "young"  $^{228}\text{Ra}$  results from the contact, a few months earlier, of the MOW with the Spanish continental shelf in the vicinity of the Gibraltar Straits. The decay of  $^{228}\text{Ra}$ , used as an internal tracer of water mass motion, leads to a transit time of the Meddy *Yseult* of less than 1 year since its formation.

### Introduction

Mediterranean Outflow Water (MOW) produces a warm and saline layer at around 1000 m in the NE Atlantic Ocean. This Mediterranean Water displays chemical and biological properties which are different from those of the surrounding Atlantic Water [Savenkoff *et al.*, 1993]. MOW can propagate as eddies, called Meddies. Meddies are mesoscale features, typically 40-100 km in diameter with mean life around 3 years [Armi *et al.*, 1988; Rossby, 1988]. Their formation process begins when MOW crosses the Gibraltar Strait. The presence of Meddies has implications for lateral dispersion of heat, salt, and pollutants [Marshall, 1988].

To explain the biological and chemical composition of Meddies, it is necessary to define the characteristics of the MOW which constitutes most of the water mass of Meddies and to evaluate the translation velocity. The time elapsed since the formation of a Meddy is useful for interpreting the composition of the saline water and for evaluating its influence on the surrounding environment.

Much effort has been devoted to the study of Meddies off the Canaries archipelago. Most of this has involved hydrodynamic study of the motion of one Meddy, tracked with drifting buoys [Armi *et al.*, 1989]. The quantitative evaluation of large-scale mixing across ocean basins is also possible from the determination of naturally occurring radioisotopes,  $^{228}\text{Ra}$  ( $t_{1/2} = 5.75$  years) and  $^{226}\text{Ra}$  ( $t_{1/2} = 1600$  years) [Kaufman *et al.*, 1973; Sarmiento *et al.*, 1982; Moore and Santschi, 1986]. Both Ra isotopes are supplied to the ocean by deep-sea and continental shelf sediments. A water mass in contact with sediment is enriched with Ra diffusing

out of sediments. The  $^{228}\text{Ra}/^{226}\text{Ra}$  activity ratio will only change by dilution and radioactive decay after the water is isolated from sedimentary input. The  $^{228}\text{Ra}/^{226}\text{Ra}$  ratio will decrease with increasing distance from sediments and with depth within the thermocline [Yamada and Nozaki, 1986; Reid *et al.*, 1979; Moore and Todd, 1993]. The  $^{228}\text{Ra}$  half-life is thus suitable for studying oceanic processes occurring on a 1- to 30-year timescale.

The objective of the work described herein has been to determine the temporal evolution of Meddies. The study concerns their origin, and in particular the Deep Mediterranean Water, and the comparison of two Meddies at different states. We used  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  tracers and associated physical measurements. Significant efforts have been necessary to improve sampling and analytical methods for radium isotopes.

### Methods

Radium determinations were made on water samples collected from different sites in the NW Mediterranean Sea and NE Atlantic Ocean to follow Deep Mediterranean Water motion. The geographical location of stations is shown in Figure 1. To survey Meddies, which are anomalous in their temperature and salinity characteristics compared with the surrounding water masses (typically  $> 11.9^\circ\text{C}$  and 36.2 practical salinity unit (psu)). These profiles led us to select two sites for which temperature and salinity indicated the presence of Meddies at intermediate depths:  $36^\circ 27' \text{N} - 10^\circ 35' \text{W}$  (Medatlante 1 - January 1989 - *Nicole*) and  $36^\circ 30' \text{N} - 15^\circ 35' \text{W}$  (Medatlante 2 - August 1989 - *Yseult*).

In seawater the very low  $^{228}\text{Ra}$  content of about  $5 \times 10^{-3}$  disintegrations per minute per liter (dpm  $\text{L}^{-1}$ ) implies either the extraction of Ra from large volume of seawater or more sensitive measurement methods [Orr, 1988]. First, we used large volumes of seawater sampled with a submersible,

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Paper number 95JC03308.  
0148-0227/96/95JC-03308\$05.00

battery-powered pumping system developed at Centre des Faibles Radioactivités (CFR). In this way, 300 to 800 L of water were filtered in situ. The water was forced through a membrane filter (0.4- $\mu\text{m}$  pore size), then through a  $\text{MnO}_2$ -coated cartridge previously prepared at the laboratory with a procedure based on that of Reid et al. [1979], and finally through a flowmeter to record the volume of filtered water. The cartridge efficiency for Ra was obtained from an independent 30-L sample on which  $^{226}\text{Ra}$  specific activity was measured.

For radiochemical analyses, the cartridge was soaked with hot concentrated HCl in the presence of a small amount of reducing agent (hydroxylamine hydrochloride). When the digestion was performed, the cartridge was removed and rinsed with deionized water. Ra was isolated by coprecipitation with a Ba carrier. Our  $\text{BaCO}_3$  carrier has a radium activity less than 0.005 dpm/g. The precipitate was recovered, rinsed with 8N HCl and dried prior to analysis by  $\gamma$  spectrometry. The two standards used for the calibration of the  $\gamma$  detector are (1) a mock-up of sediment and U and Th U.S. standards from National Bureau of Standards at 1000 ppm and (2) a matrix formed by precipitation of sulphates in the presence of a Ba solution enriched with  $^{226}\text{Ra}$  standard (Harwell) [Reyss et al., 1995]. The efficiency of Ra extraction

by  $\text{BaSO}_4$  precipitation was obtained in two ways: by calculating the weight ratio of recovered  $\text{BaSO}_4$  to introduced  $\text{BaCO}_3$  (corrected by a mass factor of 1.18) and by measuring  $^{225}\text{Ra}$ , counted along with  $^{228}\text{Ra}$ , this spike being added prior to extraction. Extraction efficiencies are in the range 75-100%, with good agreement between the two methods. On the 30-L sample, we isolated  $^{226}\text{Ra}$  by coprecipitation with  $\text{Ba}(\text{SO})_4$ . The subsequent treatment was the same as described above.

A 220-cm<sup>3</sup> low-background, well-type gamma ray detector in the Laboratoire Souterrain de Modane (LSM - Centre National de la Recherche Scientifique (CNRS)/Commissariat à l'Énergie Atomique (CEA)), allows us to measure  $^{228}\text{Ra}$  from only 30 L. of seawater with a precision of  $\pm 15\%$  for activity levels lower than  $5 \times 10^{-3}$  dpm L<sup>-1</sup>. Thus during Medatlante 2, we used only 30-L samples on which we isolated and measured  $^{226}\text{Ra}$  and  $^{228}\text{Ra}$  as described above. This new experimental strategy allows a larger number of analyses with reduced shiptime and laboratory effort.

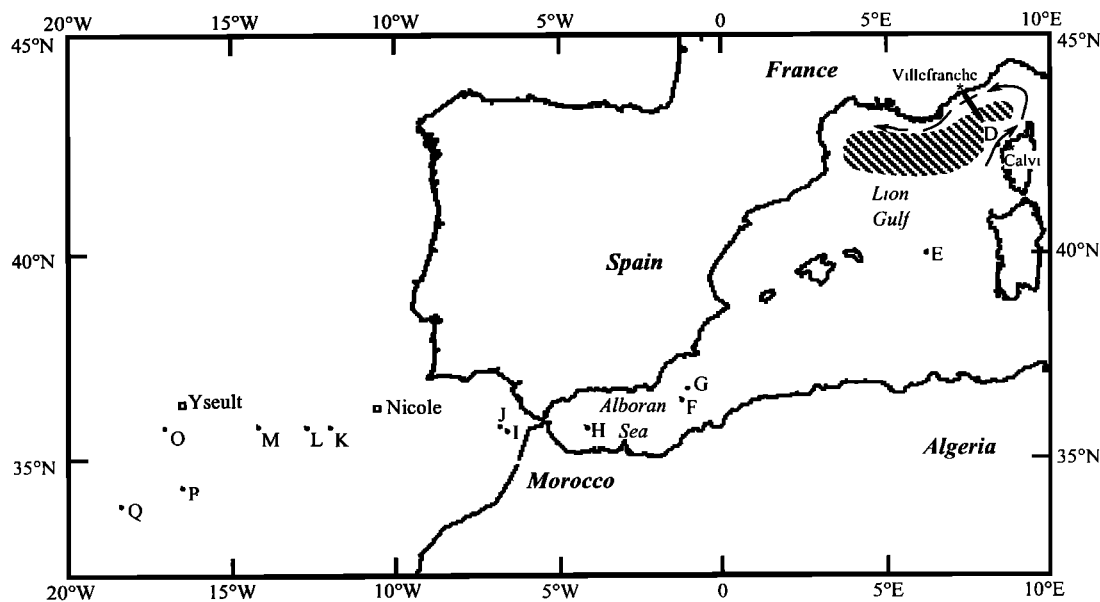
## Results and Discussion

### Mediterranean Basin

**Ligurian Sea.** Radionuclide concentrations are listed Table 1. The Western Deep Mediterranean Water is one of the

**Table 1.** Results

Stations	Depth, m	$^{226}\text{Ra}$ , 10 <sup>-2</sup> dpm L <sup>-1</sup>	$^{228}\text{Ra}$ , 10 <sup>-2</sup> dpm L <sup>-1</sup>	$^{228}\text{Ra}/^{226}\text{Ra}$	Salinity, psu	Temperature, °C	
<i>Dyfamed</i>							
Transect Bay	10	12.25 ± 3.75	4.00 ± 0.90	0.324 ± 0.122			
4 km	10	9.90 ± 0.40	2.68 ± 0.35	0.270 ± 0.035			
15 km	10	9.81 ± 0.33	2.61 ± 0.42	0.266 ± 0.044			
33 km	10	8.52 ± 0.32	1.79 ± 0.42	0.210 ± 0.050			
52 km	10	8.18 ± 0.18	0.91 ± 0.24	0.110 ± 0.029			
D	42°44N-8°31E	30	9.70 ± 0.47	1.42 ± 0.47	0.146 ± 0.052	38.767	18.815
		1000	13.15 ± 0.70	0.85 ± 0.25	0.064 ± 0.016	39.179	12.851
<i>Medatlante 1</i>							
F	36°41N-1°12W	10		0.174 ± 0.009	36.774	15.426	
		1000	12.87 ± 0.62	0.49 ± 0.04	0.038 ± 0.004	38.429	12.798
I	35°53N-6°35W	450	8.42 ± 0.30	0.67 ± 0.07	0.079 ± 0.009	35.977	12.489
J	35°52N-7°06W	200	7.32 ± 0.12	0.76 ± 0.05	0.103 ± 0.006	35.777	13.466
Nicole	36°27N-10°35W	500	6.62 ± 0.32	0.50 ± 0.07	0.076 ± 0.010	35.578	11.000
		600	7.00 ± 1.00	0.45 ± 0.08	0.064 ± 0.006	35.876	11.613
		800	9.28 ± 0.35	0.56 ± 0.09	0.062 ± 0.010	36.082	11.344
		1000	9.50 ± 0.50	0.77 ± 0.12	0.081 ± 0.004	36.448	12.126
		1200	9.65 ± 0.13	0.90 ± 0.09	0.093 ± 0.009	36.457	11.771
		1700	9.49 ± 2.60	0.19 ± 0.17	0.020 ± 0.018	35.678	7.249
<i>Medatlante 2</i>							
E	40°12N-6°08E	10	8.53 ± 0.21	1.79 ± 0.19	0.209 ± 0.022	37.331	25.064
		1000	13.13 ± 0.11	0.61 ± 0.07	0.046 ± 0.005	38.436	12.847
G	36°44N-1°10W	10	8.11 ± 0.24	1.55 ± 0.24	0.191 ± 0.030	36.555	21.459
H	36°00N-4°10W	10	8.16 ± 0.24	1.80 ± 0.22	0.221 ± 0.027	36.429	21.398
K	36°00N-12°00W	10	6.55 ± 0.10	1.24 ± 0.12	0.189 ± 0.018	36.440	23.238
L	36°00N-12°43W	10	6.97 ± 0.23	1.41 ± 0.23	0.202 ± 0.034	36.400	23.288
M	36°01N-14°12W	10	6.80 ± 0.24	1.05 ± 0.27	0.154 ± 0.040	36.431	23.116
W	35°00N-17°01W	10	5.84 ± 0.13	1.12 ± 0.16	0.192 ± 0.027	36.681	23.209
P	34°30N-16°30W	10	6.22 ± 0.11	0.91 ± 0.11	0.146 ± 0.018	36.427	23.450
Q	34°00N-18°25W	10	6.38 ± 0.44	1.10 ± 0.14	0.172 ± 0.025	36.576	24.194
Yseult	36°30'N-15°35'W	10	5.91 ± 0.13	0.88 ± 0.15	0.149 ± 0.028	36.430	23.780
		100	7.02 ± 0.14	1.05 ± 0.17	0.150 ± 0.026	36.088	15.094
		450	9.26 ± 0.15	0.41 ± 0.13	0.044 ± 0.014	35.623	11.389
		850	9.21 ± 0.19	0.53 ± 0.18	0.058 ± 0.019	35.923	10.459
		1100	8.50 ± 0.25	0.70 ± 0.10	0.083 ± 0.012	36.122	10.416
		1400	9.52 ± 0.30	0.28 ± 0.24	0.029 ± 0.025	35.602	7.281
		1800	8.46 ± 0.40	0.25 ± 0.29	0.030 ± 0.035	35.198	4.740
		2600	12.23 ± 0.30	0.22 ± 0.25	0.018 ± 0.020		
		2900	14.15 ± 0.23	0.33 ± 0.19	0.023 ± 0.013		
		3100	15.04 ± 0.20	0.42 ± 0.16	0.280 ± 0.010		

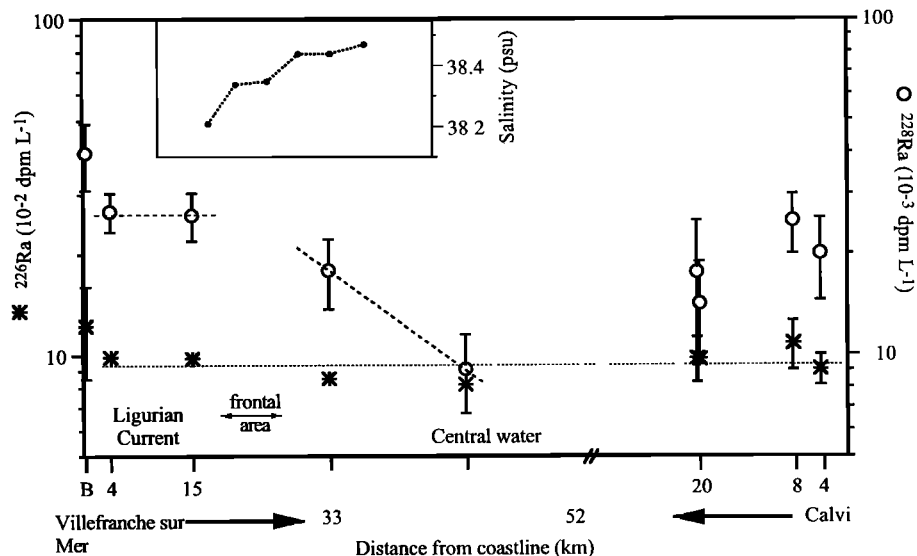


**Figure 1.** Location of samples collected in the NW Mediterranean Sea and in the NE Atlantic Ocean. All sample locations and data are listed in Table 1. The open symbols show position of Meddies *Nicole* and *Yseult* sampled during Medatlante cruises. The hatched area is the deep water formation area. The arrows show the location of the Ligurian Current in the studied area.

major constituents of the MOW. This Deep Water is formed in the northern area of the western basin (Gulf of Lion, Ligurian Sea). Deep water renewal occurs each year during winter and early spring when the cooling of superficial waters increases their density and induces vertical convection [Jacques and Treguer, 1986]. Therefore at first we studied the Deep Water formation area (Figure 1).

Figure 2 shows salinity data (L. Prieur and J. Raunet, personal communication, 1989) and radium activities in surface waters along a transect between Villefranche and Calvi in the Ligurian Sea. As a comparison, we presented radium activities in Villefranche Bay: both radium isotopes are at higher levels due to the proximity of the shallow sediments (<40 m). Along the transect, surface  $^{226}\text{Ra}$  activities are

relatively constant, varying from  $(8 \text{ to } 10) \times 10^{-2} \text{ dpm L}^{-1}$ . The half-life of  $^{226}\text{Ra}$ , 1600 years, is too long to allow an observable decay over short timescales. On the other hand,  $^{228}\text{Ra}$  activities decrease by a factor of 3 from the coast to the central site. The  $^{228}\text{Ra}$  distribution reflects the presence of two distinct surficial water masses, separated by a frontal area, as highlighted by salinity curve. The Ligurian Current, which flows between the coast and a distance of approximately 30 km offshore [Béthoux et al., 1988], is enriched in Ra by input from interstitial waters of coastal sediment. This input sustains high activities around  $2.5 \times 10^{-2} \text{ dpm L}^{-1}$  into the current body. More than 30 km offshore, the central water mass contains  $^{228}\text{Ra}$  activities typical of open ocean waters. At the central site of the transect, surface water is unaffected by input due to



**Figure 2.** Ra versus distance from coastline relationship for surface samples of Villefranche-Calvi transect in the Ligurian Sea. The salinity is expressed in psu.

the distance from sediments, either coastal or deep, the water column depth being around 2500 m.

Seawater  $^{228}\text{Ra}$  data may be used to calculate apparent horizontal eddy diffusion coefficients [Kaufman *et al.*, 1973]. The observed gradient in  $^{228}\text{Ra}$  concentrations in surface water between coastline and central sites probably reflects eddy diffusion of  $^{228}\text{Ra}$  perpendicular to the mean advective flow of the Ligurian Current. In the simplest case, the  $^{228}\text{Ra}$  distribution can be treated as a balance between eddy diffusion and radioactive decay of  $^{228}\text{Ra}$ . At steady state

$$K_y \frac{d^2c}{dz^2} - \lambda c = 0 \quad (1)$$

With boundary conditions  $c = c_0$  at  $z = 0$  and  $c = 0$  at  $z = \infty$ , the solution of (1) is [Knauss *et al.*, 1978]

$$c = c_0 e^{(-y/y^*)} \quad y^* = \sqrt{\frac{K_y}{\lambda}} \quad (2)$$

$$K_y = \left[ \frac{y}{\ln(c_0/c)} \right]^2 \times \lambda \quad (3)$$

$\lambda$  the decay constant of  $^{228}\text{Ra}$ ,  $y$  the distance between the two points,  $y^*$  the scale length and  $K_y$  the coefficient of horizontal eddy diffusion.

We calculate  $K_y$ , the apparent horizontal eddy diffusion coefficient, between 4 and 15 km offshore which may represent the diffusion of  $^{228}\text{Ra}$  into the Ligurian Current water body. Even though the extent of the Ligurian Current varies seasonally, with a maximum extension of about 20-25 km offshore, these two points are always in the central current body [Béthoux *et al.*, 1988]. Into the current body,  $^{228}\text{Ra}$  activities give a  $K_y$  of around  $7 \times 10^6 \text{ cm}^2 \text{ s}^{-1}$ , with a characteristic scale length of about 420 km (Figure 2). As the central water could be affected by deep water renewal during winter, such calculation is not realistic for the data obtained at 33 and 52 km offshore. Nevertheless, the decrease of  $^{228}\text{Ra}$  between the current body and the central water contents, beyond the influence of the Ligurian Current, indicates that Ligurian Current acts as a barrier against the propagation toward the central water of species diffusing from coastal shelf.

In the same Deep Water formation area, at 1000 m depth,  $^{228}\text{Ra}$  activity is relatively high with levels of  $0.85 \times 10^{-2} \text{ dpm L}^{-1}$ , may be in relation with the winter vertical convection (the superficial seawater which flows down during these events supplies  $^{228}\text{Ra}$ ). By comparison, Atlantic or Pacific  $^{228}\text{Ra}$  concentrations at the same depth are usually much lower, less than  $0.5 \times 10^{-2} \text{ dpm L}^{-1}$  [Moore *et al.*, 1985].

**Western Deep Water circulation in the Western Mediterranean basin.** The western Deep Water (DW), which is formed by deep convection in the north area of the Western Mediterranean basin, flows toward the Straits of Gibraltar, below 600 m depth with nearly constant hydrological characteristics:  $T = 12.74^\circ\text{C}$  and  $S = 38.44 \text{ psu}$  [Gascard and Richez, 1985]. The western DW was studied from its formation area up to its outflow through the Gibraltar Straits. This water mass, identified by its temperature and salinity signature, was sampled at 1000 m (Figure 1) : (1) south of the formation area (E), and (2) at the east side of the Alboran Sea (F). Radium-226 activities are relatively constant with values of about  $13 \times 10^{-2} \text{ dpm L}^{-1}$ , which is not surprising, taking into account the short residence time of water in this basin [Andrié and Merlivat, 1988].

For  $^{228}\text{Ra}$ , on the other hand, we observe a continuous decrease in activity outside the formation area as Deep Water

propagates to the west side of the basin. These variations reflect the aging of the Deep Water during its motion from the NW basin to the Alboran Sea. More particularly, the comparison between sites E and F indicates a significant decrease of  $^{228}\text{Ra}$  from  $(0.61 \text{ to } 0.49) \times 10^{-2} \text{ dpm L}^{-1}$  at 1000 m. As the western Deep Water propagates westward without significant dilution, the decrease in  $^{228}\text{Ra}$  activity can be ascribed to the decay of the isotope and can be used to calculate a transit time of the water mass between the two sites. The hypothesis is that deep advection is horizontal and without mixing. Moreover the depth of 1000 m in a 2500-m column allows us to neglect diffusive input of Ra from sediment. The equation is derived from the decay law

$$t = \lambda^{-1} \ln \frac{A_0}{A} \quad (4)$$

$A_0$  and  $A$  the activities of Ra at both sites,  $t$  the time elapsed which corresponds to the observed decay of  $^{228}\text{Ra}$ , and  $\lambda$  the decay constant of  $^{228}\text{Ra}$ .

With the values  $A_0 = (0.61 \pm 0.07) \times 10^{-2} \text{ dpm L}^{-1}$ ,  $A = (0.49 \pm 0.04) \times 10^{-2} \text{ dpm L}^{-1}$ ,  $\lambda [^{228}\text{Ra}] = 0.1205 \text{ y}^{-1}$ , we obtained a mean transit time of the Western Deep Water from the Southern Lion Gulf (E) to the Alboran Sea (F) of 1.8 years, with range from 0.6 to 2.9 years. Taking into account the shortest distance between the two sites, considered as a direct transect, it is then possible to calculate a mean translation velocity of the deep water mass of about  $1.5 \text{ cm s}^{-1}$  (0.9-4.5). Andrié and Merlivat [1988] established the residence time of the water in the Western Mediterranean basin with transient tracers. These authors calculate a transit time of the deep water, with the same hypothesis (deep horizontal advection, no exchange), of 6.5 years between the Sardinia Straits and Alboran Sea: such time implies a translation velocity of  $0.5 \text{ cm s}^{-1}$ . Despite the fact that these two studies concern two distinct transects and are based on different tracers ( $^{228}\text{Ra}$ , 5.75 years;  $^3\text{He}$ , 12.4 years), the estimated transit times are not drastically different. Both are in agreement with the assumption of a rapid turnover of the Mediterranean water mass already inferred from the mean of salt and heat budgets.

#### Northeastern Atlantic Ocean

**Surface activity.** Off the Gibraltar Straits, Atlantic surface water contains relatively constant values of Ra. Ratios of  $^{228}\text{Ra}/^{226}\text{Ra}$  previously determined in the same area show a discrimination between coastal and open ocean water masses [Van Geen *et al.*, 1991]. Surface Atlantic  $^{228}\text{Ra}/^{226}\text{Ra}$  activities obtained during Medatlante cruises are plotted versus salinity on Figure 3. The range of activity ratios is comparable with that measured in earlier work in the Gulf of Cadiz [Van Geen *et al.*, 1991]. These authors have defined three zones: (1) Spanish shelf water with activity ratios up to 0.57, (2) more saline Moroccan shelf water with ratios not exceeding 0.26, and (3) surface Atlantic water within the Strait with ratios less than 0.2. Our surface ratio values are in good agreement with the third group even for distant points.

**Excess of  $^{228}\text{Ra}$  in Meddies.** After investigation of the Mediterranean Deep Water, we have focused on two Meddies surveyed during Medatlante cruises. The first was localized at  $36^\circ 27' \text{N}$ ,  $10^\circ 35' \text{W}$ , characterized by a warm, salt, surface mixed layer (Figure 4). Below 100 m depth, temperature and salinity decrease rapidly to reach minimum values ( $T=11^\circ\text{C}$ ,  $S=35.578 \text{ psu}$ ) at a depth of 500 m. Below 500 m, salinity increases to a maximum at 1200 m (36.457 psu) and indicates that a Meddy called *Nicole* lies between 700 and 1500 m

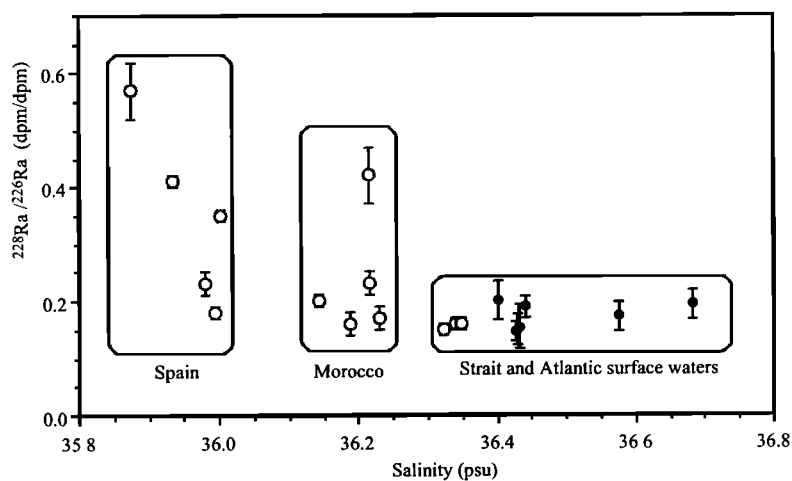


Figure 3. Ratio of  $^{228}\text{Ra}/^{226}\text{Ra}$  versus salinity relationship for surface samples. Open circles indicate values published by Van Geen *et al.* [1991].

depth. *Nicole* appears to be a large lens about 100 km in diameter. At its base, T and S values are characteristic of North Atlantic Deep Water. This hydrologic structure is reflected in the distribution of  $^{228}\text{Ra}$  between 500 and 1700 m depth. Because of its supply by diffusion from sediments,  $^{228}\text{Ra}$  activities usually display strong vertical gradients [Moore, 1969]. Moreover,  $^{228}\text{Ra}$  activities are often lower than  $5 \times 10^{-3}$  dpm  $\text{L}^{-1}$  in deep and intermediate waters [Moore *et al.*, 1985; Trier *et al.*, 1972]. Indeed, under the thermocline,  $^{228}\text{Ra}$  decreases rapidly to reach activities less than  $5 \times 10^{-3}$  dpm  $\text{L}^{-1}$  at 500 and 600 m. At greater depths,  $^{228}\text{Ra}$  increases up to  $9.0 \times 10^{-3}$  dpm  $\text{L}^{-1}$  at 1200 m (Figure 4). These high concentrations of  $^{228}\text{Ra}$  measured in Meddy water are in contrast to the low levels in the surrounding Atlantic water.

Such high levels indicate the presence of a water mass which has been recently in contact with sediments.

Meddies mostly constitute MOW which flows out into the Atlantic. At the Gibraltar Straits, MOW flows out of the Mediterranean basin at depth because of its greater density in comparison with Atlantic inflow. Around 150 m depth, the shear between the two layers flowing in opposite directions leads to eddy mixing. The effect is to reduce the salinity of the MOW [Minas *et al.*, 1991]. This dilution would also influence the Ra concentration. Sediment-water exchange is another process that could affect the Ra concentration of the MOW. During the crossing of the Straits, MOW is in contact with the sediments which presumably releases Ra and causes the Ra content of MOW to rise. Evaluating Ra input to the

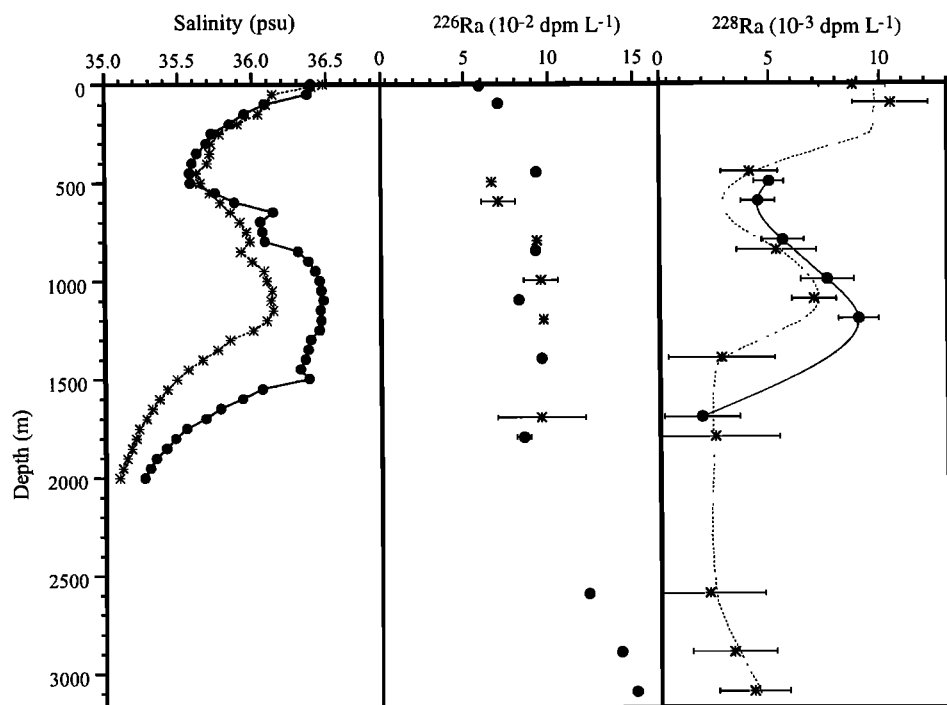


Figure 4. Vertical profiles of salinity (psu),  $^{228}\text{Ra}$  and  $^{226}\text{Ra}$  ( $10^{-3}$  and  $10^{-2}$  dpm  $\text{L}^{-1}$ ) at the site *Nicole* ( $36^{\circ}30'\text{N}$  -  $15^{\circ}35'\text{W}$ , open circles, water column depth 4750 m) and *Yseult* ( $36^{\circ}27'\text{N}$  -  $10^{\circ}35'\text{W}$ , crosses, water column depth 3170 m).

MOW in the vicinity of the Straits is complicated, because the exchange of water itself through the Straits of Gibraltar is a complex process which involves mixing of various water masses (Surface Atlantic water, Levantine Intermediate water and western Deep Water [*Gascard and Richez, 1985*]). The comparison of Ra data ( $36^{\circ}41'N-1^{\circ}12'W$  (F) and  $35^{\circ}53'N-6^{\circ}35'N$  (I) stations) on the Mediterranean and Atlantic sides of the Straits clearly indicates an increase in activities of both Ra isotopes. If we correct the Atlantic water contribution for *Nicole*, the excess  $^{228}\text{Ra}$  activity at 1200 m is  $35\pm 5\%$  in comparison to Mediterranean deep water. There is therefore strong evidence that input of Ra occurs when MOW is in contact with the sediment during the crossing of the Straits and continues as long as this water stays in contact with shallow sediment of the Spanish continental shelves.

The second Meddy was sampled at  $36^{\circ}30'N$ ,  $15^{\circ}35'W$  in summer 1989. In the upper layer, this site is similar to the *Nicole* site and exhibits a surface mixed layer of high temperature and salinity (Figure 4). Below the mixed layer, temperature and salinity decrease rapidly. Down to 500 m depth, they both increase to reach maximum values at 1100 m which corresponds to the center of the lens. This Meddy, called *Yseult*, is about half the size and is characterized by weaker salinity and temperature maxima than *Nicole*. Below 1500 m, T and S are typical of the deep Atlantic. As for *Nicole*, the  $^{228}\text{Ra}$  concentration maximum is at the surface. Radium-228 diminishes below the thermocline, reaches high levels,  $(0.7\pm 0.1)\times 10^{-2}$  dpm  $L^{-1}$  at 1100 m, at the *Yseult* immersion. As for the T and S signature, the maximum  $^{228}\text{Ra}$  activity is lower than at *Nicole*. Two processes could explain this reduction. One is dilution by surrounding Atlantic waters. According to *Armi et al. [1989]*, dilution limits the mean life to 2 or 3 years. In fact, as previously mentioned, surrounding Atlantic water at such depths contains lower levels of  $^{228}\text{Ra}$  than the MOW, as a result of the absence of recent input. The second explanation is a longer advection time from the shelf source and therefore a decay of  $^{228}\text{Ra}$ . Below 2500 m, the gradual increase of  $^{228}\text{Ra}$  could be ascribed to diffusion from sediment which lies at 3170 m.

Unlike  $^{228}\text{Ra}$ , the two profiles activities of  $^{226}\text{Ra}$ , with its long half-life, are only slightly affected by the presence of Mediterranean water. The supply by diffusion from the sediments is negligible in comparison to ambient  $^{226}\text{Ra}$  levels. Surface activities of  $^{226}\text{Ra}$  off Gibraltar are in the range of  $(5.9 \text{ to } 7.0)\times 10^{-2}$  dpm  $L^{-1}$ , slightly lower than those reported for West Atlantic upper waters [*Moore, 1987*]. This may reflect lower input from coastal shelf and reduced area of continental shelves on the eastern compared with the western side of the Atlantic.

**Translation velocity of Meddy *Yseult*.** The decrease of  $^{228}\text{Ra}$  can be used to calculate the advection velocity of the MOW as was done for western Mediterranean Deep Water. The hypothesis is that we can use *Nicole* as a reference. *Nicole* has been localized in the vicinity of the Meddies formation area, and we can assume that Ra activities are relatively constant in the MOW. The calculation takes into account the two factors which explain the decrease of  $^{228}\text{Ra}$  between Meddies *Nicole* and *Yseult*: the decay of  $^{228}\text{Ra}$  and the dilution of MOW by Atlantic water. The effect of Atlantic water on  $^{228}\text{Ra}$  activity can be corrected by calculating the dilution using temperature and salinity data. The calculated dilution of the Mediterranean water at 1100 m depth in comparison with the surrounding Atlantic water is 38%.

When this dilution factor is applied to the  $^{226}\text{Ra}$  activity, the calculated  $^{226}\text{Ra}$  is  $(8.50\pm 0.15)\times 10^{-2}$  dpm  $L^{-1}$ ; this value is in agreement with the measured value of  $(8.20\pm 0.20)\times 10^{-2}$  dpm  $L^{-1}$  (Table 1). For  $^{228}\text{Ra}$ , the value calculated in the same manner is  $(0.75\pm 0.07)\times 10^{-2}$  dpm  $L^{-1}$  which can be compared to the measured activity of  $(0.70\pm 0.10)\times 10^{-2}$  dpm  $L^{-1}$ . Due to the large errors on these values, the potential decay of  $^{228}\text{Ra}$  does not clearly appear. However, it indicates that *Yseult* is probably young, less than 1.9 years. With additional sampling, age calculation could be better constrained.

The studied area includes many submarine obstacles such as Ormond, Gettysburg, and Josephine seamounts and the Madeira archipelago. The possible Meddy pathways around these seamounts covers a minimum distance of 350 miles between the two sites studied. Assuming a mean translation velocity of  $3 \text{ cm s}^{-1}$  for Meddies [*Armi et al., 1989*], we can calculate a minimum transit time of 8.2 months. These two estimates seem to indicate that the transit time is probably less than 1 year, in agreement with the estimation based on biological tracers [*Savenkoff et al., 1993*].

## Conclusions

This study demonstrates the usefulness of Ra isotopes as internal tracers for following the MOW from the Mediterranean deep water formation area to the NE Atlantic Ocean. In the Western Mediterranean Sea, the observed gradient in  $^{228}\text{Ra}$  activities in Ligurian Sea surface seawater may be attributed to the transport of  $^{228}\text{Ra}$  away from the coast by eddy diffusion perpendicular to the Ligurian Current. Simpler one-dimensional, steady state models do not produce realistic estimates of horizontal mixing. More extensive sampling programs are needed to realize the full potential of this tracer for oceanic mixing rates. In its formation area, the  $^{228}\text{Ra}$  activity of the Deep Mediterranean Water is as high as open ocean surface waters values. This may be explained by the annual renewal of  $^{228}\text{Ra}$  during winter convection. As the Deep Water flows westward, the observed decay of  $^{228}\text{Ra}$  with distance from the source allows us to calculate a mean translation velocity of the deep water mass of the order of  $1.5 \text{ cm s}^{-1}$  which is consistent with a rapid renewal time of the western Mediterranean basin.

In the Atlantic Ocean, the presence of Meddies *Nicole* and *Yseult* is strikingly reflected by  $^{228}\text{Ra}$  profiles in the water column. Indeed, the Meddies contain activities of  $^{228}\text{Ra}$  which are significantly different from surrounding waters. This feature reflects the presence of the MOW which has recently been in contact with sediment in the vicinity of the Gibraltar Straits. After correction of the dilution of *Yseult* water by the surrounding Atlantic water using salinity and  $^{226}\text{Ra}$  data, it is possible to calculate a lower limit of the velocity of *Yseult* of around  $1 \text{ cm s}^{-1}$ . This result, along with its biological signature [*Savenkoff et al., 1993*] seems to indicate that *Yseult* has an age of less than 1 year. With additional sampling, better constrained ages could be obtained. The use of such internal tracers is perhaps the only way to constrain the age of these features without following them for several years. This work has been possible with the improvement of a low-background, high-efficiency detector which has been placed in the LSM and allows us to measure  $^{228}\text{Ra}$  on small volume samples with reduced errors and laboratory work.

**Acknowledgements.** This is a contribution to the France-JGOFS program financed by the CNRS (Dyamed, Medatlante). During the Medatlante cruises, vertical conductivity-temperature-depth profiles were established by C. Bournot, J. Raunet, and D. Tailleux. We thank the logistic staff of the LSM for its help in working up the counting installation. The help of G. Copin-Montégut was useful for retrieving salinity data of the Ligurian surface water. We also thank M. Bender and P. Newton for useful comments. This is CFR contribution 1684.

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(Received April 1, 1993; revised August 15, 1994; accepted February 13, 1995.)



