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Fate and Distribution of Linear Alkylbenzene Sulfonates in the Littoral Environment

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A study was conducted of the fate and behavior of LAS in a shallow coastal zone adjoining the Bay of Cádiz (southwest of Spain). The area is subjected to strong tidal currents and receives directly the discharge of untreated wastewaters of a population of about 100 000 inhabitants. LAS showed a nonconservative behavior due to intense biodegradation and due to sorption and settling of material in suspension. The sorption to suspended matter affects the way in which its dispersion takes place in zones with high turbidity, since this fraction may represent up to 60% of the total quantity present in the medium. The surfactant character of LAS is also illustrated by a strong sorption at the water−atmosphere interface. In zones close to urban effluent discharge points, this translates into a steep vertical gradient in the LAS concentration, with values found in the top 3−5 mm of water depth some 2 orders of magnitude greater than those found at a depth of 0.5 m. Over a large part of the area studied, the LAS concentration in the sediment and in the water column can be described by means of the Freundlich's equation.

Introduction

Since the utilization of linear alkylbenzene sulfonates (LAS) as an anionic surfactant in the manufacture of domestic detergents began in the 1960s, emission to surface and coastal waters has been acknowledged. This has stimulated many recent studies in which the concentration of LAS in seawaters and sediments has been measured, with the objectives of establishing its environmental behavior (*1*−*8*). Most of them refer to the transport processes of LAS in estuaries and its subsequent incorporation into the coastal waters. However the direct discharge of untreated effluent containing LAS to seawater has not been studied. It is clear why there is such interest in understanding the environmental behavior of this surfactant. The rate of LAS utilization is very high (e.g., 4.0 g per inhabitant per day in Spain and up to 11.0 g in Japan), and like all surfactants LAS can interact with biological membranes.

In the same way as happens with many other organic substances, the behavior of LAS depends largely on its molecular properties and its time of residence in the medium into which it is discharged. This latter factor is, in turn, conditioned by its intrinsic biodegradability and by its fate and distribution among the various environmental compartments. It is now known that the rate of degradation in the sea is slower than that in continental waters (*4*, *9*) and that this rate depends to a large extent on the physicochemical characteristics of the medium, especially the redox potential, temperature, and salinity (*10*−*12*). Recently, Rubio et al. (*13*) have shown that LAS has a strong tendency to be sorbed by marine sediment in a way similar to the process occurring in continental systems (*14*), and that the sorption increases in intensity with an increase in the salinity of the water. Laboratory studies have also shown that this sorption is practically irreversible (*13*); therefore, the sediments act as a sink for the LAS that reaches the marine medium.

Both the sorption capacity and the rate of biodegradation increase, as the length of the alkyl chain of the particular LAS homologues increases (*7*, *9*). These processes affect the net reactivity of LAS in the environment. As the residence time increases, a relative enrichment of short-chain homologues occurs in the water column.

The objective of this study is two-fold: first, to evaluate the way by which the characteristics/physicochemical properties of this surfactant substance determine which transport mechanism are involved in its dispersion in littoral zones and second, to establish the role played by the sediments in the removal of LAS from the water column.

The research has been carried out in the Bay of Cádiz, a temperate coastal ecosystem whose population consumes annually more than 1000 ton of LAS. The site selected for study is a shallow narrow channel subjected to strong tidal currents, into which untreated urban wastewater is discharged. This last factor is of notable interest, since in these circumstances the LAS reaching the environment shows a composition of homologues that is very close to that of the original commercial product.

Experimental Section

Chemicals. The liquids used as eluents in the chromatographic measurements were water and methanol, both of chromatography quality, purchased from Merck (Darmstadt, Germany). Sodium perchlorate was also purchased from Merck. The C_{18} reversed-phase and SAX minicolumns were purchased from Supelco. The HPLC separation was performed with a C_8 column of 25 cm length, 0.46 cm ID, and 10 μm particle size purchased from Teknokroma. The LAS standards, of composition similar to the commercial product used in Spain and with a low dialkyltetralinsulfonates (DATS) content (<0.5%), were supplied by Petroquimica Española SA. The proportional composition of the different homologues is as follows: C_{10}, 3.9%; C_{11}, 37.4%; C_{12}, 35.4%; C_{13}: 23.1%; C_{14}, 0.2%.

Study Area and Sampling. The zone selected for study was the Channel of Sancti Petri, a branch of the sea joining the southern part of the Bay of Cádiz with the Atlantic Ocean, which provides seawater to an area of salt-ponds of about 6000 Ha (Figure 1). It is a channel of 18 km length and of very reduced depth—between 3 and 6 m. It is subjected to a semidiurnal tidal regime, with an average amplitude of 2.56 m. The tidal wave enters from both ends of the channel, with a delay of 0.5 h at the north; the two waves meet at a

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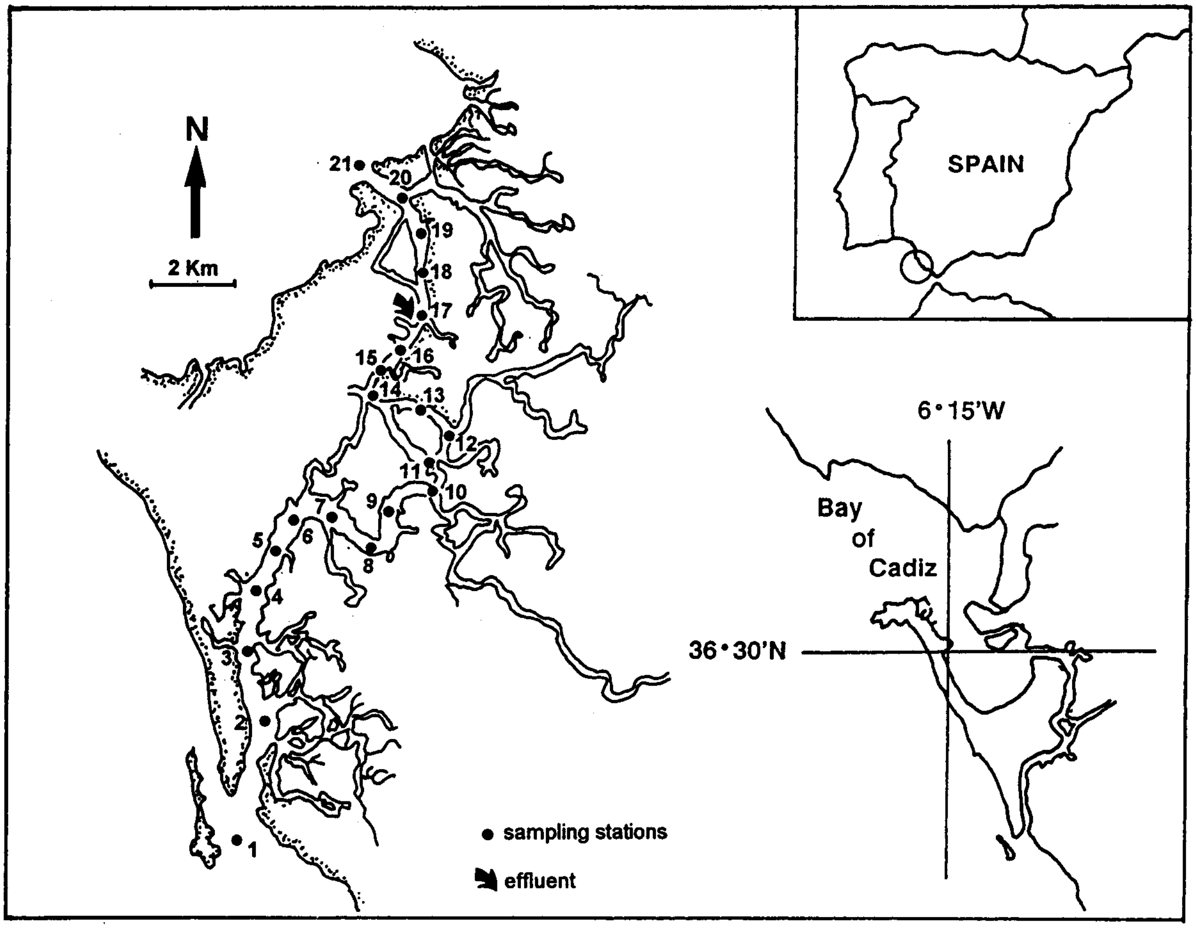

FIGURE 1. Schematic of the Bay of Cádiz showing positions of sampling stations.

TABLE 1. Main Physicochemical Characteristics of the Medium during the Samplings Made

sampling date	tidal conditions	tide amplitude (m)	temperature (°C)	salinity	dissolved oxygen (μM)	pH (NBS)	suspended solids (mg/L)
1991							
Oct 4	high tide	2.33	20.9 ± 0.7	36.35 ± 0.25	214.3 ± 29.0	8.01 ± 0.09	103 ± 13
Oct 23	flowing	3.02	17.6 ± 0.4	35.17 ± 0.42	195.1 ± 41.9	7.95 ± 0.07	122 ± 19
Oct 23	high tide	3.05	14.3 ± 0.8	35.68 ± 2.03	193.6 ± 57.1	7.91 ± 0.18	105 ± 14
Nov 29	ebbing	1.50	14.2 ± 0.4	36.27 ± 0.35	177.6 ± 57.0	7.89 ± 0.07	107 ± 21
Nov 29	low tide	1.70	14.4 ± 0.3	36.37 ± 0.39	167.7 ± 48.8	7.91 ± 0.08	116 ± 13
1992							
Feb 11	ebbing	1.42	13.1 ± 0.5	36.35 ± 0.39	172.4 ± 46.7	7.82 ± 0.10	89 ± 24
Apr 22	ebbing	1.76	17.9 ± 0.3	37.57 ± 0.42	188.7 ± 49.5	8.13 ± 0.08	139 ± 65
Apr 22	low tide	1.76	18.5 ± 0.4	37.89 ± 0.54	171.0 ± 39.3	8.14 ± 0.08	143 ± 33
Jul 8	high tide	1.65	23.1 ± 1.3	37.23 ± 0.70	159.1 ± 48.5	7.82 ± 0.09	157 ± 34
Jul 8	ebbing	1.68	24.0 ± 1.0	37.52 ± 0.96	126.4 ± 54.7	7.77 ± 0.14	134 ± 26
Sep 9	low tide	2.31	23.9 ± 0.3	39.54 ± 1.29	118.5 ± 43.8	7.81 ± 0.11	140 ± 24
Sep 9	flowing	2.20	24.1 ± 0.6	38.63 ± 1.43	121.3 ± 47.3	7.79 ± 0.10	154 ± 22

point 5 km from the Bay. It is at exactly this point that the urban effluents of the town of San Fernando, of 100 000 inhabitants, is discharged untreated.

Sampling stations, 21 in number, were established at the locations shown in Figure 1. Samples were taken on 13 occasions throughout 1991 and 1992, at varying tidal conditions. Two boats were used simultaneously for the sampling, and in all cases, less than 45 min was taken to complete the sampling. At all the stations the water temperature, the salinity (*15*), the concentration of dissolved oxygen (*16*), the pH (NBS scale), and the suspended solids (gravimetrically) were measured. Table 1 gives the main characteristics of the medium during the samplings made.

The samples of water were taken with Ruttner bottles (Hydro-Bios, Kiel) previously acid-washed and then were transferred to polyethylene bottles; a quantity of formaldehyde sufficient to obtain an HCHO concentration of 0.3M (≃ 1% v/v) was added. On some occasions, samples of surface water (between 0 and 3−5 mm depth) were taken using a sampler (Hydro-Bios, Kiel). All samples were kept in the dark at 4 °C until their analysis.

The sediment samples (0−10 cm) were taken with a Van Veen drag and then transferred to the laboratory at 4 °C. Later they were dried at 80 °C in a heater with air circulation and then milled and strained through a 160 μm sieve. To obtain the solids in suspension, samples of water of 50−60

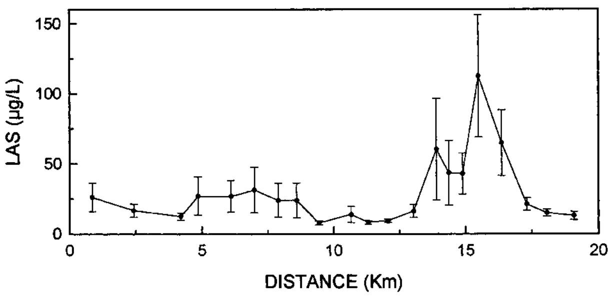

FIGURE 2. Spatial distribution of the average LAS concentration in water, for all the samplings made. The origin in the distances is starting (distance equal to 0) from the south of the Sancti Petri Channel.

L, previously fixed with formaldehyde, were centrifuged continuously at 20 000 rpm with a flow rate of 60 mL/min. The wet samples obtained were then processed in the same way as the sediment.

Pretreatment of the Samples. The samples of water (250 mL) were filtered (AP 40 Millipore), acidified at a pH of 3, and purified and concentrated by solid-phase extraction, using consecutively minicolumns of the hydrophobe type (C_{18}) and strong anionic exchange (SAX) type, following the previously described procedure (*17*).

For the analysis of the sediments, quantities of approximately 15 g were placed in Soxhlet extraction cartridges and the surfactant was extracted over 16 h with methanol. Next, the methanolic extract was evaporated until dry in a rotavapor, and the dry residue was redissolved with 200 mL of hot water in an ultrasonic bath. The samples of solids in suspension, about 1 g in weight, were processed in the same way as the sediment, using a microscale Soxhlet extraction apparatus. The solutions obtained from this treatment, both of the sediments and of the solids in suspension, were processed in an identical way to the samples of water. Recovery of the LAS in the solid-phase extraction stage was estimated at 95.4 ± 1.4%, and the reproducibility shows a relative standard deviation of between 2 and 3% for the range 10−1000 μg L^{-1}.

LAS Analysis. The separation and quantification of the various homologues of LAS were carried out with a reverse-phase liquid chromatograph with a fluorescence detector (Waters 470). The mobile phase was methanol/water (80:20 v/v), to which $NaClO_4$ was added at a concentration of 10 g L^{-1}. As the stationary phase, a Lichrosorb RP-8 column with a particle size of 10 μm was used. The separation (isocratic regime) and quantitation were done in accordance with previously described procedures (*18*−*20*).

Results and Discussion

Dispersion of LAS in the Water Column. The average values of the LAS concentration obtained in all the samplings made are shown in Figure 2. No well-established seasonal variation was observed, therefore the high values of the standard deviation found at all the stations must be attributed mainly to the variation in the flow volume with the tides and to the variations in the domestic utilization of LAS depending on the time of day and on the day of the week (*21*).

The highest values are recorded in the part of the channel nearest to the urban effluent discharge point, where the LAS concentration can occasionally exceed 1500 μg L^{-1} with the tide ebbing at 0.5 km to the discharge point. There is an appreciable reduction in concentration toward both ends of the channel, owing to biodegradation and dilution produced by the tidal inflows; the values obtained here (10−30 μg L^{-1}) are similar to those obtained by various authors in low contaminated places, such as the outer zone of the Bay of Tokyo (*22, 23*). Occasionally (high tide with a high coefficient), little urban sewage discharges are produced in the channel area located between 5 and 8.5 km. This fact can explain that the LAS concentrations measured in this area are much higher than those measured in the adjacents areas.

In the area near the effluent discharge point, the surfactant properties of LAS are demonstrated in the manner by which the dispersion of LAS takes place. The data shown in Figure 3A demonstrate the existence of a strong sorption at the water−atmosphere interface. In this place (station 17), no similar vertical gradient was found for salinity, therefore it can be discounted that the vertical distribution found for the LAS might be produced by the surface spreading of the effluent plume as a result of its lower density. At more distant stations, at which the flow regime is more turbulent (Figure 3B), the vertical gradient in the LAS concentration is not detectable. At station 10 (Figure 3B), an inversion can even be observed in the distribution found near the effluent discharge of untreated wastewater, which could be imputed to the existence in this area of a certain stratification in the water column caused by a continuous supply/outflow of freshwater with a lower LAS content. It is in agreement with the vertical profiles of salinity measured in the same site and at the same time (data not showed).

The behavior of the LAS in the zone studied is clearly nonconservative. This result had been reported by Takada y Ogura (*1*) in a previous study perfomed in an estuarine area where the salinity variations were high. Figure 4 illustrates that a significant loss of the surfactant occurs in the dissolved phase, particularly at the salinity values found near the effluent discharge of untreated wastewater. Three causes or mechanisms may be postulated to explain the observed behavior as previously reported by several authors: (i) biodegradation in the dissolved phase; (ii) sorption by the particulate material (solids in suspension and sediments); and (iii) precipitation, presumably in the form of calcium salts. This third process ocurrs on the sediments closest to the point of discharge of the urban effluents, although it does not fully explain the nonconservative behavior of the LAS. In effect, if the sampling stations closest to the effluent discharge are excluded, the lack of linearity between the LAS concentration and the conservative index of the mixture used is maintained, albeit less accentuated.

The existence of an intense biodegradation in waters of the Bay of Cádiz has been demonstrated in a previous study (*20*), in which high concentrations of long-chain ($11 > n > 7$) sulfophenylcarboxylic acids (SPC) were discovered; the relative values of these were consistent with the mechanisms proposed by Schöberl (*24*), for the first stages of the degradation of LAS.

The nonconservative behavior of LAS is also determined by its sorption by the particulate matter in suspension. Table 2 gives the results obtained for the LAS concentration in the solids in suspension in a sampling carried out at low tide with a low tidal coefficient, where there is little appreciable variation in salinity between the various stations. It can be observed that the LAS concentration in the particulate matter is high. The content of the water here in suspended solids is very high, and this means that the quantity of LAS transported in the undissolved form must be high. This suggests that the generalized practice of filtering the samples before their pretreatment to concentrate the total LAS should be avoided/discontinued, since the result of such filtering would be to commit serious errors by default.

The value of the partition coefficient between the particulate matter and the water is high due to the high sorption capacity of LAS. In Table 2, it can be observed that the value of the partition coefficient increases in proportion to the time of residence of the LAS in the medium. This fact can be interpreted to mean that the sorption of LAS has a slow kinetic, as has been shown in laboratory trials of sorption of

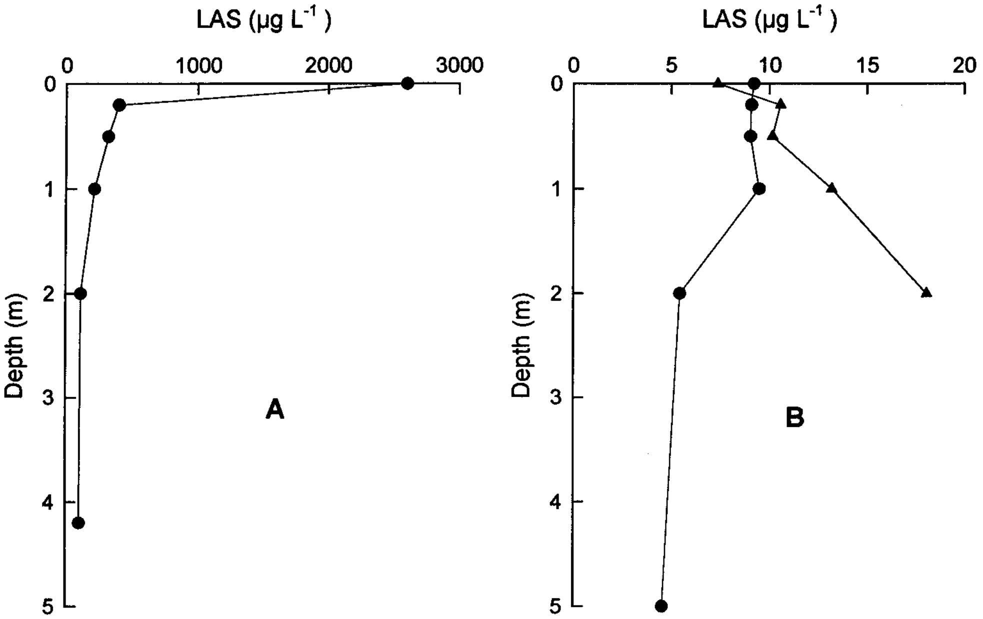

FIGURE 3. Vertical profiles of dissolved LAS concentration measured in water layer: (A) station 17; (B) station 20 (circles), and station 10 (triangles).

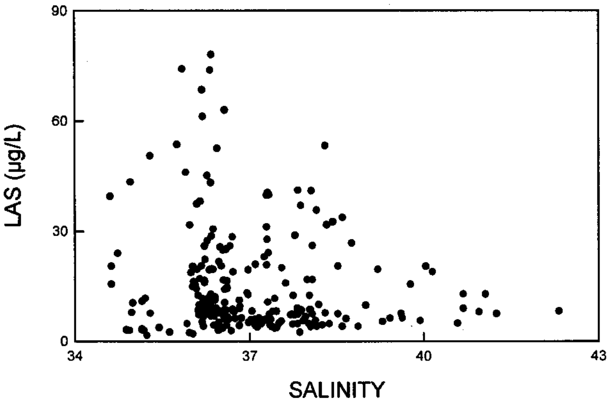

FIGURE 4. Representation of the concentration of dissolved LAS against salinity, for all the samplings made.

sediment (*25*). As a consequence of this fact, the percentage of LAS associated with the particulate matter increases in line with the distance of the sampling stations from the point of effluent discharge, reaching 60% at the farthest stations. Also, the magnitude of sorption is closely correlated with the ionic strength of the medium, which agrees with the results obtained in sorption tests with marine sediments (*13*).

The extent to which LAS is adsorbed by the particulate matter, as well as the rate of biodegradation, maintains a relationship with the length of the alkylic chain of the LAS. From this it can be deduced that the proportions of the homologues of the LAS that remain dissolved in the water should vary in a regular way with the time of residence of the surfactant in the medium. Figure 5 shows the distribution of homologues in the top layer of the water for those three stations at which the vertical gradient of concentration was determined (Figure 3). It can be observed that, in general terms, as the length of the residence time of the LAS in the water column increases, there is an increase in the proportions of the shorter-chain homologues (C_{10} and C_{11}) and a decrease in the proportions of the longer-chain homologues.

Agreement between the theoretical predictions and the experimental data is relevant given that the data is from fieldwork—when the vertical evolution of the various homologues in the area close to the urban effluent discharge point is analyzed. From Table 3, it can be observed that the surface sorption not only is very intense (Figure 3A) but also takes place preferentially on the homologues of longer alkylic chain. The results shown in Table 3 probably correspond to the combined effect of a series of circumstances that coincided in this sampling, particularly the absence of wind and a low tidal coefficient, which produced a regime of nonturbulent mixing between the wastewaters and those of the seawater tidal flow. Nevertheless, this fact illustrates the important part played by the molecular properties in the environmental behavior of organic substances; this should be borne in mind when their dispersion is studied, at least in the littoral environment.

Furthermore, the appreciable difference found between the distribution of homologues near the urban effluent discharge point and the commercial form of LAS (Figure 5) confirms the idea proposed by Moreno and Ferrer (*26*) that the LAS undergoes an intense process of degradation during its passage through the drainage system before being discharged into the environment.

Accumulation of LAS in the Sediment. The distribution of LAS found in the top layer of the sediments is shown in Figure 6. It can be observed that the values found near the effluent discharge point are very high due to the direct discharge of LAS, higher even than those reported for highly contaminated parts of the Lagoon of Venice (*27*) or of the Bay of Tokyo (*1*). The LAS concentration decreases sharply, the farther the sampling station is from the effluent discharge point, with values of 1.5−2.0 ppm found at the two entrances to the channel.

In general terms, the evolution of the LAS concentration in the sediments is similar to the average found for the water, and both values show a close relationship over a large part of the channel. Figure 7 shows this relationship for the stations in the northern part of the channel, which is the place with the highest concentration levels. Both variables, the concentration in the sediment (C_s) and the concentration in the water (C_w), show a good fit ($r = 0.92$) to following the equation:

$$C_s = K(C_w)^{1/n}$$

which corresponds to the expression of Freundlich's isotherm. This is the model that has been satisfactorily used

TABLE 2. Concentrations of LAS in Water and in Solids in Suspension[a]

station	DDO[b] (km)	salinity	dissolved LAS (μg/L)	particulate LAS (μg/L)	coefficient of partition	% LAS in suspended solids[c]
1	13.36	36.018	5.3	72.0	13.58×10^3	59.5
3	10.79	35.916	7.2	86.3	11.99×10^3	55.9
5	8.89	35.436	5.7	60.5	10.61×10^3	54.0
10	4.33	34.875	9.4	50.9	5.41×10^3	41.4
14	1.09	34.624	2.7	14.4	5.33×10^3	39.8
17	0.50	34.402	1687.2	5941.0	3.52×10^3	30.1
18	1.37	34.992	12.8	99.7	7.79×10^3	46.5

[a] The coefficient of partition tabulated corresponds to the quotient between the LAS concentrations in the particulate phase and in the dissolved phase, expressed in the same units. [b] The variable DDO is the distance in km of each sampling station from the point of discharge of the untreated wastewaters from the town of San Fernando. [c] The percent of LAS is related to total amount of LAS measured in the environment, and it has been calculated taking into account the suspended solids content measured in each station.

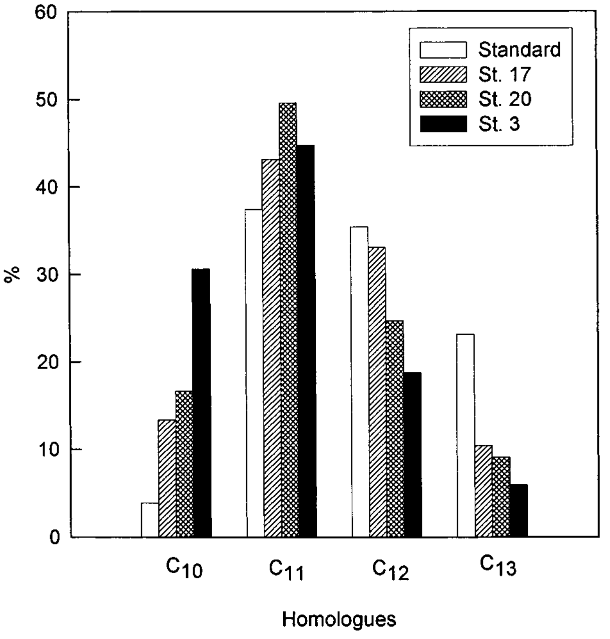

FIGURE 5. Proportional distribution (%) of the LAS homologues at three stations, with increasing residence time (stations 17 < 20 < 3).

TABLE 3. Percentages of the Homologues of LAS in the Water Column, at Different Depths

depth (m)	C_{10} (%)	C_{11} (%)	C_{12} (%)	C_{13} (%)
0.0–0.005	13.37	43.12	33.08	10.43
0.2	11.84	48.03	31.61	8.52
0.5	11.72	47.67	31.99	8.63
1.0	13.31	50.36	29.46	6.87
2.0	14.78	50.67	28.49	6.06
4.2	15.07	51.65	27.75	5.53

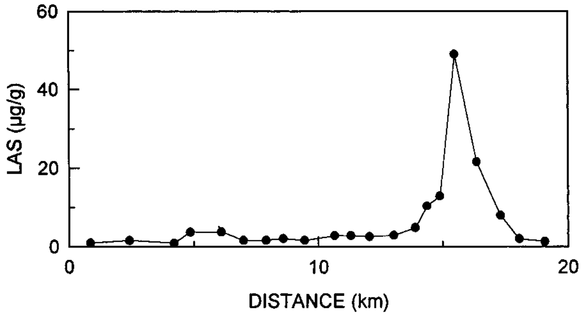

FIGURE 6. Spatial distribution of the concentration of LAS in sediment.

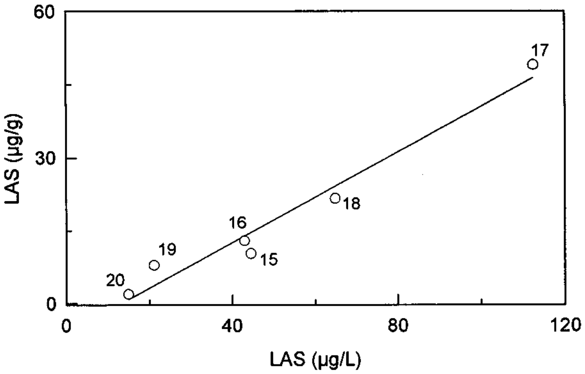

FIGURE 7. Representation of the concentration of LAS in sediment against the concentration of LAS in water, in the northern part of the zone studied.

before to describe the results of trials of sorption of LAS by both continental (*14*) and marine (*13*) sediments. In our case, the values obtained for *K* and *n*, when C_s is expressed in μg g^{-1}, and C_w in μg mL^{-1}, are 66 and 1.39, respectively. These values are similar to those obtained for the shorter chain homologues in the trials carried out on LAS sorption by marine sediments with characteristics similar to those sampled in this study (*13*). In other parts of the study zone, the relationship between the LAS concentration in the sediment and that in the supernatant water is similar to that described for the northern part of the zone, although it has not been possible to establish a statistically significant functional relationship between the two variables. Probably, this is mainly due to the greater variability in the physical characteristics (granulometry) of the sediment existing in the southern part of the channel.

The influence of the length of the alkylic chain on the process of sorption is demonstrated by the distribution of homologues shown by the LAS incorporated into the sediments (Figure 8). Differently from that which occurs in the water, the proportion of each homologue between C_{10} and C_{13} in the sediment increases in line with the number of carbon atoms in the chain. Appreciable quantities of the homologue C_{14} have even been found, despite its proportion in commercial LAS being very low (0.2%) and despite its greater biodegradability. As has been suggested on the basis of results obtained from sorption trials carried out in the laboratory (*13*), this fact has a significant environmental repercussion, since it produces the transfer preferentially of the more toxic homologues in water (*7*) to an environmental compartment where their persistence is very high. The finding of the more biodegradable homologue in aerobic conditions (C_{14}) in the sediment confirms the suggestion that the degradation of LAS in anoxic conditions is extremely slow.

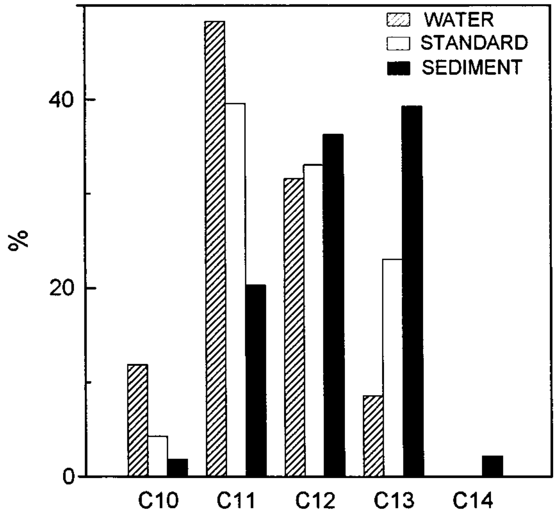

FIGURE 8. Average values of the distribution of homologues of LAS in water and in sediment, for the northern part of the Sancti Petri Channel.

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Literature Cited

(1) Takada, H.; Ogura, N. *Mar. Chem.* **1992**, *37*, 257.
(2) Takada, H.; Ishiwatari, R.; Ogura, N. *Estuarine, Coastal Shelf Sci.* **1992**, *35*, 141.
(3) Takada, H.; Ogura, N.; Ishiwatari, R. *Environ. Sci. Technol.* **1992**, *26*, 2517.
(4) Terzic, S.; Hrsak, D.; Ahel, M. *Mar. Pollut. Bull.* **1992**, *24*, 199.
(5) Terzic, S.; Ahel, M. *Bull. Environ. Contam. Toxicol.* **1993**, *50*, 241.
(6) Matthijs, E.; Stalmans, M. *Tenside, Surfactants, Deterg.* **1993**, *30*, 29.
(7) Prats, D.; Ruiz, F.; Vázquez, B.; Zarzo, D.; Berna, J. L.; Moreno, A. *Environ. Toxicol. Chem.* **1993**, *12*, 1599.
(8) Terzic, S.; Ahel, M. *Mar. Pollut. Bull.* **1994**, *28*, 735.
(9) Terzic, S.; Hrsak, D.; Ahel, M. *Water Res.* **1992**, *26*, 585.
(10) McAvoy, D. C.; Eckhoff, W. S.; Rapaport, R. A. *Environ. Toxicol. Chem.* **1993**, *12*, 977.
(11) Quiroga, J. M.; Sales, D. *Bull. Environ. Contam. Toxicol.* **1990**, *44*, 851.
(12) Quiroga, J. M.; Sales, D.; Gómez-Parra, A. *Toxicol. Environ. Chem.* **1992**, *37*, 85.
(13) Rubio, J. A.; González-Mazo, E.; Gómez-Parra, A. *Mar. Chem.* **1996**, *54*, 171.
(14) Amano, K.; Fukushima, T. *J. Environ. Sci. Health* **1993**, *28*, 683.
(15) Gómez-Parra, A.; Forja, J. M. *Mar. Chem.* **1994**, *45*, 53.
(16) Grasshoff, K.; Ehrhardt, M.; Kremling, K., Eds. *Methods of Seawater Analysis*; Verlag Chemie: Weinheim, Germany, 1983.
(17) González-Mazo, E.; Quiroga, J. M.; Sales, D.; Gómez-Parra, A. *Toxicol. Environ. Chem.* **1997**, *59*, 77.
(18) Nakae, A.; Tsuji, K.; Yamanaka, M. *Anal. Chem.* **1981**, *53*, 1818.
(19) Di Corcia, A.; Marchetti, M.; Samperi, R.; Marcomini, A. *Anal. Chem.* **1991**, *63* (3), 1179.
(20) González-Mazo, E.; Honing, M.; Barceló, D.; Gómez-Parra, A. *Environ. Sci. Technol.* **1997**, *31*, 504.
(21) Quiroga, J. M.; Sales, D.; Gómez-Parra, A. *Water. Res.* **1989**, *23*, 801.
(22) Hon-nami, H.; Hanya, T. *Water Res.* **1980**, *14*, 1251.
(23) Kikuchi, M.; Tokai, A.; Yoshida, T. *Water Res.* **1986**, *20*, 643.
(24) Schöberl, P. *Tenside, Surfactants, Deterg.* **1989**, *26*, 86.
(25) Matthijs, E.; De Henau, H. *Tenside, Surfactants, Deterg.* **1985**, *22*, 299.
(26) Moreno, A.; Ferrer, J. *Tecnol. Agua* **1990**, *66*, 41.
(27) Marcomini, A.; Pavoni, A.; Sfriso, A.; Orio, A. A. In *Proceedings of the 3rd International Conference on Environmental Contamination;* CEP Consultants Ltd.: Edinburgh, **1988**; 94.

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