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Modelling the influence of oil content on optical properties of seawater in the Baltic Sea

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The accuracy and correct interpretation of optical parameters of seawater depend on the complete information osn the interactions between seawater components and the light field. Among components influencing the radiative transfer, the droplets of oil can cause over- or underestimation of modelled and measured optical quantities, especially in closed seas and coastal zones. Oil content in the Baltic Sea varies from several ppb in the open sea to several ppm in estuaries or ship routes. Oil droplets become additional absorbents and attenuators in seawater causing changes in apparent optical properties. These changes can potentially enable remote optical detection of oil-in-water emulsion in visible bands. To demonstrate potential possibilities of such optical remote sensing, a study of inherent optical properties of two types of crude oil emulsion was conducted, i.e. high absorptive and strongly scattering *Romashkino*, and low absorptive and weakly scattering *Petrobaltic*. First, the calculations of spectral absorption and scattering coefficients as well as scattering phase functions for oil emulsions were performed on the basis of Lorentz-Mie theory for two different oil droplets size distributions corresponding to a fresh and 14-days aged emulsions. Next, radiative transfer theory was applied to evaluate the contribution of oil emulsion to remote sensing reflectance $R_{rs}(\lambda)$. Presented system for radiative transfer simulation is based on Monte Carlo code and it involves optical tracing of virtual photons. The model was validated by comparison of $R_{rs}(\lambda)$ simulated for natural seawater to $R_{rs}(\lambda)$ from in situ measurements in Baltic Sea. The deviation did not exceed 10% for central visible wavelengths and stayed within 5% for short and long wavelengths. The light *Petrobaltic* crude oil in concentration of 1 ppm causes typically a 10-30% increase of R_{rs} while the heavy *Romashkino* reduces R_{rs} for 30-50%.

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1 INTRODUCTION

Optical properties of water bodies carry important information about the composition, dynamics and processes in seawater. This information is widely used to derive ocean parameters for climate study and environmental management purposes. Currently, satellite ocean colour data are used to monitor and understand climate change on global and local scales, to forecast and observe natural phenomena of societal consequence, such as harmful algal blooms, hurricanes or sand storm), and to gather useful information for offshore and coastal zone management [3, 8].

Satellite ocean colour data are verified and validated by referencing *in situ* data. Spectral remote sensing reflectance, $R_{rs}(\lambda)$, determined from top-of-atmosphere radiance after atmospheric correction, is the primary data product used for the generation of higher level products (such as chlorophyll-a concentration). As a consequence, access to accurate in situ $R_{rs}(\lambda)$ is essential for the assessment of primary data products from satellite ocean colour missions [19, 20]. To meet the growing requirements of accurate satellite measurements (approaching 2% uncertainty) it is necessary to apply local methods and models that account for all seawater constituents, such as mineral particles [18], coloured dissolved organic matter (CDOM) [6], micro-bubbles [21], or oil droplets [12].

Radiative transfer in seawater is the physical basis for quantitative ocean optics including ocean colour remote sensing. The conservation of energy in the interaction of light and water is described by the radiative transfer equation (RTE). The time-independent form of RTE for horizontally homogenous water, widely used in oceanography, is expressed by the following formula [10]:

$$\cos\theta \frac{dL(z,\theta,\varphi,\lambda)}{dz} = -c(z,\lambda)L(z,\theta,\varphi,\lambda) + \int_{4\pi} L(z,\theta',\varphi',\lambda)\beta(z,\psi,\lambda)d\Omega' + S(z,\theta,\varphi,\lambda) \quad (1)$$

where:
$L(z,\theta,\varphi,\lambda)$ — radiance as a function of depth z in the sea, zenith angle θ, azimuth angle φ, and wavelength λ,
$c(z,\lambda)$ — total light beam attenuation coefficient equal to the sum of absorption coefficient and scattering coefficient: $c(z,\lambda) = a(z,\lambda) + b(z,\lambda)$,
$\beta(z,\psi,\lambda)$ — volume scattering function (describing angular

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distribution of scattering process),
ψ — scattering angle between the direction of incident light (θ, φ) and the direction of scattered light (θ', φ'),
$S(z,\theta, \varphi, \lambda)$ — source function describing emission and inelastic scattering into the beam (such as fluorescence or bioluminescence).

Oil droplets become additional absorbents and attenuators in many water regions as a consequence of increasing shipping activities and oil transportation. Their presence can considerably affect optical properties of near-surface water, especially in the the areas more exposed to pollution, like coastal zones, estuaries, marine transportation routes or oil fields [15, 16]. The main types of oil pollution in seawater are crude oils, lubricate oils and fuel oils, flowing in with the rivers or coming from ship routine activities and discharges. While a small fraction of oil from these sources dissolves within seawater (0.2 – 0.7%), most persists as dispersed droplets (oil-in-water emulsion). Oil emulsion amounts to over 80% of the total oil pollution in Baltic Sea estimated to be 76 thousand tons per year (HELCOM, 1993). According to the MARPOL convention, ship discharge waters may legally contain up to 15 ppm of oil. In some regions the limits are more restrictive, e.g. up to 5 ppm in Canadian inland waters.

There is a need of gathering comprehensive datasets containing all the information necessary for a complete radiative transfer calculation, which is more important in optically complex waters. Inherent optical properties (IOPs) such as absorption $a(\lambda)$, scattering $b(\lambda)$, and attenuation $c(\lambda)$ are essential inputs for radiative transfer models for computing light fields in seawater [11, 22]. However, the quantitative use of these data requires taking into account all local seawater constituents. Currently there is insufficient data and no applied methods to estimate the influence of dispersed oil on the upwelling light field of seawater [14]. The presence of oil-in-water emulsion is usually not taken into consideration.

2 METHOD

Numerical radiative transfer simulations are used to predict the upwelling light stream using given seawater inherent optical properties. Light propagation is computed for specified conditions and allows evaluating the influence of each factor on remote sensing reflectance separately [9]. The above mentioned inherent optical properties of oil-in-water emulsions have been implemented into a system of radiative transfer simulation based on Monte Carlo code in order to estimate their influence on remote sensing reflectance [4]. Monte Carlo methods, developed during the second world war, are now widely used for radiative transfer modelling purposes, particularly to solve the time-dependent radiative transfer equation or to address 3D problems, such as modelling the influence of sensor geometry on measured parameters, corrections for self-shadowing of sensors and measurement platforms, or analysis of light propagation in turbulent media [2]. Monte Carlo methods effectively account for multiple scattering by mathematically following many photon packets until they contain a negligible amount of energy. The Monte Carlo code involves optical tracing of photons within a given solid sector of upper hemisphere, on the basis of probability of visible light absorption and scattering by seawater constituents, including oil droplets. It allows conducting single-wavelength simulations limited to the wavelengths of which the seawater IOPs are known. It does not include inelastic scattering.

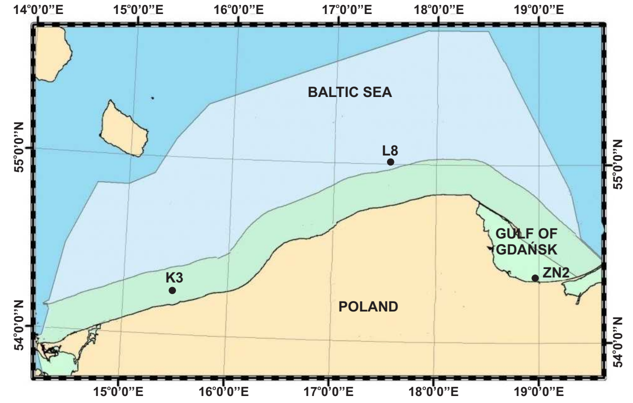

FIG. 1 The map of *in situ* measurement stations in the Baltic Sea.

Oil dispersed in seawater forms spherical droplets and bulk light scattered from droplet populations has been accurately modelled with single-scatter formulations of the RTE [1, 12]. Lorentz-Mie theory allows computing the IOPs of oil emulsion on the basis of complex spectral refractive index of light in particles matter and particle droplets size distribution.

Radiative transfer simulations were conducted using *in-situ* input data from the region of Southern Baltic Sea in order to address presented method to the natural environment. Baltic Sea is a „special area" under MARPOL Convention, in which, according to the International Maritime Organization, for technical reasons relating to their oceanographical and ecological condition and to their sea traffic, the adoption of special mandatory methods for the prevention of sea pollution is required. For this study, three stations with different optical properties were chosen (Figure 1): in the Gulf of Gdansk (ZN2), in open Baltic Sea (L8), and in the estuary (K3). Measurements of light absorption and attenuation were conducted in April and May 2012 using in-situ spectrophotometer AC-9 (WET Labs Inc.) onboard R/V Oceania, owed by the Institute of Oceanology of Polish Academy of Sciences.

2.1 Model description

A layered seawater model formulated into Monte Carlo code was created in order to discretize continuous values of IOPs for the purpose of conducting radiative transfer simulations (Figure 2). The depth of each layer was chosen separately for every *in situ* station considering the depth-dependence of absorption and scattering coefficients (see Appendix). Only the surface layer (usually 0-5 m) was artificially polluted by *Petrobaltic* and *Romashkino* crude oil emulsions in two different droplet size distributions and in concentrations of 1-10 ppm.

The boundary conditions are: (i) downwelling irradiance calculated from the incident solar light zenith angle (modelling the sun height), the coefficient of sky diffusion and given number of incident photons; (ii) sea surface optical characteristics described by Fresnel formulas and statistical distribution of wave slopes, parameterized by the wind speed (Cox and Munk distribution) [23]; (iii) sea bottom reflectance coefficient. Simulations were conducted for the sea surface characterized by a typical wind speed of 5 m/s, clear sky conditions (70% direct sun irradiance, 30% diffusive sky irradiance), and a lambertian seafloor typical for the Baltic Sea sand (2% reflected light, 8% diffused light).

The model output data is the remote sensing reflectance R_{rs} calculated as the ratio of the water-leaving radiance Lw to the downward sky irradiance E_d:

$$R_{rs} = \frac{L_w}{E_d} \tag{2}$$

L_w was calculated within the half angle of 7^o, what corresponds to the field of view of the radiometer used for validation of the results (Ramses radiometer, Trios GmbH).

2.2 Input data for radiative transfer model

The input data for the radiative transfer simulations are the inherent optical properties of all seawater components and the boundary conditions. The optical model of water body consists of three elements:

1. Pure water —- with spectral absorption coefficient given by Pope and Fry (1997) and spectral scattering coefficient given by Smith and Baker (1981).
2. Common natural components of seawater — particles and dissolved organic matter described by total spectral absorption and attenuation coefficients measured in the Baltic Sea and scattering phase functions adapted from Petzold (1972) and measured locally in the Baltic Sea [9, 7, 5].
3. Oil droplets of two types of crude oil —- with optical characteristics calculated on the basis of Lorentz-Mie theory (Figure 3). The input data used in calculations consist of the complex refractive index of light and the size distribution of oil droplets. The relative complex refractive index of light m_{rel} for oil droplets suspended in water is defined as follows:

$$m_{rel} = n' - ik = \frac{n_c}{n_w} - ia\frac{\lambda_0}{4\pi n_w} \tag{3}$$

where:
n_c, n_w — real refractive indexes of crude oil and seawater respectively,
a — absorption coefficient of oil,
λ_0 — wavelenght of light in vacuum.

The real part of the refractive index n' describes the change of the light speed after passing through the border of two media. It impacts the computed oil volume scattering function. The imaginary part k contains information about the absorption of light in oil. Spectral variability of both, the real and imaginary parts of complex refractive indexes of considered crude oils are presented in the Figure 3.

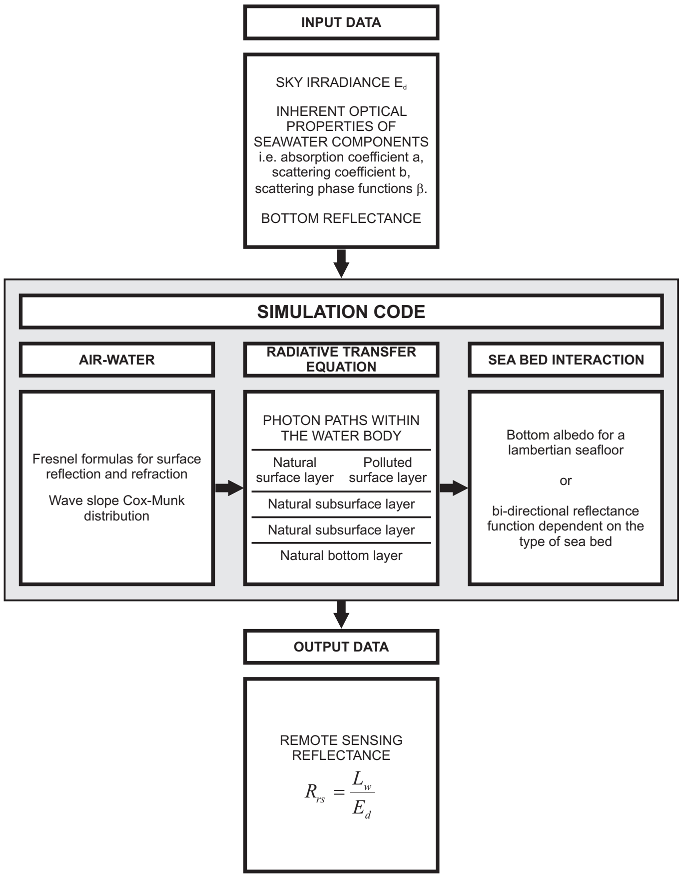

FIG. 2 General scheme of the optical model of seawater framed into Monte Carlo radiative transfer simulation code.

It was determined, that size distribution can be described by a log-normal size distribution function [17]:

$$f(r) = A\,exp\left(-\frac{ln^2\frac{r}{r_0}}{2\sigma^2}\right) \tag{4}$$

where:
A — quantity parameter,
r_0 — radius of the size distribution peak,
σ — parameter of the shape of size distribution.

Two sets of parameters were used in the calculations: r_0 = 0.25 µm with parameter σ = 1.02 (corresponding to a freshly homogenized emulsion) as well as r_0 = 0.08 µm with σ = 0.87 (corresponding to the emulsion after 14 days of ageing). The results of calculations of spectral IOPs of oil-in-water emulsions are shown in the Figure 4 and Figure 5.

The influence of oil droplets on the seawater IOPs depends not only on the type and concentration of oil, but also on the degree of droplet break-up (see Figure 6). In this study we present two types of crude oil transported through the Baltic Sea characterized by different optical properties. The transparent *Petrobaltic* represents light crude oils and it is extracted offshore in the Southern Baltic region. The real part of it's refractive index varies spectrally from 1.47 to 1.49 in the visible bands and slightly depends on temperature [15]. The imaginary parts is highly variable (10^{-5} – 10^{-2}) and implies strong variability in absorption. The opaque *Romashkino* heavy crude oil, extracted onshore in the Volga-Ural basin in Russia, also represents the pollution from river inflows. The real part of

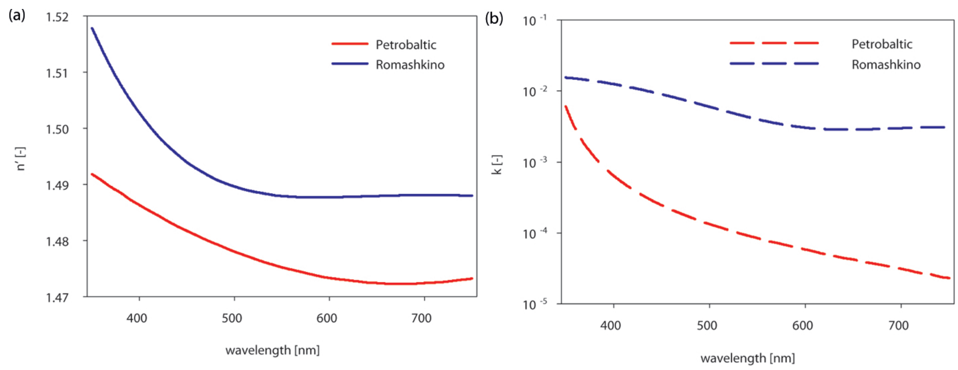

FIG. 3 The spectra of real part n' and imaginary part k of the complex refractive index of light of *Petrobaltic* and *Romashkino* crude oils.

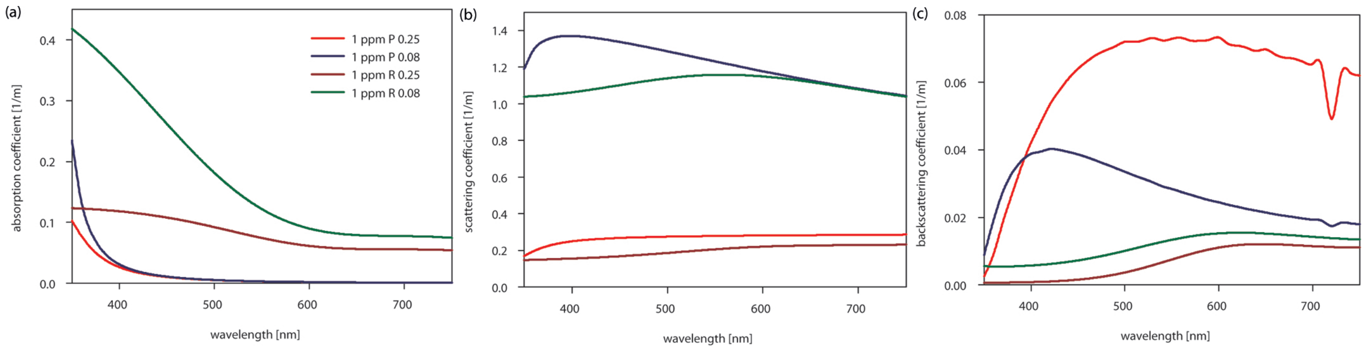

FIG. 4 Spectral absorption, total scattering and backscattering coefficients of oil-in-water emulsions *Petrobaltic* (P) and *Romashkino* (R) in concentration of 1 ppm calculated on the basis of Lorenz-Mie theory for two different droplets size distributions marked by the value of radius of the size distribution peak r_0 (in µm).

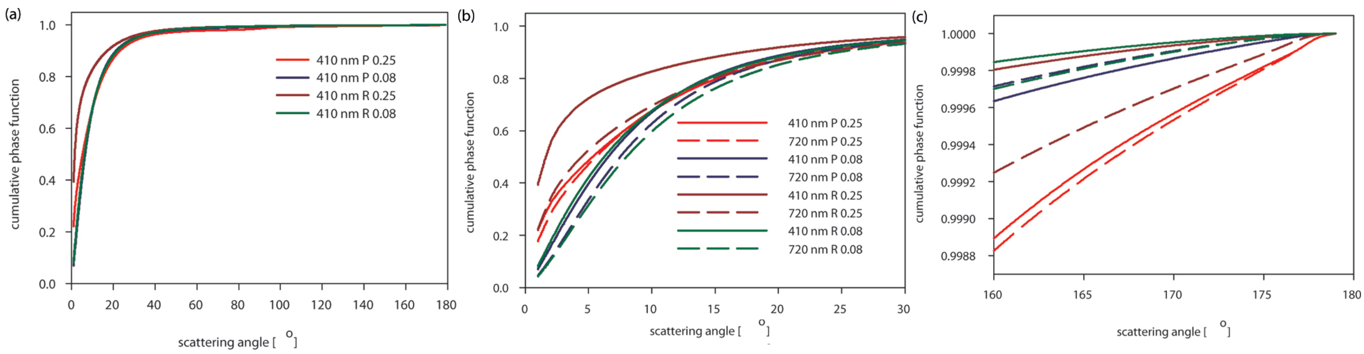

FIG. 5 Phase functions of oil-in-water emulsions in the cumulative form. Left graph shows the variability with the type of oil and droplets size distribution. Central and right graphs show the wavelength dependence for small and large scattering angles respectively.

it's refractive index is higher and varies spectrally from 1.49 to 1.52 in the visible bands. The imaginary part is less variable ($\sim 10^{-2}$) and results in higher absorption. The study shows that *Petrobaltic* droplets in concentration of 1 ppm slightly increase seawater absorption (for less than 3% in the visible bands), and more significantly increase the total scattering coefficient. The impact on scattering coefficient depends on the real part of the complex refractive index (i. e. the type of oil) and size distribution of oil droplets. It varies from almost 50% for larger droplet radii (r_0 = 0.25 µm) to quadrupled values for small radii (r_0 = 0.08 µm). *Romashkino* droplets in the same concentration increase the total absorption coefficient from over 30% (large droplets) to about 65% (small droplets), and give respectively over 30% contribution to the scattering coefficient from large droplets and a 3-4 fold contribution from small droplets. The impact on absorption reaches maximum in the central part of the visible light, and the impact on scattering slightly increases with wavelength.

3 RESULTS AND DISCUSSION

Model accuracy was preliminarily evaluated by comparison of in situ $R_{rs}(\lambda)$ with simulated $R_{rs}(\lambda)$ for natural seawater. The best fit (up to 5% of percentage deviation) was achieved for the wavelengths of 410, 488 and 510 nm. Model accuracy is strongly affected by the choice of scattering phase function (see Figure 7), whereas small modification of the boundary conditions seems to be insignificant.

Results of modelling the influence of oil content on $R_{rs}(\lambda)$ show the magnitude of impact of several factors: the type of oil product, i.e. the IOPs of oil-in-water emulsion (Figure 8), the concentration of dispersed oil (Figure 9), the size distribution of oil droplets (Figure 10) as well as the optical properties of natural components of seawater (Figure 8). Fresh *Petrobaltic* emulsion (larger droplets) cause in average a 30% increase of R_{rs} by the concentration of 1 ppm. The effect grows propor-

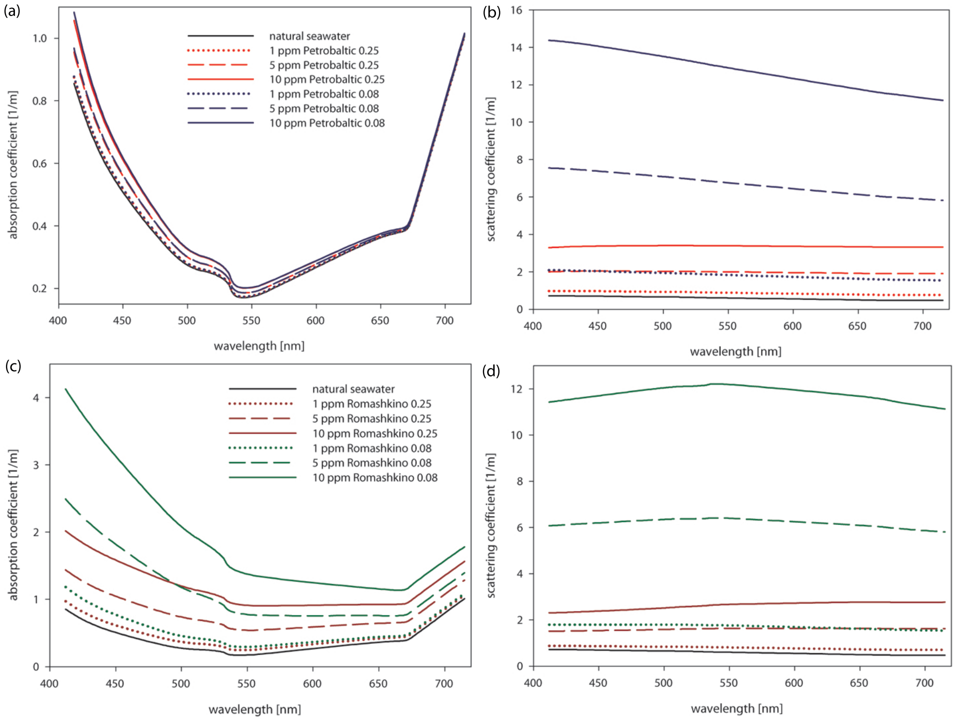

FIG. 6 Spectral absorption and scattering coefficients for the surface layer of natural seawater at the station L8 (black line) and seawater artificially polluted by *Petrobaltic* and *Romashkino* droplets (colour lines).

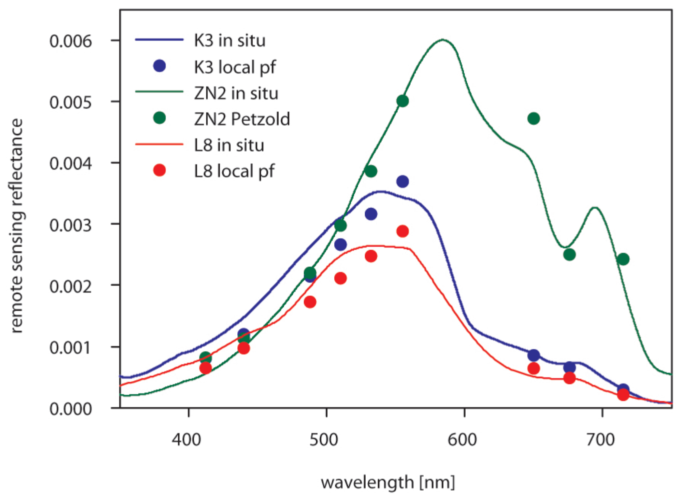

FIG. 7 Comparison between $R_{rs}(\lambda)$ measured *in situ* (solid lines) and modelled (dots) using different scattering phase functions.

tionally with concentration. However, the same concentration of 14 days aged emulsion (smaller droplets) gives a doubled 60% increase of R_{rs} which grows more rapidly with concentration, reaching 8-fold by 5 ppm and 20-fold by 10 ppm. Remote sensing reflectance increases with the backscatter ratio and decreases with the increase of absorption coefficient. Therefore, in case of *Petrobaltic* crude oil, mainly its scattering properties influence R_{rs}.

The influence of *Petrobaltic* emulsion is more noticeable in barely absorptive seawater (e.g. station K3), while *Romashkino* causes more significant drop of R_{rs} in weakly scattering environment (e.g. station ZN2).

In the contrary, the influence of *Romashkino* emulsion is more complex: we observe a reduction of R_{rs} in the range of 400-600 nm (15-20% for 1 ppm) and a gain for longer waves (10-15% for 1 ppm). The effects increase slightly in both directions with oil concentration and more remarkably with the degree of droplets break-up, showing the combination of absorption and backscattering influence. The reduction of R_{rs} in the short-wave part of spectrum is caused by the high absorption coefficient, but for the longer waves the contribution of backscattered light becomes dominant.

4 CONCLUSION

The remote sensing reflectance from seawater, which is the input of algorithms for deriving many ocean parameters from remote sensing methods (including satellite remote sensing) can be influenced by dispersed oil in the near-surface water column. Interpretation of reflectance spectra requires a simultaneous multi-parameter analysis of light propagation in seawater. The influence of most parameters on $R_{rs}(\lambda)$ is nonlinear and often superimposed by other factors. However, it can be studied separately in terms of numerical radiative transfer simulations. The contribution of different types of oil to $R_{rs}(\lambda)$ can be theoretically estimated by including optical properties of oil droplets to radiative transfer modelling. The presented study connects radiative transfer process in seawater with the inherent optical properties of dispersed oil. It creates the tools to evaluate the potential contribution of oil content to the upwelling light measured with remote sensing methods.

In general, the presence of high-absorptive and low-backscattering oil droplets is easily observed on any marine water background, as they cause a significant decrease of R_{rs}. Those features usually imply opaque oils with large-

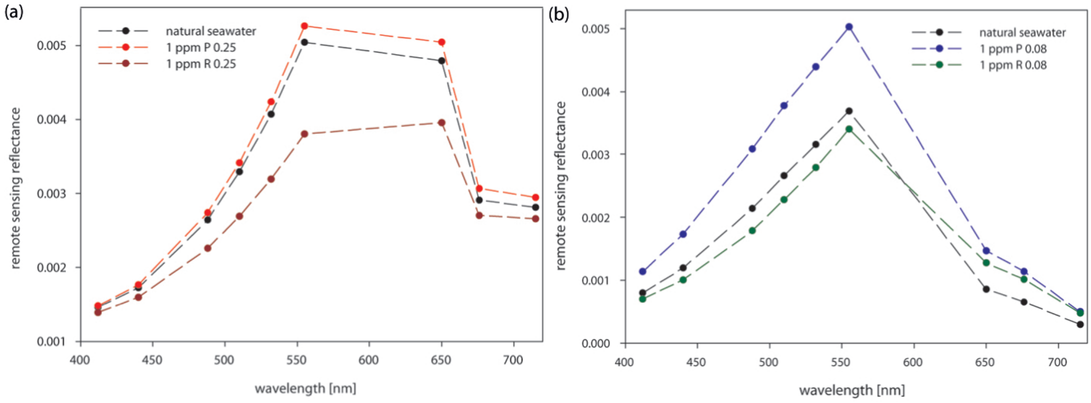

FIG. 8 Modelling the effect of the type of oil on $R_{rs}(\lambda)$. Input data for simulations are taken from stations ZN2 (left graph) and K3 (right graph).

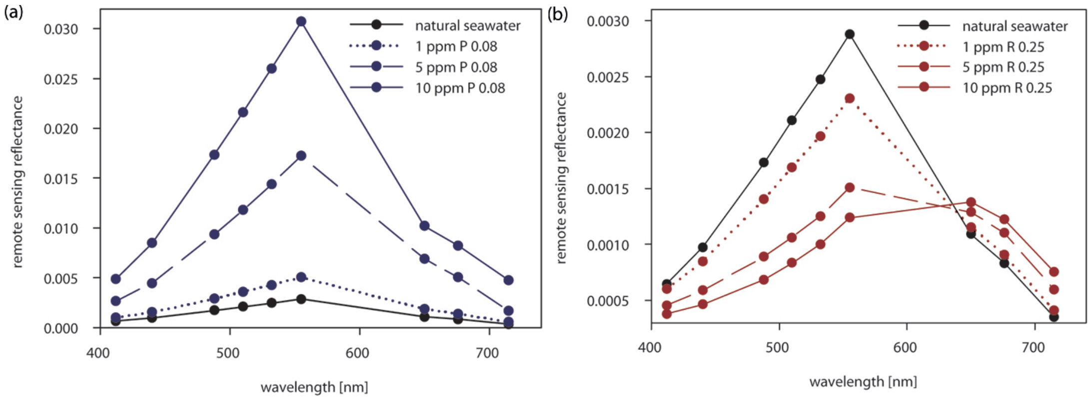

FIG. 9 The effect of the concentration of oil on R_{rs}. Input data for simulations are taken from station L8.

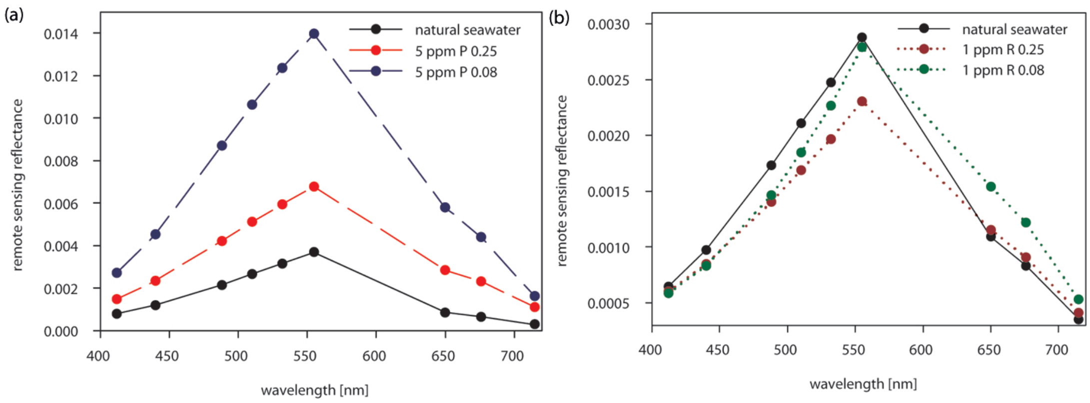

FIG. 10 The effect of the size distribution of oil droplets on $R_{rs}(\lambda)$. Input data for simulations are taken from stations K3 (left graph) and L8 (right graph).

sized droplets, which scatter light more efficiently into near-forward directions. On the other hand, high backscatter fraction (usually observed for small-sized particles) as well as low absorption coefficient (characteristic for transparent oils) increase R_{rs}. In case of oils with combined optical properties, e.g. high-absorptive and high-backscattering, the impact on R_{rs} becomes more complex and more concentration-dependent, but is still possible to evaluate.

Quantitative radiative transfer computations can contribute to the improvement of satellite algorithms accuracy for the determination of parameters derived from the water-leaving radiance, regarding coastal zones, estuaries, main marine transportation routes and oil extraction areas. It can also improve the accuracy of shipboard and offshore R_{rs} measurements, which are used for the purpose of calibration of satellite data and for seawater monitoring. It is also possible that it will enable the remote detection of oil-in-water emulsion. As a next step, environmental measurements of the natural sea water polluted by oil emulsion will be performed in order to confirm modelled results.

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APPENDIX

wave length	ABSORPTION COEFFICIENT				SCATTERING COEFFICIENT			
	Station K3							
[nm]	I layer 0–2 m	II layer 2–4 m	III layer 4–7 m	IV layer 7–10 m	I layer 0–2 m	II layer 2–4 m	III layer 4–7 m	IV layer 7–10 m
412	0.8847	0.8490	0.7593	0.6900	0.9180	0.7985	0.5543	0.2913
440	0.5883	0.5625	0.4893	0.4290	0.8967	0.7785	0.5487	0.2897
488	0.3153	0.2995	0.2540	0.2137	0.8550	0.7440	0.5390	0.2857
510	0.2317	0.2210	0.1883	0.1580	0.8410	0.7400	0.5410	0.2877
532	0.1740	0.1655	0.1403	0.1190	0.8107	0.7140	0.5267	0.2810
555	0.1237	0.1170	0.0977	0.0817	0.7783	0.6870	0.5120	0.2760
650	0.0357	0.0330	0.0243	0.0177	0.6653	0.5875	0.4577	0.2560
676	0.0377	0.0335	0.0240	0.0123	0.6373	0.5635	0.4407	0.2470
715	0.0000	0.0000	0.0000	0.0000	0.6340	0.5615	0.4467	0.2543
	Station L8							
[nm]	I layer 0–5 m	II layer 5–12 m	III layer 12–16 m	IV layer 16–24 m	I layer 0–5 m	II layer 5–12 m	III layer 12–16 m	IV layer 16–24 m
412	0.8373	0.8390	0.7198	0.6230	0.7145	0.7171	0.4408	0.2301
440	0.5578	0.5600	0.4653	0.3843	0.6992	0.7026	0.4360	0.2319
488	0.2983	0.2987	0.2400	0.1883	0.6580	0.6633	0.4245	0.2301
510	0.2202	0.2197	0.1765	0.1388	0.6462	0.6511	0.4230	0.2328
532	0.1642	0.1630	0.1325	0.1048	0.6187	0.6237	0.4078	0.2278
555	0.1135	0.1126	0.0910	0.0715	0.5930	0.5987	0.3963	0.2270
650	0.0305	0.0304	0.0220	0.0154	0.4967	0.5051	0.3508	0.2135
676	0.0282	0.0290	0.0213	0.0105	0.4762	0.4843	0.3378	0.2050
715	0.0000	0.0000	0.0000	0.0000	0.4683	0.4773	0.3433	0.2134
	Station ZN2							
[nm]	I layer 0–4 m	II layer 4–11 m	III layer 11–22		I layer 0–4 m	II layer 4–11 m	III layer 11–22	
412	2.556	1.139	1.726		3.539	1.445	8.219	
440	1.836	0.744	1.175		3.431	1.388	8.116	
488	1.025	0.383	0.645		3.529	1.362	7.816	
510	0.783	0.278	0.480		3.594	1.368	7.673	
532	0.604	0.210	0.356		3.614	1.334	7.504	
555	0.461	0.149	0.246		3.634	1.320	7.321	
650	0.213	0.064	0.099		3.575	1.211	6.546	
676	0.446	0.109	0.156		3.312	1.187	6.342	
715	0.000	0.000	0.000		3.707	1.225	6.193	

TABLE 1 Table of the in situ data collected at the field stations: K3, L8 and ZN2 using AC9 Wetlabs spectrophotometer (calibrated to pure water).

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