

Fate of a Xylene Slick at Sea and Influence of Meteorological Conditions

Stéphane Le Floch, Mélanie Fuhrer, François Merlin - Cedre, Brest, France
 Laurent Aprin, Pierre Slangen - Ecole des Mines d'Alès, Alès, France
 Magali Mouries - Nymphéa Environnement, Cassis, France
 André Laflamme - Transport Canada, Ottawa, Ontario, Canada
 Stephane.Le.Floch@cedre.fr

Abstract

Shipping is a valuable solution for transporting large quantities of non-perishable goods over long distances.

While the shipping of liquids in bulk is still largely dominated by oil, the percentage of chemicals is sharply rising in certain regions of the world, such as the English Channel through which 30 million tonnes were transported a year in 1988, compared to 150 million tonnes a year in 2000 (*HASREP*, 2005). This increase in maritime traffic induces an increase in the risk of accidents and, consequently, an increased risk of spills of cargo at sea (collision, grounding, etc.). Furthermore, the wide diversification of the chemical nature of products transported, their highly varied physico-chemical properties that generated very different behaviours at sea, their potentially hazardous nature both for humans and for the ecosystem as a whole, constitute a set of factors that significantly complicate the operational response strategy to be defined in the case of a spill. So as to better identify the risks which certain chemicals transported by sea may present, the notion of Hazardous and Noxious Substances (HNS) emerged in Europe in 2000.

Depending on the substances concerned, location and characteristics of the incident, response can therefore range from the "leave alone" option, as in the case of the Maltese chemical tanker *Balu* which sank in the Bay of Biscay in 2001, releasing her 8,000 tonnes of sulphuric acid, to the deployment of major on-land and at-sea response means, as in the case of the chemical tanker *Ievoli Sun* which sank in the Channel in 2000, for which response operations included both response at sea (treating the cargo) and monitoring of potential pollution on the shoreline (sampling of coastal waters and air quality measurements).

Several international, regional and national authorities, aware of the risk connected with the transport of chemicals by sea, have published operational guides to describe the possible response options. For instance, IMO has published several manuals (IMO, 1987, 1992), REMPEC has done the same for the Mediterranean (REMPEC, 1996, 1999, 2004); we can also mention the Helsinki Baltic Sea Convention manuals (HELCOM, 1991, 2002, 2007), the North Sea Bonn Agreement manuals (Bonn Agreement, 1985, 1994) as well as on a national level in France the *Cedre* response guides each of which is specifically dedicated to a given chemical.

Alongside this information, which ranges from general (i.e. the case of a guide on response at sea to hazardous substances), or specific (i.e. guides that characterize the behaviour and identify the risks related to a given product), the Standard European Behaviour Classification system or SEBC code determines the short-term behaviour of any chemical as long as its physico-chemical characteristics are known. This tool is of the utmost importance for operational staff in charge of response, as it can be used in an emergency to indicate the main components of the

ecosystem that will be affected. However, experience has made clear that this code can be tricky to use as it can be difficult to understand how the environmental factors (wind, sunshine, water and air temperature, etc.) will affect the categorization of the chemicals. Indeed these parameters have a very significant influence on the short-term fate of chemicals in the marine environment, in particular on their evaporation kinetics (transfer from the water surface to the atmosphere), their dissolution kinetics (transfer from the water surface to the water column) and their transport and transformation processes in the aqueous phase.

1 Behaviour of Substances with Low Reactivity

In the case of a spill, response follows the following three generally accepted scenarios: i) response is not possible because the spill occurred in a geographical environment that is incompatible with reasonable response times; ii) response is not possible due to the reactivity of the substances (major, imminent danger); iii) response is possible.

If, in the latter scenario, the substance spilled is not particularly reactive, it is important to bear in mind that the slick formed will evolve. Immediately after its accidental release into the marine environment, the substance will tend to move into different components of the ecosystem, i.e. the atmosphere, the sea surface, the water column and the seafloor. These different forms of distribution are gathered under the umbrella term of “fate”, for which we generally distinguish short term behaviour (a few hours) and longer term behaviour (up to several years).

1.1 Short-term Behaviour

Short-term behaviour covers a duration ranging from a few minutes to a few hours, and it must be known in order to implement response measures and techniques. It was in light of this observation that the Standard European Behaviour Classification, or SEBC code, was developed (Cedre, 1988; Bonn Agreement, 1994; GESAMP, 2002).

This code classifies the behaviour of chemical substances according to their state and some of their physical properties, i.e. density, vapour pressure and solubility. The state of the substance refers to whether it is in the form of a gas, a liquid or a solid at 20°C. The density, defined as the mass per unit volume compared to that of seawater (1.03 g.cm^{-3} , at 20°C), determines whether a substance will float or not. The vapour pressure is defined as the partial vapour pressure of a compound in equilibrium with its pure condensed phase (liquid or solid). It is generally accepted that a floating substance will not evaporate if its vapour pressure is lower than 0.3 kPa and it will evaporate rapidly if its vapour pressure is higher than 3 kPa. For dissolved substances, evaporation occurs when the vapour pressure is greater than 10 kPa. Solubility is defined as the maximum abundance of a compound per unit volume in the aqueous phase when the solution is in equilibrium with the pure compound in its initial state. It is usually measured at 20°C, at a pressure of 1 atmosphere, in distilled water (zero salinity). The criteria used are different according to the physical state of the substance. A substance is considered insoluble if its solubility is less than 0.1% for liquids and less than 10% for solids. At the other end of the scale, the phenomenon of dilution will take place if the solubility is greater than 5% for liquids and greater than 100% for solids.

These limits are presented in Figure 1 and can be used to identify 12 types of behaviour, which are summarized in Table 1.

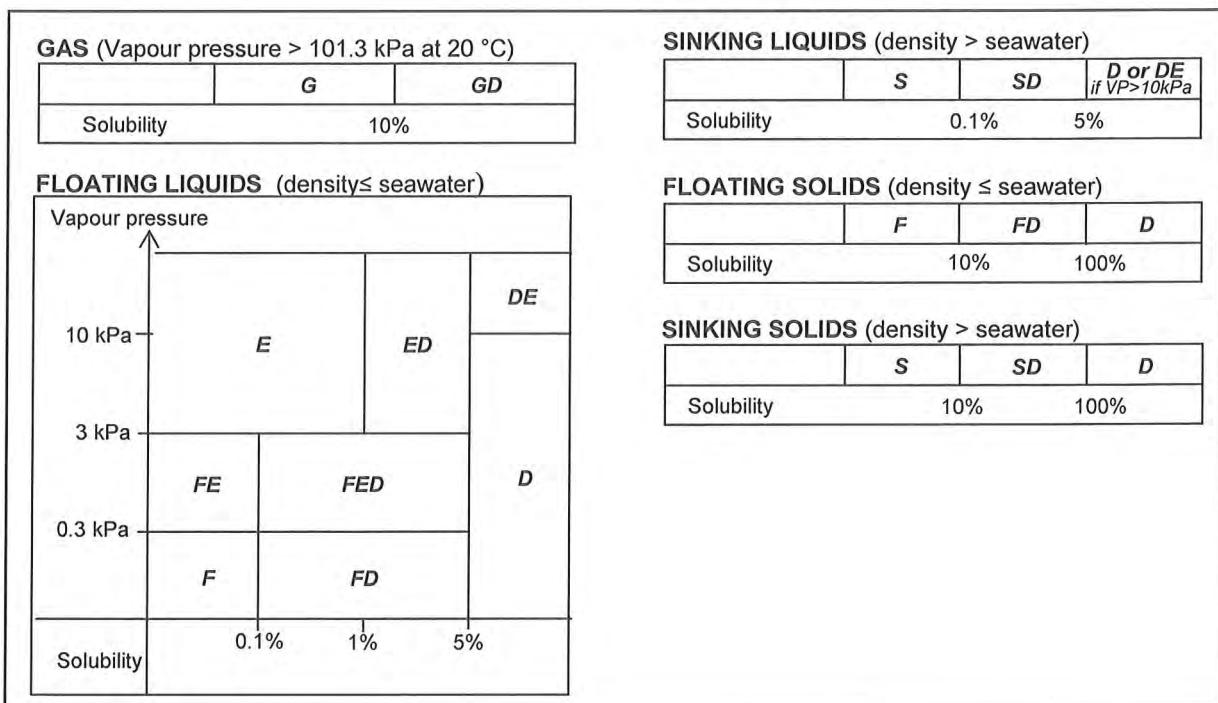


Figure 1 SEBC Classification of Chemical Products according to their Physical State and Physical Properties

Table 1 The 12 Behaviour Groups according to the Standard European Behaviour Classification System

Main group		Subgroup	
G	Gas	GD	Gas that dissolves
E	Evaporator	ED	Evaporator that dissolves
F	Floater	FE	Floater that evaporates
		FD	Floater that dissolves
		FED	Floater that evaporates and dissolves
D	Dissolver	DE	Dissolver that evaporates
S	Sinker	SD	Sinker that dissolves

For example, benzene is classed E (vapour pressure = 10 kPa at 20 °C), i.e. it is a liquid chemical that immediately evaporates following a spill.

1.2 Longer Term Behaviour

In terms of response, it is also useful to be aware of longer term behaviour as it provides information on the fate of a pollutant that cannot be removed from the environment through response operations. The residual chemical will spread through

the atmosphere and/or the water column where it may become associated with sediment particles in suspension before settling. In water and air, the ultimate fate of chemicals will depend on the processes of dispersion, breakdown and other physical, chemical or biological transformations. These processes can extend over periods ranging from a few months to several years when the substance remains in the environment.

In terms of behaviour, all these phenomena will contribute, in the best case scenario, to the disappearance of the product and, in the worst case scenario, to its accumulation or even its transfer into another part of the ecosystem.

To come back to our example of benzene, following a major spill, even though this product is ranked E according to the SEBC code, it is not completely insoluble ($\text{solubility} \approx 1.7 \text{ g.L}^{-1}$ i.e. $\leq 0.17\%$) and it is therefore logical to suppose that a percentage of the quantity spilled will enter the aqueous phase, causing water contamination, whereby the greater the quantity spilled, the greater the contamination. However, as benzene has a short half-life in the aqueous phase ($t_{1/2} = 5.6 \text{ H}$), according to the theoretical calculation developed by Lyman et al. (1982), this dissolved quantity will rapidly be returned to the atmospheric compartment, and, in the long term, the near unique consequence of a benzene spill will be contamination of the atmosphere. The pollution will therefore persist in the environment, and will mainly result in the production of phenols, nitrophenols, nitrobenzene, formic acid and peroxyacetyl nitrate according to photo-oxidation processes.

1.3 Influence of Environmental Factors

Spill response fundamentally requires rapid identification of the compartment(s) of the ecosystem in which the pollutant will accumulate so as to implement the appropriate means to reduce, or even eliminate, its impact. While the SEBC code provides initial elements of response, the operational personnel in charge of response must interpret this classification critically so as to assess whether the specificities of the environment in which the incident has occurred will alter the results or not. This critical interpretation may go as far as altering the categorization of a substance as in the case of the industrial incident at the Jilin plant in China.

On 13 November 2005, an explosion occurred in a petrochemical plant resulting in a spill of around a hundred tonnes of various substances into the Songhua River. Analyses conducted by the Chinese Research Academy of Environmental Science showed that the water contained benzene and nitrobenzene concentrations 30 to 100 times higher than the accepted standards, of 0.01 mg.L^{-1} and 0.017 mg.L^{-1} respectively (Ambrose, 2006; UNEP, 2006). The pollution was carried by the current and concentrations 34 times higher than standard levels were found in Harbin, 380 km downstream of Jilin, and 10 times higher than standard levels in Jiamusi, 550 km downstream. This decrease can be explained both by the processes of dilution and adsorption of the chemicals by organic matter present in the water, either in suspension, or on the banks (Levshina et al., 2009). These unexpectedly high concentrations can be explained by the particularly harsh weather conditions on site at this time of year (negative temperatures [$< -10^\circ\text{C}$] and river partially frozen) which restricted evaporation, promoting dilution.

In the context of its general research program, *Cedre* conducted trials in floating cells to fully understand the influence of environmental parameters on the behaviour of benzene. These *in-situ* trials involved three 25-L spills of benzene, in floating pens anchored in Brest estuary, during which transfers between the slick and the atmosphere, and the slick and the water column were studied. Evaporation was monitored using a PID (Photo Ionization Detector) and dissolution was characterised by *in-situ* measurements at four different depths (0.5, 1, 1.5 and 2 m), using an onboard spectrofluorometer (SF-UV Turner Design 10AU), completed with sampling of the water column followed by GC-MS analysis. These floating cells are equipped with a weather station that records the wind speed and direction, sunshine, rainfall and air temperature.

The observations made during this series of trials showed that benzene slicks disappear completely from the water surface in less than 4 hours. Evaporation was intense during the first hour, then decreased, coming to a total stop after 3 hours. For the first hour, the average benzene concentration was 325 ppm, then 133 ppm during the second hour, and finally, practically nil by the third hour (3 ppm, Figure 2). These rapid evaporation kinetics represent the behaviour of an evaporating liquid, which correlates perfectly with its classification E according to the SEBC code. The high degree of variation in amplitude between minimum and maximum concentrations over time (550 ppm during the first hour then less than 240 during the second) has two explanations: the decrease in the surface area of slicks over time (decrease in exchange interface) and a decrease in wind speed (between the beginning of the trial and T+3H the wind speed dropped from 30 km.h⁻¹ to less than 10 km.h⁻¹). The higher the wind speed, the greater the evaporation, and often higher wind speeds are associated with swirling winds, explaining the large gaps between minimum and maximum concentrations. Within this experimental context, the wind factor did not affect the SEBC result for benzene as, on the contrary, it favoured transfer kinetics from the surface into the atmosphere, justifying its classification as an evaporator (E).

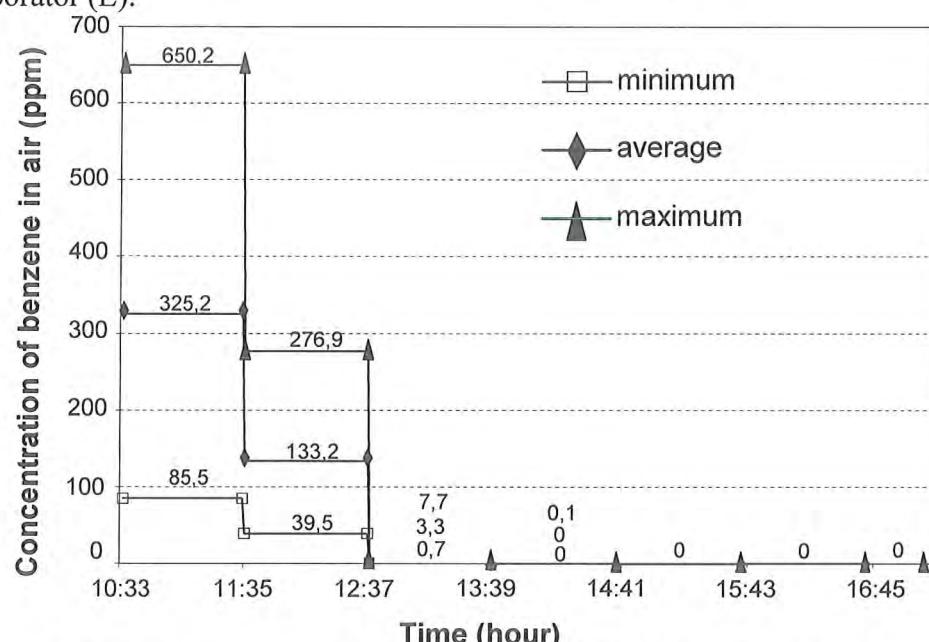


Figure 2 Variations in Benzene Concentrations in the Atmosphere

In terms of dissolution, at T+2h, benzene was detected at the four different depths with concentrations of between 1.5 mg.L^{-1} at 0.5 m and 1 m and, 1 mg.L^{-1} at 1.5 m and 2 m. From T+6h, it was possible to consider the benzene concentration in the water column as background levels (Figure 3). These concentration levels are lower than benzene's solubility limit and support its SEBC classification as E. However, these results must be taken in the context of the weather conditions at the time, i.e., an outside temperature of 15°C and water temperature of 10°C ; very different conditions to those in the case of the Jilin incident.

Thus in the event of a spill, benzene's main behaviour will be type E, i.e., it will accumulate in the atmosphere, and the greater the wind speed, the faster it will accumulate. However, in the case of extreme temperatures, as illustrated by the Jilin case, dissolution will prevail, underlining the need to adjust the interpretation of the SEBC code, established from data obtained in standardized laboratory conditions. Although this point is discussed in documents which employ the SEBC code, it remains largely underestimated.

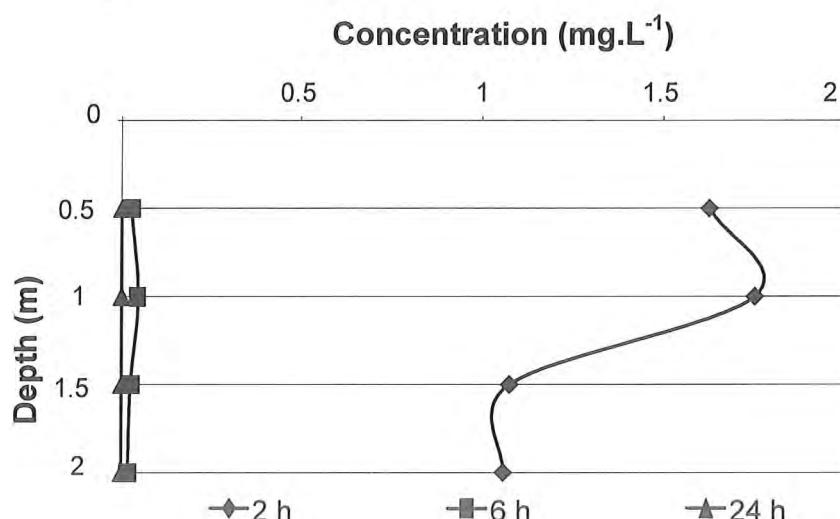


Figure 3 Evolution of Benzene Concentrations in the Water Column

2 Fate of a Xylene Slick at Sea

The efforts made to study the behaviour of xylene when spilled at sea can mainly be explained by two facts: on the one hand, this substance is one of the most commonly transported cargos along the French coastline, both in terms of tonnage and frequency (ranking ninth in the list of 100 chemicals most commonly transported in Europe; HASREP, 2005), and on the other hand, it is identified by the SEBC code as FE, a Floater-Evaporator, i.e., a product for which response is possible.

2.1 Physico-Chemical Properties of Xylenes

This chemical was chosen to be studied due to its physico-chemical characteristics (floater), its widespread use in industry (solvent widely used in the manufacture of paints, varnishes, glues, printing inks, insecticides, etc.) and its

consequent high level of transportation by sea (tonnage and frequency), as well as following an analysis of past accidents, in which it features with the *Grape One* incident (1993).

Xylene is produced from oil-based raw materials from catalytic reforming or catalytic cracking. Three chemical molecules are in fact often grouped under the term xylene: ortho-xylene, meta-xylene and para-xylene. In 1995, the total production of xylene in Western Europe represented 2.7 million tonnes (Ullmann, 1996).

In terms of hazards for human health, the inhalation of vapours can cause sore throats, drowsiness, nausea, irritation of nasal passages and the throat as well as breathing difficulties. In the case of high concentrations, there is a risk of depression of the central nervous system, or even death.

In terms of behaviour in the marine environment, xylene is described as a product which, once spilt on the water surface (density = 0.8), will evaporate almost completely. Xylene vapours are heavier than air and travel at the water surface. A small part may dissolve (solubility $\approx 0.2 \text{ g.L}^{-1}$).

2.2 Experimental Protocol

The experimental protocol followed was the same as for the benzene experiments: 25 L spill in a 9 m² cell, monitoring of evaporation with a PID and of dissolution by *in-situ* measurements (SF-UV turner design type 10AU) completed by sampling at various depths.

Two series of trials, in different climatic conditions, were conducted so as to investigate the influence of environmental parameters: series No.1, calm, sunny weather; series No.2, cloudy weather and rough sea.

2.3 Results

Observations of Xylene's Behaviour

The observations made during series No.1 were very different to those of series No.2: for No.1, the slicks disappeared rapidly preventing surface sampling (Figures 4 and 5), whereas for No.2 the slicks persisted at the water surface and emulsions were able to be identified (Figure 6 and 7).

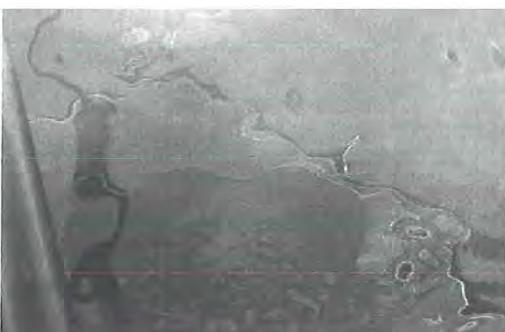


Figure 4 Xylene Slick Immediately After Spill



Figure 5 Xylene Slick 4 Hours After Spill in Calm Weather



Figure 6 Xylene Slick 4 Hours After Spill in Rough Sea

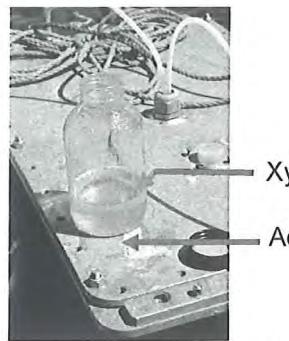


Figure 7 Sample Taken During series No.2, Presence of an Emulsion at the Water Surface

On-site Sea and Weather Conditions

For series No.1 weather was calm (sunny, little wind and percentage humidity in air systematically less than 70%). For series No.2, the sea state was slightly rough according to the Douglas Sea Scale (degree 3) with the presence of wind and a humidity percentage in air of around 80% (Figures 8, 9 and 10).

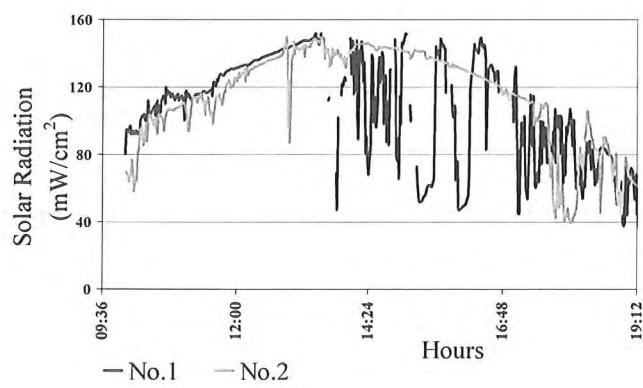


Figure 8 Evolution of Solar Radiation during Xylene Spills

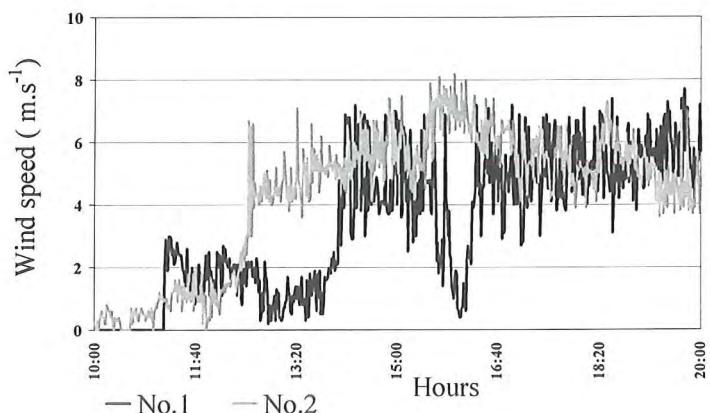


Figure 9 Evolution of Wind Speed during Xylene Spills

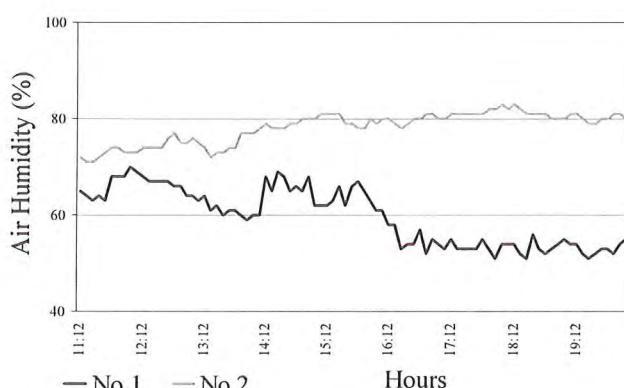


Figure 10 Evolution of Air Humidity during Xylene Spills

Emulsification

Surface sampling during series No.2 detected the presence of emulsions. At T+7h, the water content was practically identical for each of the slicks, at around 55%. However, these emulsions were not persistent as the slicks had totally disappeared from the water surface by the following day. This process is directly dependent on the on site weather conditions: emulsification of the slick can only occur if surface agitation is sufficient, i.e., in the absence of wind and waves, a xylene slick will not emulsify, while it may form a significant but unstable emulsion in the presence of wind and waves.

Evaporation

The formation of a gas cloud above the slick was monitored with a PID. The results obtained show a significant difference for xylene concentrations in air between series No.1 and No.2 spills. During series No.1, the persistence time of the slick at the water surface was lower than that of series No.2 due to immediate, intense evaporation. The xylene vapour concentrations were 5 to 10 times higher for series No.2 (Figure 11).

Xylene isomers are particularly photosensitive molecules with very short atmospheric half-lives, of less than 3 hours (Brice and Derwent, 1978). During the trials, the evaporation kinetics were higher, i.e., higher atmospheric xylene concentrations with peaks three times greater than the ELV estimated at 100 ppm (Ineris, 2006), for the highest level of solar radiation (Figure 8), which confirms the importance of this parameter, meaning that in the event of a spill in calm, sunny conditions, the evaporation process will be intense.

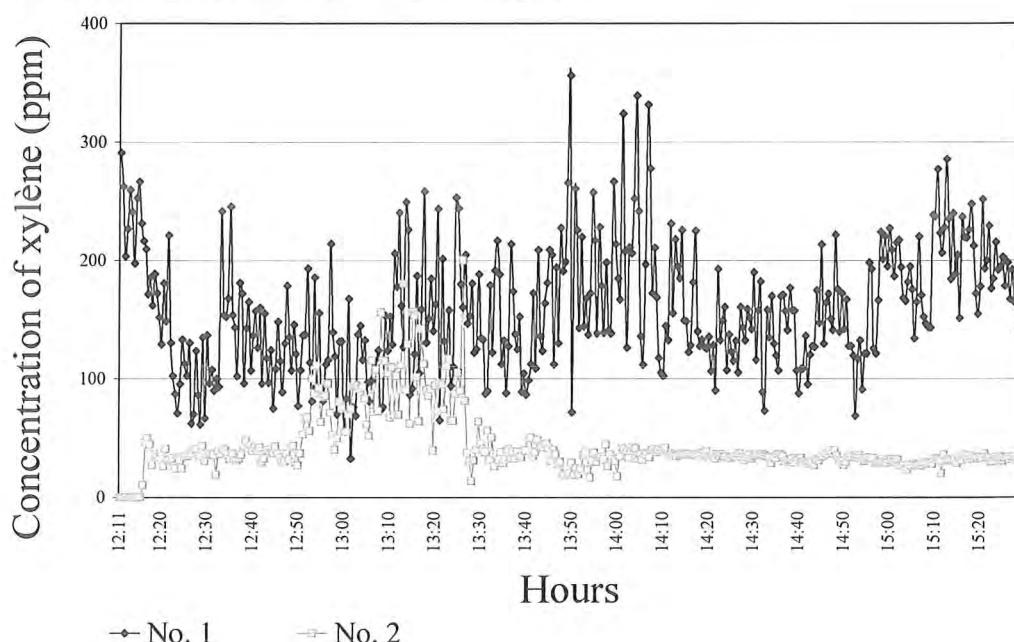


Figure 11: Atmospheric Xylene Concentrations during Spills No.1 and No.2.

However, this result should not alter the influence of the wind factor on evaporation processes as it is the predominant factor for the majority of chemicals.

Evaporation kinetics are positively correlated with wind speed. In the case of these trials, this influence was apparent in series No.2 with a peak in atmospheric xylene concentrations between 12:50 and 13:30 (Figure 11), which can be explained by the appearance of wind on site at that time (wind speed greater than 2 m.s^{-1} from 12:00). The drop in gas concentrations after 13:30 can be explained by the emulsification processes that slowed down evaporation phenomena.

Dissolution

Dissolution during series No.1 was significantly lower than that measured during series No.2. The concentrations measured were systematically lower than 1 mg.L^{-1} , i.e., below xylene's theoretical solubility estimated at 200 mg.L^{-1} but higher than the PNEC of p-xylene estimated at $26 \mu\text{g.L}^{-1}$, while No.2 concentrations were around 10 mg.L^{-1} during the first 5 hours following the spill, and the substance was still detected on the second day.

Behaviour of Xylene

During these trials, several 25 L xylene spills were reproduced on different dates so as to work in different climatic conditions. During series No.1, the onsite conditions were calm (sunny weather, no wind) while during series No.2 wind was present and the air humidity level was high ($> 70\%$). The results obtained highlighted the dual behaviour of this chemical which did not persist for long at the water surface due to a high evaporation rate in the climate conditions of series No.1, whereas it persisted for longer in the case of sufficient surface agitation to cause emulsification of the slick, resulting in the secondary effect of promoting dissolution at the expense of evaporation. The behaviour of this product is therefore not unique but directly depends on the sea and weather conditions onsite at the time of the spill, in particular surface agitation, wind, sunshine and water temperature.

These results were obtained for small spills (25 L) with controlled spread: the slick could not exceed 9 m^2 , which is the surface area within a floating cell. However, in the case of a real spill, the spread of the substance will determine the intensity of exchanges between the atmosphere and the water column, i.e., the greater the surface area of the slick, the higher the evaporation and dissolution rates may potentially be. To overcome this constraint, *Cedre*, in collaboration with the *Ecole des Mines d'Ales* and the company Nymphéa Environnement, and with logistical support from the French Navy, organized an experiment in the open sea designed to investigate the behaviour of xylene *in-situ*.

3 Experimentation in the Mediterranean

3.1 Experimental Location

The experiment was carried out at the location defined by $42^{\circ}50'N$ and $005^{\circ}05'E$, which is 50 miles off the French shoreline outside the Ligurian current in the Mediterranean Sea.

3.2 Experimental Protocol

It was decided to release 2000 L of xylene. Discharge was carried out via a floating cylinder launched using the crane of the ship. The product was transferred into the cylinder by pumping (using electric pumps ATEX), then 2 m^3 was

transferred, the crane lifting the cylinder thus releasing the totality of the product at the water surface. The different steps are described in Figure 12.

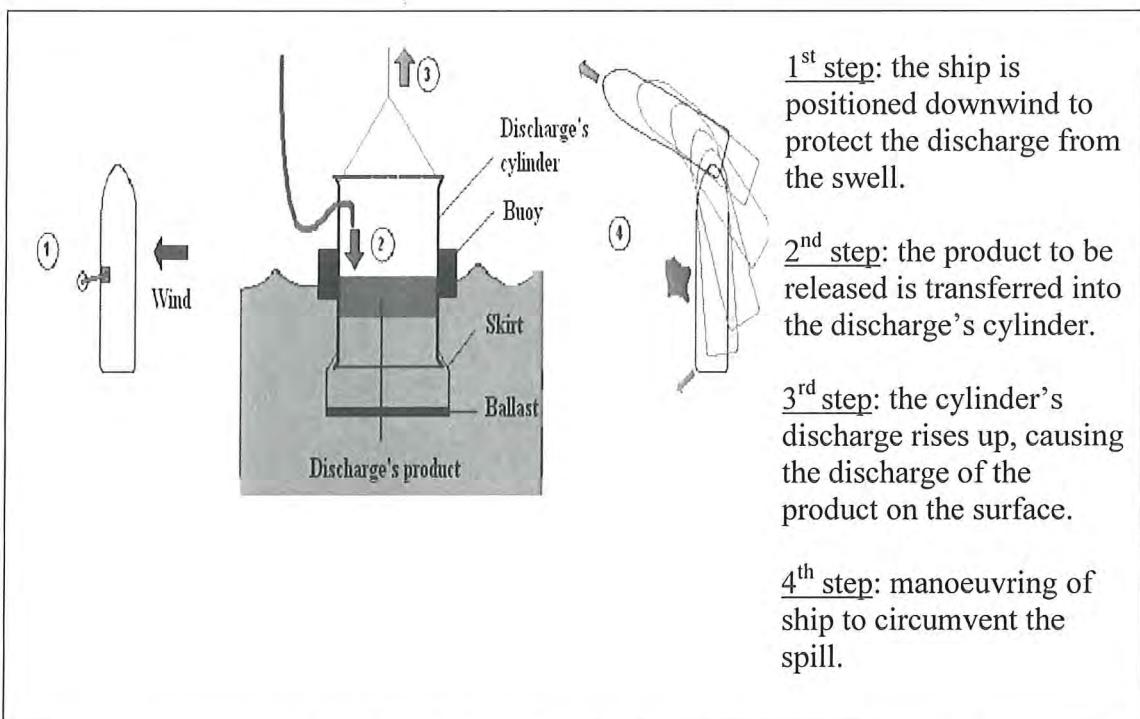


Figure 12 Description of the Discharge Technique of the Products

For behaviour monitoring, the same techniques were used: PID for the air and SF-UV plus sampling for the seawater. In addition, the drift of the slick was tracked by using Argos buoys.

For the air, the formation and displacement of the gas clouds were evaluated with two PID detectors which were located at two different levels (2 and 3 m high) on a Remote Operated Vehicle (ROV). The ROV was operated directly from the supply ship. These detectors were combined with a Global Positioning System (GPS).

For the seawater column, *in-situ* measurements were performed with a SF-UV onboard a zodiac at two different depths (1 and 2 m).

3.3 Results

The drift of the xylene slick over time is shown in Figure 13. The on-site measurements are represented by the crosses, with T0 corresponding to the first measurement (Figure 13). The slick was detected at the sea surface for less than 2 hours.

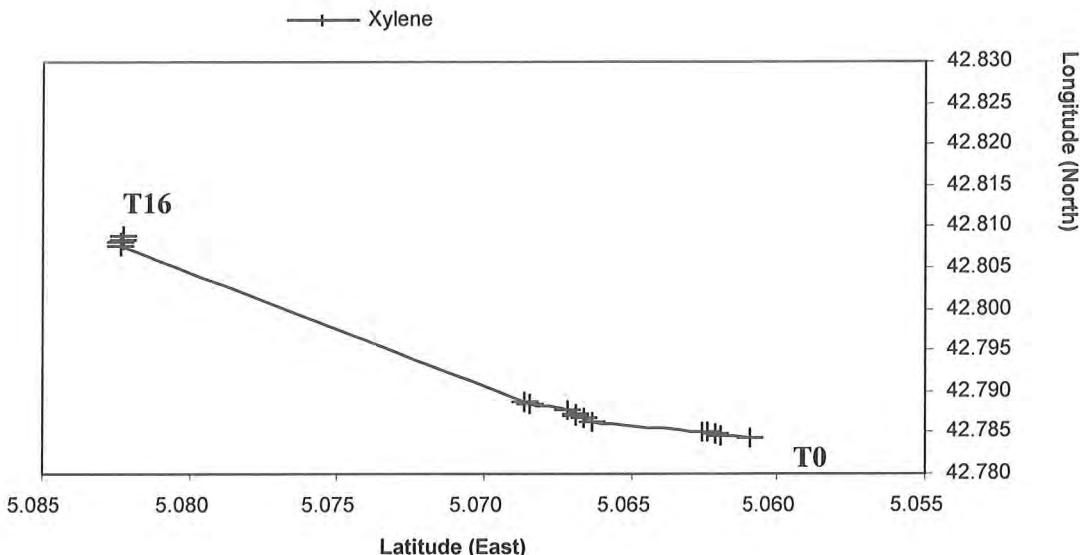


Figure 13 Drift of Xylene and n-Butanol Slicks (1st day)

For the atmosphere, sensor acquisition was started before the xylene was released by positioning the ROV close to the discharge's cylinder.

The most important point is that concentrations were detected for only 37 minutes (Figure 14). There is a similar trend of concentration measurements between PID 1 and PID 2 with a period of 10 minutes with high concentrations, then no concentration measurements for 10 minutes, followed by further detection of low values (lower than 100 ppm) for 12 minutes. The blank period was probably due to the difficulty in positioning the ROV on the slick and measuring the xylene evaporation during the test. It is clear than the main objective was to monitor the evaporation rate from the slick and not to track the drift of a xylene cloud.

Comparison between the two detectors shows similarities with an abrupt rise in concentration that reached 700 ppm for PID 2. This concentration is lower than the IDLH value for xylene (900 ppm) but was still a dangerous concentration for responders. Figure 14 shows a difference between the detectors with lower concentration values for PID 1, which is located higher than PID 2. This observation confirms that xylene vapours are heavier than air and are more concentrated in a zone located near the sea surface.

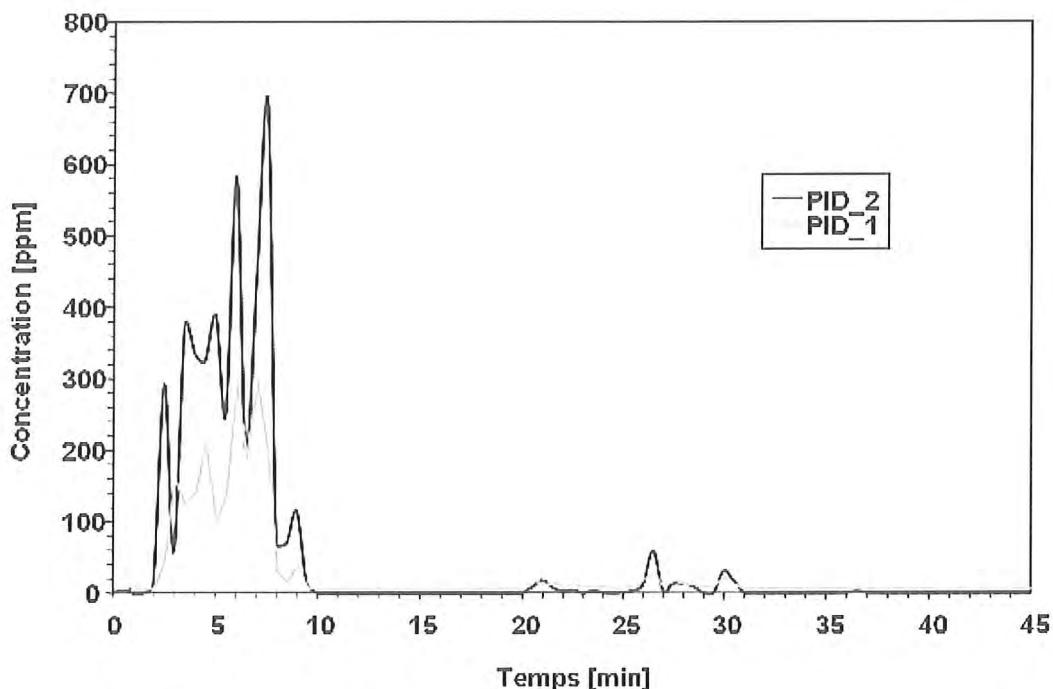


Figure 14 Evolution of the Xylene Concentration Measurements in the Atmosphere

Figure 15 shows the evolution of the sea water concentration of xylene. Even if the product was detected in the water during the first 3 hours, concentrations were never higher than $20 \mu\text{g.L}^{-1}$.

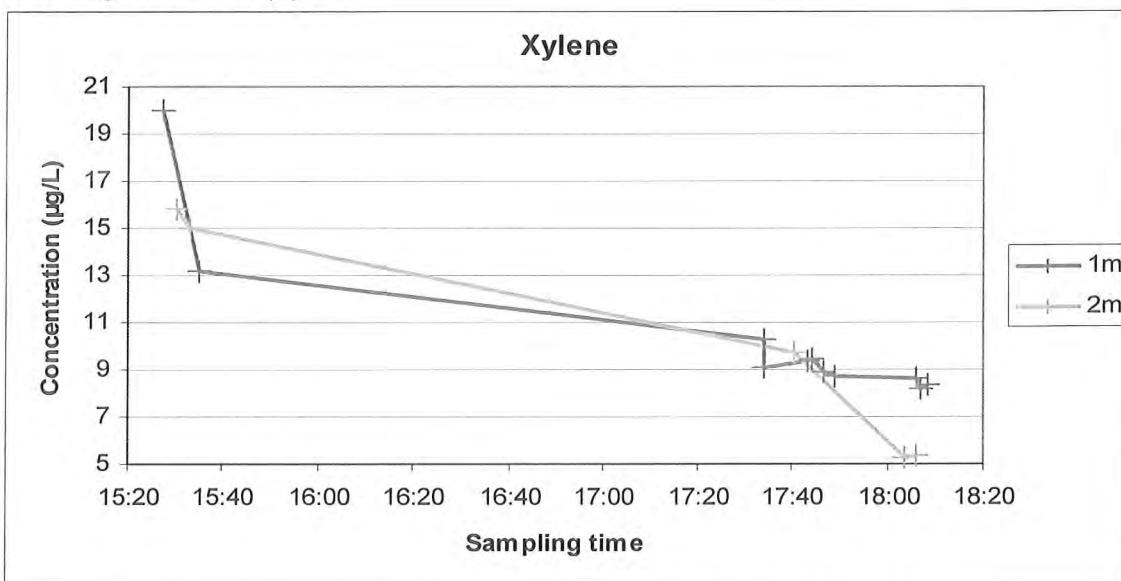


Figure 15 Evolution of Xylene Concentrations in Sea Water Over Time

3.4 Behaviour at Sea

Although all the results have not yet been processed, particularly the remote sensing data that should enable an estimation of the evolution of the slick surface area over time, an initial behavioural trend has become apparent.

In the conditions of this trial, calm sea and sunny conditions with no wind, it appears that the persistence of the xylene slick at the water surface is limited in time: after less than 2 hours, the slick can no longer be seen and, due to maximum spreading, it was never possible to sample the chemical (absence of emulsion). As for the vapours, the cloud remained just above the water with a maximum concentration at an altitude of 1 m and, less than an hour after the spill, the concentrations were lower than the PID detection limit, making them no longer quantifiable. However, even though the concentrations in water were lower than this substance's solubility limit, xylene was detected up to three hours after the spill, at both depths sampled.

4 Conclusion

In the event of an incident resulting in a chemical spill at sea, those in charge of emergency response must draw upon data from the literature to determine the fate of the pollution and the most appropriate response strategy. Among the available tools, the SEBC code is very useful as it can be used to rapidly identify the part of the environment in which the pollutant will mainly accumulate, based on laboratory data. Despite many warnings, however, the influence of environmental factors is underestimated, which may lead to mistaken interpretations. Thus, in the Jilin incident, the authorities had to deal with pollution of the aqueous component although the spilled product was classified as an evaporator, i.e., pollution of the atmosphere.

It was therefore decided that the influence of sea and weather conditions on a widely transported substance, xylenes, would be characterized. The results obtained show the product's dual behaviour according to surface agitation which is directly dependent on wind and waves. In terms of response, this means that we must take into account problems related to the following:

- Spills of floating-evaporating products (FE) on calm water bodies, with a wind speed of less than 3 m.s^{-1} and an outdoor temperature greater than 10°C ; In practice, response to the formation of a gas cloud should mainly consist of recovering the slick with sorbent or foam covering and tracking its displacement in the atmosphere as it is toxic, or even explosive, especially as these vapours have a density greater than that of air.
- Spills of floating-evaporating-dissolving products (FED), particularly in the presence of waves and wind. Emulsification of the slick will not totally prevent evaporation so it must be taken into consideration. In these weather conditions, however, response should also consider contamination of the water column and the related catchment issues, as well as the possible impact on flora and fauna especially as xylene has a low LC₅₀.

These trials once again highlight the need to consider weather conditions when studying the behaviour of a chemical at sea. Direct consultation of the SEBC classification code is insufficient alone to identify the component of the environment that will be directly affected by the pollution. However, it appears necessary to complete this type of trial with laboratory tests that will better identify the meteorological limits at which the behaviour of a product will change rapidly. For instance, it could be useful to determine the minimum energy required for the

substance in question to form an emulsion, then to correlate this with a sea state. Although these *in-situ* trials are a valuable source of information, they can only be carried out in good weather conditions so as to minimize risks for operators. They are therefore only representative of a given situation, often far removed from the reality of a spill situation in which the weather conditions are frequently difficult (storms).

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