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Estimate of the Slick Thickness for Leaked Heavy Oil from the Sunken Nakhodka in the Sea of Japan

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Abstract : In order to estimate the thickness of leaked oil from the sunken Tanker Nakhodka in the Sea of Japan, the relationships between the slick thickness and the upward radiance from the surface of water at selected wavelengths were investigated by experiments in a test water tank. The obtained relationships and *in situ* oceanographic measurement data were used for the estimate of actual oil slick thickness. In the range of slick thickness of 0.0005 – 0.0019 mm, white and shiny stripe patterns are observed and the appearance is white. This is considered to be caused by the irregular reflection, which results from the oil droplets due to the small surface tension of the dirty city water. Above the slick thickness of 0.0027 mm, the white area decreases gradually with the increase of oil and becomes entirely brown at the thickness of 0.075 mm. When the thickness increases further, a tint of dark brown increases. No definite relationship is found between the slick thickness and the dominant wavelength of chromaticity point of the water. The relationships between the upward radiance and the oil slick thickness are as follows:

$$Y = \alpha X^{-0.0637} \quad (Lu/Ed\,(412)\;/\;Lu/Ed\,(510) \leqq 1.4)$$
$$Y = \beta X^{0.143} \quad (Lu/Ed\,(412)\;/\;Lu/Ed\,(510) > 1.4)$$

where Y : upward radiance at 510nm of oil slick
X : oil slick thickness (mm)
α, β : constant value calculated from upward radiance at 510nm of sea water at critical point
Lu : upward radiance Ed : downward irradiance
412, 510: wavelength (nm)

Near the sunken site, the oil slick thickness is estimated to be 0.014 mm in 1997 and 0.017 mm in 1998.

Keywords : *Slick thickness, Leaked heavy oil, Sunken Nakhodka, Critical point*

1. Introduction

In January of 1997, there was a large-scale heavy oil leak from the ruptured Tanker Nakhodka located at the western region of the Sea of Japan. The oil that was leaked then spread rapidly by action of wind and currents towards the northern coastal areas of Honshu Island and inflicted great damage on the fisheries. (KATSURAGI, 1999)

At that time it was estimated that the volume of the leaked oil was 5000–6000 kL based on the volume of the boat hold. However, it was very difficult to grasp the volume of oil at each different location because the leaked oil dispersed in the form of slicks. In addition, the thickness of oil slicks was very thin and the slick layers fragmented into many pieces. It is extremely important to accurately grasp the spread range and the volume of spilled oil for the preservation of environment and marine life, and the protection of the fisheries. The information is also important for the removal of

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spilled heavy oil and its treatment and/or disposal.

So far, there has been a lot of research for the detection of oil slicks and the determination of the spread range for spilled oil. For the detection, the infrared method (FORNACA *et al.*, 1995) and the fluorescence method (HOVER and PLOURDE, 1995) were employed. For the spread range, the remote image sensing from satellites was used because it can cover wide area to obtain extensive information (TSENG and CHIU, 1994). CHUBAROV *et al.* (1995) used laser light and BROWN *et al.* (1995) used laser light and ultrasonic waves for the determination of oil slick thickness. Both methods, however, are inappropriate for the determination of slick thickness for the extensive area of ocean.

In the present investigation, in order to estimate the thickness of leaked oil slicks from the sunken tanker Nakhodka in the Sea of Japan, we conducted laboratory experiments and these results are applied to estimate the thickness of leaked oil slicks.

2. Experimental Methods

The laboratory experiment was performed at the roof top of the Department of Ocean Sciences building at Tokyo University of Fisheries when the altitude of the sun was high (from 11: 30 a.m. to 3: 15 p. m., on a sunny day) in October 1997. A schematic diagram of the experimental apparatus is shown in Fig. 1. A cylindrical tank (diameter: 0.7 m, height: 0.7 m), of which the inner wall and bottom were painted black, was filled with city water to the height of 0.6m. The C-heavy oil (density: 0.96 g/cm^3) was used for the formation of oil slicks. A micropipette with the precision of 1/1000 ml was used to drop the heavy oil on the surface of the water and the water was stirred slowly so that the slick became uniform on the surface of the tank. The slick thickness was calculated by dividing the amount of dropped oil by the area of water surface. Seventeen levels of thickness were prepared between 0.0005 to 1.0 mm. A profiling reflectance radiometer, PRR600 (Biospherical Instruments Inc.) was installed at the top center of tank so as not to form the shade on the surface of water. The distance between the surface of the water and the sensor of the instrument was 0.5 m. With PRR600, the upward radiance and the downward irradiance can be measured simultaneously at the wavelength of 412, 443, 490, 510, 555 and 665 nm. The upward radiance and downward irradiance were measured for each oil slick thickness to obtain the relationship between the slick thickness and the upward radiance.

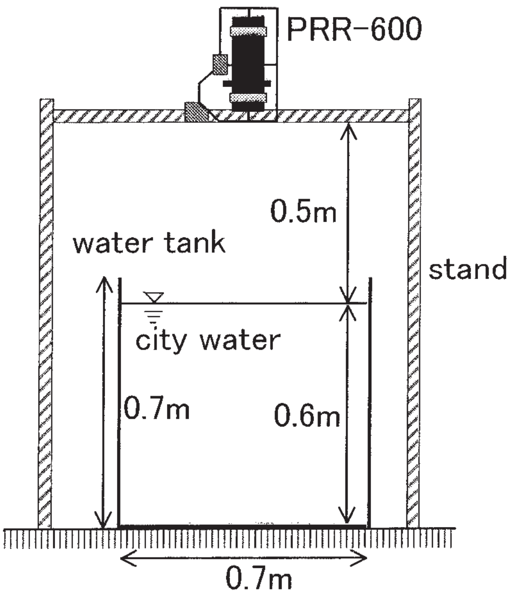

Fig. 1. A schematic diagram of the experimental apparatus.

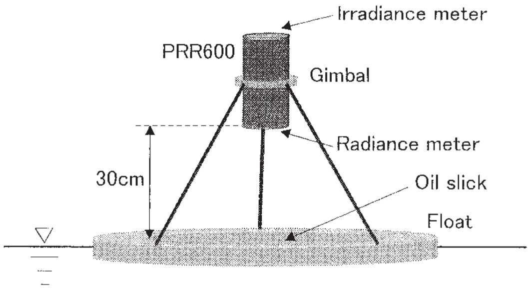

Fig. 2. A sketch of measurement on *in situ* oil slick with PRR 600.

The *in situ* oceanographic observations were carried out aboard the RT/V Umitaka-maru of Tokyo University of Fisheries in September of 1997 and 1998 around the region where the Nakhodka had sunken (37° 14.4' N, 134° 24.9' E). The profiling reflectance radiometer PRR600 was fixed to the ring float so that the light-receiving sensor was located approximately 0.3m above the surface of the water (Fig. 2). The upward radiance and downward irradiance were measured simultaneously at the selected wavelengths by setting the ring float around the region of the oil spill. For comparison, measurements were also performed for the nearby ocean surfaces where no oil slicks were observed.

3. Results and Discussion

3-1. Visual observation of water color

The experiment was started by injecting C-heavy oil at the center of water surface of laboratory tank. When the oil spread to the entire area of the water surface and the slick thickness became 0.0005 mm, many white and shiny stripe patterns were formed. When more sample oil was injected, white stripe patterns increased and the entire water surface became white at the thickness of 0.0019 mm. When oil was added further, the oil on the water surface formed masses and no longer spread easily. These results are probably due to the low surface tension of dirty city water resulting in irregular reflection of the light and droplet-like oil (SATO, 1960). When the water surface was disturbed, the masses of oil dispersed into countless spots, and formed light brown patches dotted on the water surface in white. This is considered to be the beginning of the formation of the oil slick by the mergence of droplets. When the slick becomes thicker than 0.0027 mm, the white area decreased and the brown area increased. At the slick thickness of 0.075 mm, the entire surface displayed a light brown color. Further addition of oil resulted in easy spreading. When the slick becomes thicker than 0.1 mm, the color changed to a darker brown with the increase of the thickness.

3-2. Relationship between oil slick thickness and upward radiance as a function of wavelength

The changes in downward irradiance distribution of the sun and sky during the experiment is shown in Fig. 3. (See P.120) At the beginning of experiment (time: 11:30), the curve for irradiance of the sun and sky versus the wavelength has a small value in shorter wavelength band with a peak at 510 nm. The downward irradiance decreases with time, namely with decrease in altitude of the sun. At the end of experiment (time: 15:15) , the downward irradiance at 510 nm decreased to be 30% of that at the start time, and the curve is almost similar to that at the beginning of experiment. Thus, it is natural to consider that this time-dependent change of the irradiance of the sun and sky should affect the radiance from the oil slick. Therefore, in the present analysis the radiance is expressed with the relative value by dividing the upward radiance by the downward irradiance (Lu/Ed ; Lu : upward radiance, Ed : downward irradiance).

Table 1 shows the dominant wavelength at each oil slick thickness from the chromaticity diagram. Here Lu/Ed is used. As the oil thickness increased from 0.0013 to 0.0019 mm, the dominant wavelength shifted from shorter to longer ones, and it moved back and stayed around the vicinity of 480 nm when the thickness is 0.0027–0.01 mm. When the thickness is 0.05–0.075mm, the complementary wavelength was 567 nm, and the thickness is 0.1–1.0 mm it again moved back to shorter wavelengths

Table 1. Dominant wavelength of each oil slick. Symbol indicates complementary wavelength.

Thickness of oil slick(mm)	Dominant wavelength(nm)
No slick	494.8
5.0×10^{-4}	489.0
1.3×10^{-3}	562.7
1.9×10^{-3}	570.0
2.7×10^{-3}	482.4
4.0×10^{-3}	485.5
5.0×10^{-3}	484.2
8.0×10^{-3}	482.0
1.0×10^{-2}	484.4
2.0×10^{-2}	539.4
3.0×10^{-2}	588.7
5.0×10^{-2}	*566.9
7.5×10^{-2}	*567.0
1.0×10^{-1}	457.9
1.5×10^{-1}	464.5
2.0×10^{-1}	466.0
5.0×10^{-1}	470.8
1.0×10^{0}	469.0

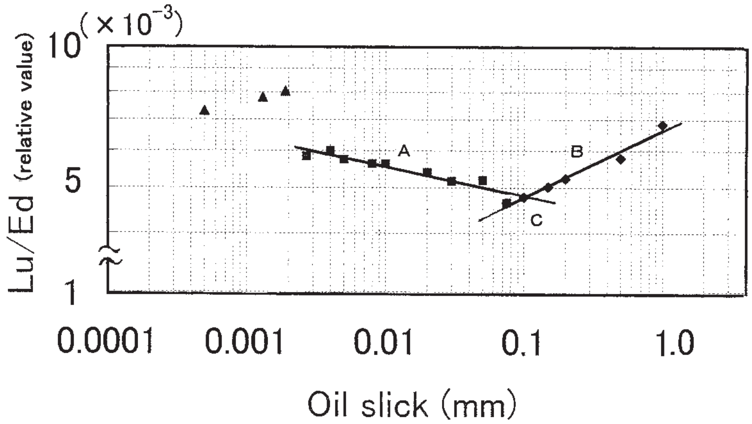

Fig. 4. Ratio of upward radiance and downward irradiance at 510nm versus oil slick thickness. A, B: regression lines. C: cross point.

around 460 nm. Thus the change of wavelength (water color) as a function of the oil slick thickness is not a monotonous increase. This result is similar to the relationship between the dominant wavelength of water color and the concentration of suspended solid (SS) and phytoplankton (Chl. *a*), as investigated by MORINAGA *et al.* (1992).

Fig. 4 shows a relationship between *Lu/Ed* at 510 nm and the oil slick thickness. The value of *Lu/Ed* without oil slick is 4.75×10^{-3}. When the oil slick thickness is very thin (0.0005–0.0019 mm), the *Lu/Ed* greatly increases from that without oil slick. As described previously in the visual observation of the color of water, this phenomenon is due to irregular reflection caused by the formation of heavy oil droplets (SATO, 1960). When the oil thickness is from 0.0027 to 0.075 mm, the *Lu/Ed* decreases with the increase of the slick thickness. The scattered light due to oil increases, but the scattered light from the water inside largely decreases with the increases of slick thickness. This is explained as follows: the transmitted light into water through the thick oil slick decreases, and the scattered light from the water inside also decreases because of the thick oil, resulting in an extreme decrease of the resultant scattered light. The phenomenon of the radiance decrease with the increase of the slick thickness is also observed at 412, 443, 490, and 555 nm with the exception at 665 nm. As to an exceptional wavelength, it can be considered that the rate of light attenuation by water itself is extremely high at longer wavelength band. MORINAGA and ARAKAWA (2000) investigated the effect of oil slick to the irradiance under the water. If we set the irradiance without oil slick at 100%, the irradiance measured at 510 nm attenuates to 37.5% at the slick thickness of 0.02 mm, showing a high attenuation rate. When the slick thickness is 0.075–1.0 mm, the *Lu/Ed* increases with the increase of the slick thickness. In this case, there is almost no scattered light from the water inside due to thick slick, and the increase of upward radiance is due to an increase of the scattered light from the oil slick. The upward radiance is considered to be the sum of the reflected light and the scattered light from the oil slick. The same trend of the radiance increase with the increase of the slick thickness is also observed at 412, 443, 490, and 555 nm.

Now we will discuss equations that are used for the calculation of the thickness of oil slick. In Fig. 4 two regression lines are drawn using the least squares method for the relationship between the oil slick thickness and the *Lu/Ed* for the above two cases. The first line and the second line is A and B, respectively, and the cross point is a critical point C. These results can be interpreted by the schematic diagrams for the optical phenomena (Fig. 5). The upward radiance without oil slick (L_{UW}) is the sum of reflected light from the water surface (L_{RW}), and scattered light from the water inside (L_W), and L_{RW} is much smaller than L_W (Fig. 5a). When there is oil slick, reflected light from the oil slick (L_{RO}) is a little larger than L_{RW} because the relative refractive index of oil is larger than that of water (Fig. 5b). Here, two cases can be considered depending upon the thickness of oil. When the oil slick is very thin (Fig. 5b (1)), the upward radiance (L_{UO}) is the sum of L_{RO} and scattered light from the water inside ($L_{W'}$) and that from the oil itself (L_O) including reflected light from the boundary of oil and water (Line A in Fig. 4). In this case $L_{W'}$ is larger than L_O. When the oil slick thickness increases, $L_{W'}$ becomes to be smaller than L_O, and eventually it becomes zero. When the oil slick thickness is rather larger (Fig. 5b(2)), the upward radiance ($L_{UO'}$) is the sum of L_{RO} and scattered light from the oil itself ($L_{O'}$) (Line B in Fig. 4). That is, when the oil slick thickness increases, only $L_{O'}$ increases, resulting in the increase of the upward radiance.

In Fig. 4 the critical point C is the point where $L_{W'}$ becomes zero. Thus, L_{UO} is equal to $L_{UO'}$. At the critical point C, the values of *Lu/Ed* as a function of wavelength are as follows: 5.57, 5.10, 4.78 and 4.45×10^{-3} at the wavelengths of 443, 490, 510, and 555 nm, respectively. The oil slick thickness at those points is 0.076, 0.097, 0.102, and 0.123 mm, respectively. That is to say, the position of critical point varies with the wavelength. When there is no oil slick, the values of *Lu/Ed* are 5.17, 4.85, 4.75, and 4.37 $\times 10^{-3}$ at each of the above wavelengths. Thus, at point C, L_{UO} is slightly larger than L_{UW}, and the difference between L_{UO} and L_{UW} varies with the wavelength. As mentioned above, this maybe is due to the difference between L_{RW} and L_{RO}. The difference is the smallest at 510 nm, and it corresponds to 0.6 % of the total value. Based on these, we obtain the following equations for the calculation of the film thickness using the *Lu/Ed* at 510 nm. The regression equation for line A (film thickness, less than 0.102 mm) is $Y = \alpha X^{-0.0637}$ and the one for line B (film thickness, more than 0.102 mm) is $Y = \beta X^{0.143}$ (Y: *Lu/Ed* (510), X : oil slick thickness, α, β: constant) . Here *Lu/Ed* (510) is ratio of upward radiance and downward irradiance at 510 nm of oil slick. The α and β are the constant values calculated from the ratio of upward radiance and downward irradiance at 510 nm of sea water at critical point.

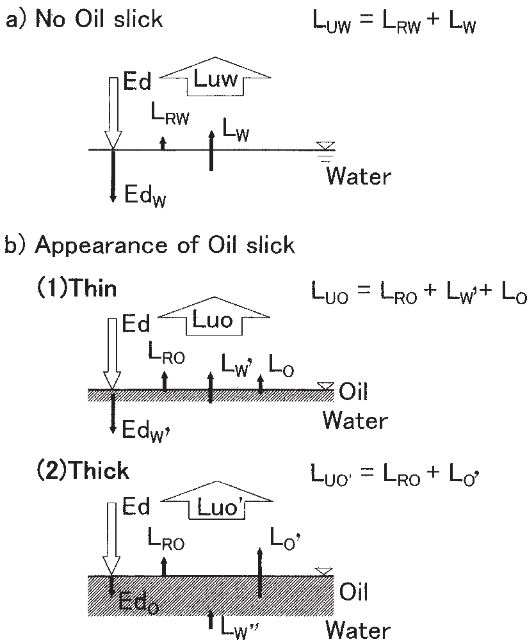

Fig. 5. Optical process by oil slick near the water surface. Ed: downward irradiance Edw, Edw': downward irradiance inside the water, Edo: downward irradiance inside the oil. Luw: upward radiance without oil slick. LRW: reflected light from the water surface. Lw: scattered light from the water inside. Luo: upward radiance through the oil. LRO: reflected light from the oil surface. Lw': scattered light from the water inside. Lo: scattered light from the oil itself. Luo': upward radiance through the oil. Lo': scattered light from the oil itself. Lw'': scattered light from the water under the oil.

When the *Lu/Ed* data of *in situ* observations is used for the ordinate, two values can be obtained for oil slick thickness (abscissa) from line A and from line B (Fig. 4). In order to solve this contradiction, the relationship between the ratio of *Lu/Ed* at two different wavelengths and the oil slick thickness is plotted in Fig. 6. The combinations of *Lu/Ed* at two wavelengths that give a large difference with respect to the thickness are *Lu/Ed* (412) / *Lu/Ed* (510) and *Lu/Ed* (443) / *Lu/Ed* (510) . For example the ratio *Lu/Ed* (412) / *Lu/Ed* (510) is 1.4 if the thickness is more than 0.1

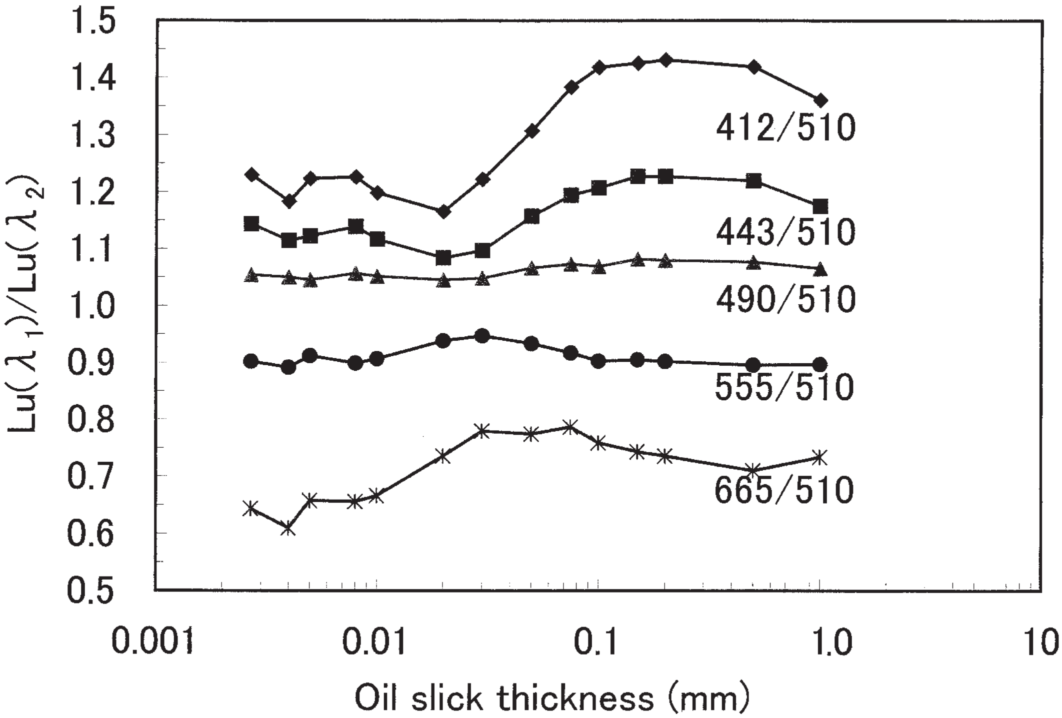

Fig. 6. Relationships between ratio of upward radiance at two wavelengths and oil slick thickness.

mm and it is less than 1.2 if the thickness is less than 0.02 mm, showing a great difference. Therefore, the applicable regression line can be determined easily if we take the ratio of the upward radiance and downward irradiance at two wavelengths, 412 and 510 nm.

3-3. Oil slick thickness for leaked heavy oil in the Sea of Japan

One of the pictures of *in situ* oil slick in September 1997 is shown in Fig. 7. This leaked oil was from the bottom of the sea, which was approximately 2500 m deep. The elapsed time after the leak from the boat hold was unknown. In the visual observation, the width of major oil slick was 300 m and the length was 1.2 km (HAMADA and AOYAMA, 1999). The oil slick was shiny, and it was obvious that the radiance in the region was much higher than that of the surrounding water.

Water temperature and salinity of the surface layer for surveyed sea area are 25.1–25.3 ℃ and 32.3–32.6 psu, respectively. Both the water temperature and salinity do not change down to the depth of 30m, indicating a mixed layer. A permanent thermocline is observed from the depth of 30m to approximately 300 m. The temperature decreases gradually below thermocline, reaching 0.5 ℃ at the depth of 500 m. On the other hand, the salinity increases gradually and reaches 34.4 psu at the depth of 100 m, and there is almost no change below that (MAEDA *et al.*, 1999).

The observed values for the oil slick are tabulated in Table 2. The upward radiance is divided by the downward irradiance and the relative values are used. Since the values of *Lu/Ed* (412)/*Lu/Ed* (510) are in the range of 1.01–1.12 and they are smaller than 1.4, the equation $Y = \alpha X^{-0.0637}$ is used for the calculation. Here Y is *Lu/Ed* (510) of oil slick and X is oil slick thickness (mm) . The value of α is calculated using that Y is 0.0192 and X is 0.102 in 1997, and that Y is 0.0100 and X is 0.102 in 1998. It becomes to be 1.66×10^{-2} in 1997, and to be 8.64×10^{-3} in 1998. Based these values, the each thickness of oil slick a, b, c, d and e is estimated to be 9.2×10^{-5}, 2.2×10^{-4}, 1.4×10^{-2}, 1.7×10^{-2}

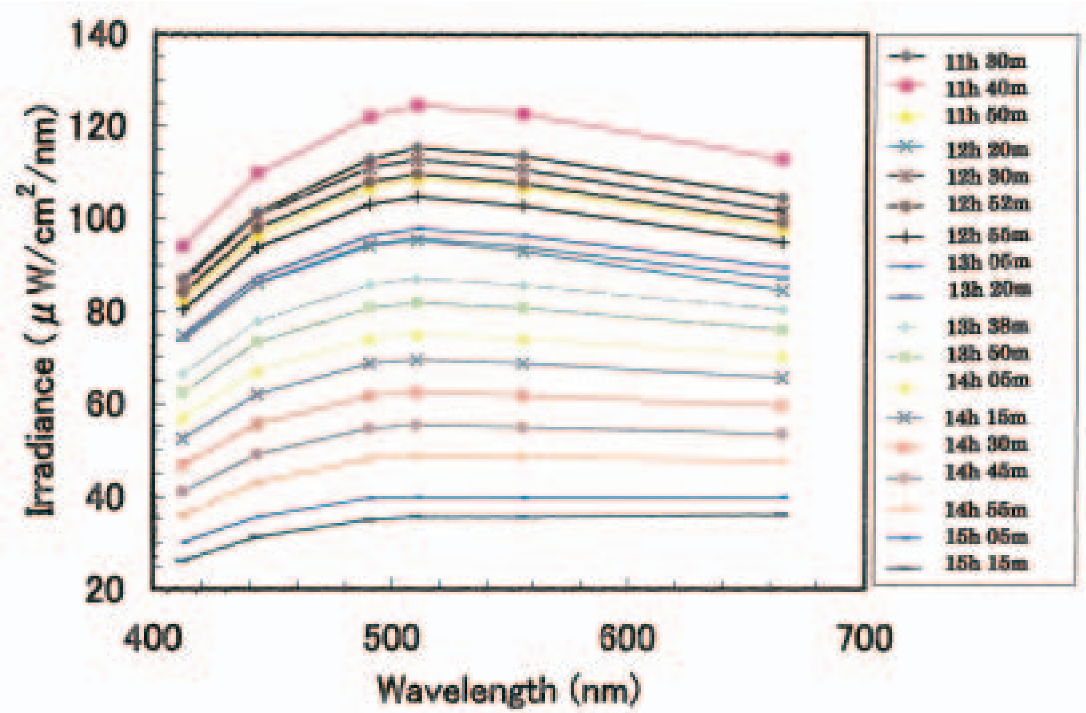

Fig. 3. Distribution of downward irradiances of the sun and sky. The numerals outside the figure indicate the time of each measurement.

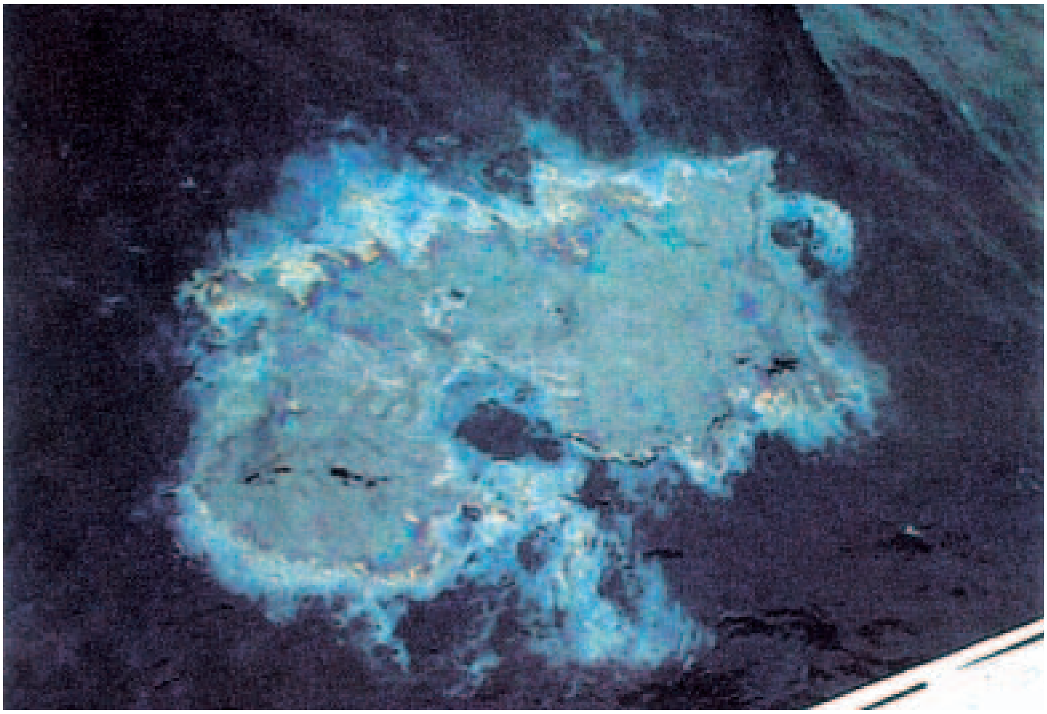

Fig. 7. Picture of *in situ* oil slick observed at September 11 in 1997. This was photographed on board by Prof. Hamada Yoshiyuki of Tokyo University of Fisheries.

Table 2. *In situ* measurement data for the oil slick in 1997 and 1998. *Lu*: upward radiance of oil slick. *Ed*: downward irradiance of oil slick. *Lu**: upward radiance of sea water. *Ed**: downward irradiance of sea water 412, 510: wavelength (nm)

Observation Month&Year	Site	Lu/Ed		Lu/Ed(412) / Lu/Ed(510)	Lu*/Ed*(510)
		412nm	510nm		
Sep.in 1997	a	0.0308	0.0300	1.02	0.0192
	b	0.0294	0.0284	1.03	0.0192
	c	0.0232	0.0218	1.06	0.0192
Sep.in 1998	d	0.0126	0.0112	1.12	0.0100
	e	0.0220	0.0216	1.01	0.0100

and 5.7×10^{-7} mm, respectively. However, the thickness values that are in the range of experiment are the oil slick c of 1997 and the oil slick d of 1998. Accordingly, they are estimated 0.014 and 0.017 mm, respectively. The estimate for the oil slick b of 1997 is considered to be accurate judging from the visual observation. Other thickness values are extremely thin. It is unknown if this is due to unique dispersion by the long term wave effect or due to a change of oil properties after a long time of the leak from the boat hold. If the properties of oil are different, the above correlation equations may not be applicable. This will be investigated in the future.

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