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Dispersed methane flux to the water column from natural gas bubble streams at the Black Sea shelf

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Gas bubble streams are detected in the water column by the presence of strong, flare shaped backscatter signals recorded during hydroacoustic single beam echosounder surveys (flares). Some of these flares even reach the sea surface. In motion bubbles get into an evolutionary process caused by a variety of effects, including gaseous exchange with surrounding water. Simplistically, kinetics of such a gas exchange can be described by the Fick's law; the direction of the transfer of any given gas through the bubble depends on partial pressure of respective the gas in bubble, Henry's law constants and the concentration of dissolved gas in the water. In general, methane gradually dissolves during the lifetime of a bubble, while other gases enter the bubble. Consequently, bubbles cause a vertical transfer of methane from the sea bottom to upper water layers and can be considered as sources of dispersed methane flux to the water column. In present work an attempt is made to trace the methane gas phase exhaustion in flares trough the water column at the Black Sea shelf.

Our approach is based upon acoustic observations and measurements carried out in 2003 and 2004 with the scientific echosounder EK-500 onboard RV Vodianitskiy as part of the EU funded project CRIMEA. For the estimation of bubble size distributions our data from direct measurements of acoustic cross-section of single bubbles were used. Data for the relation between rising speed and shrinking rate vs. bubble size were obtained by tracking of single bubbles. Modelling was used to evaluate features

of the gas transfer process induced by rising bubbles. Having initial bubble size, gas composition and water depth as starting conditions the model produces series of time based values of bubble size, gas composition, rising depth and rising speed. Acoustic observations were utilized to verify the chosen model parameters.

For seeps detected at 90 - 95 m water depth hydroacoustically measured bubble sizes ranged from 1.3 to 11.3 mm in diameter. This bubble size range was confirmed by visual observations during video and submersible inspections. We assumed a gas content of 99% methane and small amounts of nitrogen and oxygen as initial gas composition according to geochemical analyses of gas bubbles sampled by submersible just above the sea floor. To determine the entire free methane flux from the sea floor into the water column and maybe into the atmosphere we run our model for several bubble sizes classes. Then simulation data were summed up with weighting coefficients according to the respective amount of bubbles per class. As a result, vertical profiles of molar content (mkmol) and methane flux (mkmol/s m) per average statistic bubble vs. depth were obtained. To get methane flux from the whole seepage simple multiplication is required by average statistic initial number of bubbles above the bottom per unit height. Depending on the spatial extension of the seep area, point or volume backscattering methods were used to quantify the bubble amount. Of great importance for both methods is the averaging of a high amount of data in space and time. We detected an average of 400 bubbles at high intensity seep sites within a water volume of 1m thickness above the bottom. The hydroacoustically determined amount of bubbles is again in very good agreement with direct visual observations. With an average initial rising speed of 0.25 m/s, 400 bubbles escaping from the sea floor cause a methane flux of 3.45 mmol/s using average bubble methane content of 34.5 mkmol ($400 \times 0.25 \times 34.5 = 3.45 \text{ mmol/s}$). As final methane content of average bubble at the sea surface is 6.5 mkmol, only $6.5/34.5 \times 100 = 18.8 \%$ of methane can reach the atmosphere due to the methane flux into the water column on the way up to the sea surface.