

Size distributions of bubbles and aerosols

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1. INTRODUCTION

The role of bubbles in air-sea gas transfer has been described in several publications (e.g., Thorpe [1982], Merlivat and Memery [1983], Woolf [1993]) and experimental evidence has been presented [Lamarre and Melville, 1991; Wallace and Wirick, 1992; Farmer et al., 1993]. To better understand the physics of air-sea exchange, bubble size distributions were measured by the TNO Physics and Electronics Laboratory (TNO-FEL) during ASGASEX, in combination with fluxes of CO₂ and profiles of aerosol size distributions. Bubble plumes were measured with sonar techniques by UCG and SUDO, as described elsewhere in this report.

The aerosol size distribution profiles were measured together with bubble size distributions to derive a quantitative source function for sea spray aerosol as function of meteorological and oceanographic parameters. The bubble-mediated production of sea spray aerosol has been studied for many years (e.g., Blanchard [1989]). New techniques are being explored to study the bubble bursting process and the subsequent production of jet droplets in great detail [Spiel, 1991; 1994]. The results can be used to formulate the oceanic source function, provided that oceanic bubble spectra are available for a wide range of conditions. However, this is not sufficient because sea spray aerosol droplets are also produced by direct tearing from the wave crests due to wind stress in high wind speeds ($u > 9$ m/s) [Monahan et al., 1986]. We feel that from the combination of realistic bubble-mediated jet droplet source functions derived from *in situ* bubble spectra, combined with simultaneously measured aerosol particle size distributions, the effects of direct tearing and bubble-mediated production can be separated. In that case, both production mechanisms can be evaluated as function of environmental parameters and combined into a consensus source function that applies in a wide range of conditions.

In this contribution we report on the bubble measuring system (BMS) that we developed and its deployment in open sea in studies on the air-sea exchange of gases and aerosols. The measurements of aerosol size distributions are described in brief. Flux measurements are described elsewhere in this report [Kunz and De Leeuw, 1994]. In section 2 we describe the BMS and the data processing, as well as the deployment from a float in open sea. In section 3 the aerosol measurements are described. An overview of the measurements and some preliminary results are presented in section 4. The results are discussed in section 5, which also gives the first conclusions. The bubble measurements were previously presented in De Leeuw and Cohen [1994].

2. THE BUBBLE MEASURING SYSTEM

A bubble measuring system (BMS) was developed and constructed by TNO-FEL. The sizes of single bubbles are measured in sea water to obtain bubble size distributions in the diameter range from 30 μm to 1000 μm . The BMS system is schematically shown in Figure 1. The sample volume is illuminated by a diode laser and viewed by a ccd camera via a telescope. The length of the sample volume is limited by the conical tubes in which windows and lenses are mounted. The lengths of these tubes have been chosen such that all bubbles inside the sample volume are in focus. The conical shape has been chosen to reduce the creation of turbulence near the sample volume.

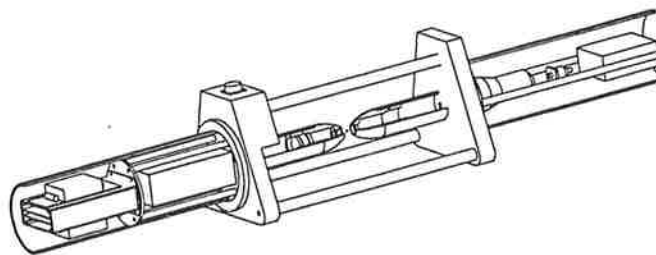


Fig. 1. The TNO-FEL bubble measuring system measures the sizes of single bubbles in the sample volume that is illuminated by the laser beam and imaged on the ccd camera through a telescope. The length of the sample volume is limited by the conical pipes in which the windows and lenses are mounted.

The camera signal is fed into a dedicated processing board for on-line analysis of the size and shape of objects in the sample volume. The processing assumes a spherical bubble shape and the aspect ratio is used as a selection criterion. Non-spherical shapes are assumed to be particles or biological species. The number of non-spherical shapes is retained in the data files. The images are also recorded on S-VHS video tape.

The BMS was calibrated with circles on paper which were photographically reduced to a known size. The smallest bubble size that we can presently measure is 60 μm , as compared to the aimed 30 μm . This smaller size could be measured by an optimized processing algorithm that will be developed for future applications, or by using a larger

magnification in the telescope (at the expense of the smaller sample volume).

During the ASGASEX campaign, power supply and data recording were located on the MPN and the BMS was deployed with a 100-m long cable to the tower. A self-contained system that telemeters the data to a receiving station ashore over a maximum distance of 10 NMi is also available.

During ASGASEX the BMS was deployed on a small float that consisted of a life-buoy and a pole extending 0.5-2 m (adjustable) below the surface. The BMS was mounted at the bottom of the pole. We deployed the BMS only at 0.5 m and at 1 m below the surface. The pole was mounted on gimbals to keep the BMS horizontal. The buoy was attached to the MPN tower with lines, at a distance of 50-100 m from the tower to reduce the influence of bubbles generated by (tidal) currents around the tower. Because the influence of the tidal current was still evident in the spectra, cf. Figure 2, a second float was anchored NW from the platform and used to keep the BMS out of the current-generated plume.

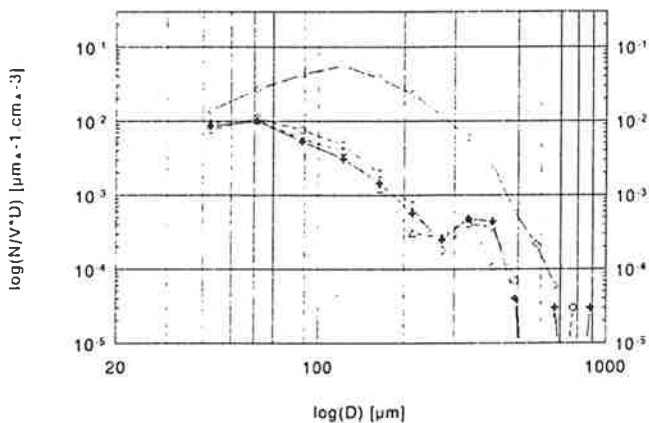


Figure 2. The influence of the bubble plume generated by the platform due to the tidal current is evident in the bubble size distributions shown in this Figure. The horizontal axis shows the bubble diameter in μm on a log scale, the vertical axis shows the number concentrations, per cm^3 , per size increment (in μm). The upper curve was measured in the bubble plume, the lower curves were measured outside the bubble plume. The number of bubbles in the plume is much larger, especially for the larger bubbles, which causes a shift of the median diameter to larger sizes. The data were collected at 0.5 m below the surface.

3. AEROSOL MEASUREMENTS

Aerosol particle size distributions were measured with two methods. An optical particle counter, a Particle Measuring Systems (PMS, Boulder, CO) CSAS 200 P that measures particles in the diameter range from 0.2-20 μm was mounted at the lower deck of MPN, at 11 m above the mean sea level. The optical particle counter was continuously operated and data were stored as 1-minute samples.

Profiles of particle size distributions were measured with Rotorod inertial impactor samplers mounted on the pulley and float system that was also used during previous experiments from MPN [De Leeuw, 1987, 1990]. The Rotorod samples particles larger than 12 μm in diameter. It consists of two polished stainless steel rods, mounted in a retracting collector head on a motor that rotates at a nominal speed of 2400 RPM. The linear velocity of the rods is 10 m/s. Particles impacted on the rods are retained by a sticky coating (silicone). Microscope images of the rods are digitized to determine the particle size distribution by computer. The profiles were measured at a distance of 13 m from the platform by using a horizontal mast extending north from the NW corner of the platform. Flow distortion by the platform structure, and the influence of waves breaking on the platform, are negligible at this position. The Rotorod samplers were mounted on a rod that slid along a vertical line extending from the tip of the mast down into the water. The measurements were made from just above the wave tops up to about 15 m. Wave following measurements were made with the Rotorods attached to a float, at 0.2 m to 3 m above the instantaneous water level.

4. RESULTS

a. Bubble size distributions

During the ASGASEX experiments, bubble size distributions were measured as 15-minute averages, in images recorded on video tape. Altogether about 36 hours were recorded, which translates into about 144 samples. The initial recordings, where the influence of the platform was evident, were deleted from the data set. Due to several other problems, also many of the images recorded later-on were not reliable and had to be deleted from the data set. After thorough validation, based on spectral shape and bubble concentrations, only 40 reliable bubble size distributions were left for further analysis.

In Figure 3 we present a comparison of one of these bubble size distributions with spectra presented in the literature. The fairly good agreement with the literature data gives confidence in our system.

A second test is the variation of the bubble concentrations with environmental parameters. Trends were observed that indicate a dependence of the bubble concentrations on fetch and wind speed. These are discussed below. Unfortunately, statistical relations cannot be derived because the data set is too small for a detailed analysis based on the subdivision for, e.g., a range of fetches or wind speeds (cf. Van Eijk and De Leeuw [1992] for an example of such an analysis of aerosol particle size distributions at MPN).

In the time series in Figure 4 we discern seven longer periods. For each period the observations are briefly described, including the meteorological characterization. We note that until 15 September the BMS was deployed at 0.5 m below the instantaneous water surface, and from 26 September the depth of the BMS was 1 m. The variation of the bubble concentrations with depth has been reported in the literature (e.g., Wu [1992]), but the present data base does not allow for such an analysis.

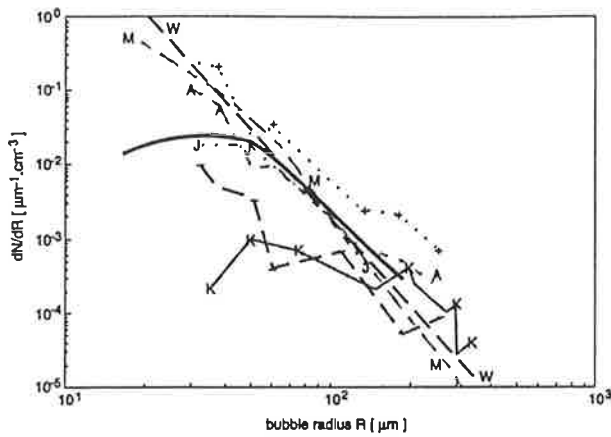


Figure 3. Comparison of bubble size distributions measured with our BMS, with spectra presented in the literature. All spectra were measured in a wind speed of 13 m/s. Other environmental parameters were different, however, as were the depths at which the data were collected and the techniques that were used. Note that bubble size is presented by the radius in μm , and therefore also the concentrations are given in number of bubbles per cm^3 per μm radius increment. The spectrum labelled K is from Kolovayev [1976], J is from Johnson and Cooke [1979], M is from Monahan [1988], W is from Wu [1989] and +, - and A are spectra from Medwin and Breitz [1989] just after wave breaking, the average and the minimum concentrations, respectively.

On September 10, 08:47-19:21, we have 6 spectra, over a total period of 10.5 hours. During this period the wind was from the south west, decreasing from 14.3 m/s to about 12 m/s. Significant wave height was 1.4 m. The thermal stratification was neutral to slightly unstable (air-sea temperature difference, or ASTD, was -0.5°C to -0.9°C). On average, the concentrations of the larger bubbles decrease, as expected. However, the concentrations of the smaller bubbles only initially decrease and then increase slightly. The latter observations are not yet understood. However, instead of wind speed, the effect of wind stress (or the drag coefficient) should be considered because these parameters describe wave breaking, and thus bubble formation, better than wind speed alone. Stress is also influenced by fetch, surface current, the angle between wind and waves, etc.

On September 13, 08:26-17:06, seven validated spectra are available spread over a period of 8.5 hours. The wind was from ESE, about 9.5 m/s. Significant wave height was 0.7 m. The thermal stratification was unstable, with ASTD -2.4°C . The bubble concentrations were fairly constant during this period, although more variation is observed in the concentrations of the larger bubbles than for the smaller ones. However, in spite of the lower wind speed as compared with the previous period on September 10, the bubble concentrations on September 13 are about a factor of two higher than on September 10, for all sizes. This effect is more pronounced for the smaller bubbles.

The increase in the concentrations is presumed to be due to the effect of fetch. The fetch in this ESE wind is much shorter than in SW winds. This results in younger waves which are steeper, resulting in more breaking. To verify these arguments, and to quantitatively describe the observed phenomena, data are required on stress and on whitecap recordings. Because the sonic anemometers were mounted on the west side of the MPN, i.e. in the wind shadow, direct measurements of the stress were not made. Whitecap data were not available at the time of writing this contribution.

On the next day, September 14, 5 validated bubble size distributions are available for an 8 hour period, 09:42-17:27. The conditions were similar to the previous day, although the wind speed was somewhat lower and decreased further (from 8.4 to 6.2 m/s). Wind direction was ESE, waves decreased from 0.8 m to 0.6 m. The thermal stratification was unstable with ASTD increasing from -4°C to -2°C . The bubble concentrations are seen to decrease only slightly, although for the larger bubbles the concentrations are very variable and no clear trend is observed.

On 26 September we have only three validated spectra over a four hour period between 15:38 and 19:20. Nevertheless this period is interesting because of the high wind speed of around 18 m/s, from the NE, with waves increasing from 3 to 3.4 m/s. Thermal stratification was unstable, with ASTD -3.1°C . The concentrations of the smaller bubbles ($\mu 214 \mu\text{m}$) are clearly higher (by about a factor of 2) than in any of the previous periods. For the larger bubbles, however, there are little or no data. This might be due to the deeper deployment of the BMS (-1 m as compared to -0.5 m during the previous periods), but this is not likely because in this high wind speed deeper mixing would be expected. Wave age might be another consideration, but presently we cannot explain the observations (or better the lack of observations) for the larger bubbles.

On the following day, September 27, the wind still was from northerly directions, but the wind speed had decreased significantly to 3-5 m/s. Waves were still high and decreasing from 1.7 m to 1.3 m. The ASTD increased from -1.6°C to -0.6°C . Four bubble spectra are available for a period of 3.5 hours between 06:06 and 09:25. The bubble concentrations were fairly constant, decreasing for the larger sizes. As expected, the concentrations were appreciably lower than during the preceding high wind period, and, except for the smallest bubbles, also lower than in other periods discussed thus far. This is explained by the weak winds and the long fetch, in which no significant whitecapping is expected.

On September 28, 06:10-21:52, we have 6 bubble size distributions over a 16 hour period, with a 10 hour interruption after the first two measurements. The wind was SSW, 8 m/s in the morning while in the evening series the wind speed started at 12.4 m/s, decreasing to 10 m/s. Significant wave heights were about 0.8 m.

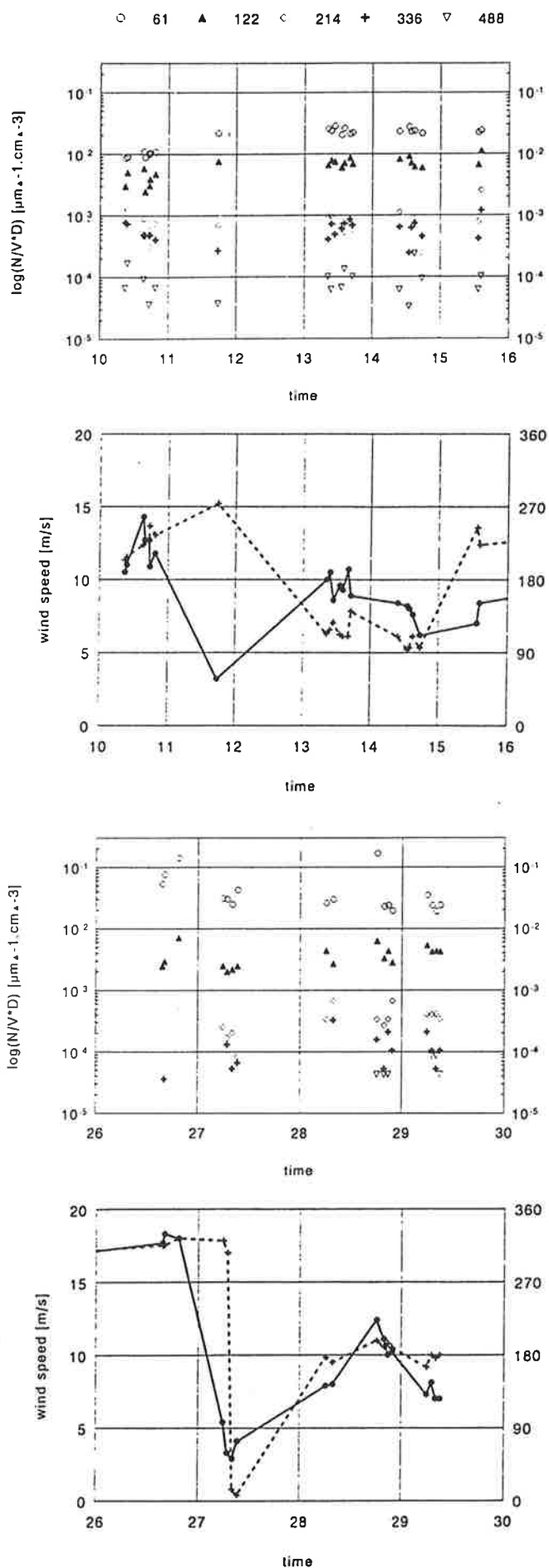


Fig. 4. Time series of bubble concentrations, for bubbles with diameters of 61 μm , 122 μm , 214 μm , 336 and 488 μm (see legend at the top), and wind speeds (solid line) and wind directions (broken line) during the measurements. The connecting lines in the wind data are not extrapolations. Note that the concentrations are given as number of bubbles per cm^3 per μm diameter increment. The upper two panels present the data for the first period from 10 to 16 September, the lower two panels are for the period 26 to 30 September. The numbers on the horizontal axes indicate the day in September.

The atmosphere was unstable, with ASTD -3.7°C in the morning and -2.7°C in the evening. The higher wind speed of 12.4 m/s at the start of the evening measurements resulted in a peak in the bubble concentrations for the smaller sizes, but not for the larger ones. In general, the concentrations were actually observed to decrease somewhat.

Presently we have no explanation for the latter observation. Maybe mixing processes are an issue both here and in the observations regarding the concentrations of the larger bubbles on the previous day, but at this moment we can only present speculations which are not supported by experimental data or models.

On September 29, 06:07-09:08, 4 bubble size distributions are available over a period of three hours. Wind speed was 7-8 m/s, from the south. Significant wave height was only 0.5 m and ASTD was -4.3°C , i.e. very unstable. The bubble concentrations are observed to decrease slightly, more for the larger bubbles than for the smaller ones.

b. Aerosol measurements

During ASGASEX eight profiles of particle size distributions were measured with the Rotorod inertial impaction samplers. These will be analyzed together with the eighteen profiles that were measured during the MAPTIP trial that was conducted from MPN immediately after ASGASEX [Van Eijk et al., 1994]. MAPTIP was focused on aerosols. An initial analysis of the ASGASEX/MAPTIP profile data base was presented in Davidson et al. [1995].

The optical particle counters were continuously operated. However, the wind directions during ASGASEX were predominantly easterly in which the particle counters were shielded by the platform. Hence the aerosol data are only reliable and representative for open sea conditions in winds with westerly components (wind directions 110° to 340° with respect to North). To obtain a statistically significant amount of data, also the aerosol size distributions measured with the optical particle counters are combined with the MAPTIP data. A preliminary analysis of the MAPTIP data set was presented in Van Eijk et al. [1995]. The primary objectives of these analyses was the validation of the MPN model, that is based on data collected during the HEXMAX experiments [Van Eijk and De Leeuw, 1992], the extension of the MPN model to a wider range of conditions and the generalization of the MPN model to other parts of the North Sea.

5. DISCUSSION AND CONCLUSIONS

In the above data presentation we have indicated trends in the bubble concentrations and the evolution of the bubble size distribution with environmental parameters. Wave breaking, and thus also the bubble concentrations resulting from the wave breaking process, are determined by wind speed and other parameters such as surface currents and fetch. Together these parameters also determine the wind stress. Unfortunately the stress could often not be directly measured during ASGASEX because all equipment had been mounted at the west side of the MPN, while during ASGASEX the winds came predominantly from easterly directions. Thus the instruments were sheltered by the MPN and reliable data could not be obtained during most of the time.

Direct measurements of the whitecapping ratio, the surface manifestation of the emerging bubble plume, were not available at the time of writing this contribution. Therefore we have thus far described our observations only in terms of the available bulk meteorological parameters such as wind speed, wind direction, significant wave height and ASTD. Another factor to consider is the surface current caused by tidal effects, and the effect of river outflows on the salinity and the surface roughness. Also biological effects should not be forgotten in this coastal North Sea area. Especially because these may also create bubbles.

Our validated data base turns out to be too small to separate and quantify individual effects on the bubble size distribution. The observation of large bubble concentrations in relatively short fetch is ascribed to the enhanced breaking of young waves that are steeper than in a well-developed wave field.

The results have not yet been interpreted in terms of air-sea gas exchange and the production of sea spray aerosol. This requires a modeling effort which we hope to pursue in the future.

The aerosol data collection was continued during the MAPTIP campaign that was conducted from the MPN in October-November 1993 [Van Eijk et al., 1993]. The ASGASEX and MAPTIP aerosol data are analyzed as one (semi-) continuous data base. These data are used to validate the MPN aerosol model that resulted from the HEXMAX data base [Van Eijk and De Leeuw, 1992], to extend this model to easterly wind directions, and to further analyze the height dependence of the aerosol size distributions. An initial attempt will be made to derive an aerosol source function for the area.

The deployment of the BMS during the ASGASEX campaign was a first test in open sea, and also the first deployment during an extended period of time. These initial tests have indicated that some improvements to the BMS are required, especially as regards the hard ware. The recordings show that the North Sea water contain many particles and organisms of biological origin. Our BMS cannot discriminate between spherical particles and bubbles. This might be a reason for the relatively large number of bubbles counted at low wind speeds, and the relatively small

sensitivity of the bubble concentrations to changes in wind speed. This asks for a detailed analysis of the recorded tapes for selected periods. If indeed many spherical shapes are counted which are not bubbles, a more sophisticated processing algorithm must be developed to discriminate between bubbles and other organisms and particles.

We feel that the bubble measuring system we developed is a useful contribution to the rare studies that are being made on bubble size distributions. The bubble concentrations and size distributions are similar to those obtained from other studies. Variations in the bubble concentrations have been explained by variations in environmental conditions. The data base is presently too small to derive quantitative relations between bubble size distributions and environmental parameters in a complicated environment such as the coastal North Sea.

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