STUDIES ON AEROSOLS IN THE MARINE ATMOSPHERIC SURFACE LAYER

A contribution to the EUROTRAC subproject ASE

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Summary

The work performed in 1992 in the framework of the EUROTRAC subproject ASE was mainly focused on three topics. The first was the extension of the modified CLUSE numerical model [Rouault et al., 1991; De Leeuw et al., 1992a] to over-ocean conditions. The modifications in the new code (SEACLUSE) include the influence of waves on the air flow and the evaporation of salt-water droplets. The second aim was to finalize the analysis of the TWO-PIE experimental data on tracer aerosol deposition on water surfaces in the presence of a simulated whitecap and associated sea spray aerosol. The CLUSE and TWO-PIE efforts are part of EUROTRAC ASE Subtopic 5: Factors determining particle dynamics over the air-sea interface, in cooperation with Riso National Laboratory, Roskilde, Denmark and Ecole Centrale de Nantes, France. The third effort was the participation in the field experiment from FPN in September 1992. The cooperation to integrate our modeling efforts with atmospheric chemistry models to study wet-phase chemistry and scavenging processes has been intensified.

The work that is part of EUROTRAC ASE subtopic 5 is described in De Leeuw et al. [1993a] and is therefore only briefly discussed in this contribution.

Aims of the Research

The aim of the research is to yield a description of the factors determining the production, the dynamics, and the deposition of the aerosol in the marine atmospheric surface layer through field experiments and laboratory investigations in combination with numerical and analytical modeling.

To achieve these goals, detailed studies are made of the production processes of sea spray aerosol, the subsequent transport from the production zone into the surface layer and the dispersal throughout the atmospheric boundary layer, including transformation due to evaporation and condensation, and the removal of both sea spray aerosol and particles of continental origin. Bubble-mediated droplet production is studied in detail through laboratory experiments and theoretical analyses of the bubble-breaking mechanism, while on the other hand adequate sea-spray source functions are sought for oceanic conditions as function of the meteorological conditions. The latter studies will finally also include effects of spume droplets that are generated by the direct action of the wind on the wave crests.

Similarly, the life cycle of the aerosol and the effect of sea spray on the surface-layer temperature and humidity profiles are investigated through models based on laboratory experiments. Field data are used for the extension to oceanic conditions, which requires adequate descriptions of the flow over the surface wave field.

The deposition velocity of sub-micron particles is not well known. Uncertainties of one order of magnitude are reported. To understand the causes for this large uncertainty, a laboratory study has been carried out to determine the effect of breaking bubbles on the deposition velocity. Breaking bubbles disrupt the laminar sublayer, which offers the largest resistance against deposition of sub-micron particles. Thus the deposition velocity should be enhanced as more bubbles reach the surface and disrupt the laminar sublayer.

Sea-spray droplets may also affect wet-phase chemistry. The study of these processes is performed in an international and multi-disciplinary cooperation between different institutes.

For all these studies, experimental data are required on the aerosol size distributions as function of height in the surface layer. Such data were obtained during the EUROTRAC ASE experiment from FPN in September 1992, where we measured for the first time the height dependence of aerosols, with different techniques, over a size range from 0.16 μm to about 100 μm in diameter. Complementary measurements were made by other participants to this experiment.

Activities During the Year

The activities in 1992 were a continuation of the work described in De Leeuw et al. [1991, 1992a]. They were mainly concentrated on three topics, i.e. the analysis of the TWO-PIE experimental data, numerical modeling and participation in the EUROTRAC ASE experiment from FPN in September 1992. The TNO contribution to the FPN experiment is described in more detail below. Our activities in the field of numerical modeling were concentrated on the optimisation of the CLUSE code for fresh-water droplets and flat water surfaces, and the extension of this code to oceanic conditions. The latter requires the replacement of fresh-water droplets by salt-water droplets which only partly evaporate, the introduction of surface waves and the associated effect on the wind field. These activities, resulting in the SEACLUSE model [Van Eijk et al., 1993], have been described in more detail in the common EUROTRAC ASE topic 5 report [De Leeuw et al., this issue]. This common EUROTRAC ASE report also describes the re-analysis of the TWO-PIE experimental data on the removal of tracer aerosol particles over a simulated whitecap, and the cooperation with Johannes Gutenberg Universitaet in Mainz, Germany, and Chalmers University of Technology in Stockholm, Sweden, to model the influence of sea-spray aerosol droplets on atmospheric chemistry. The analytical description of the jet droplet production from a rising air bubble has been completed [Dekker and De Leeuw, 1993]. Other TNO studies on air-sea exchange include the analysis of aerosol and lidar data collected near the Florida Keys [De Leeuw and Kunz, 1992], over the North Atlantic [De Leeuw et al., 1993b] and over the North Sea [Van Eijk and De Leeuw, 1992], and their application for model validation, as well as a field experiment at the North Sea near the German Frisian Island Sylt [De Leeuw et al., 1992b].

TWO-PIE. The efforts of TNO in the TWO-PIE analysis were concentrated on the re-analysis of the time-serial data on the removal of tracer aerosol particles in the presence of bubbles. Bubble fluxes, wind speeds and relative humidity were varied and the removal rate was determined for a range of particle sizes. Figure 1 shows the removal rate as function of particle size, for a wind speed of 3 m/s and a relative humidity of 90%, and four different bubble fluxes. The removal rate has been plotted because that is the parameter that was directly measured. This rate includes the removal at the tunnel surfaces other than the water surface, such as the heat exchangers and the wind generator. Since these effects are independent of bubble rate, they do not affect the final conclusions. The data in Figure 1 show no systematic dependence of the removal rate on the bubble flux. This is further illustrated in Figure 2, where the removal rate for six different particle sizes (see legend) has been plotted versus the bubble flux. The effect of bubbles on the deposition velocity is further discussed in De Leeuw et al. [1993a].

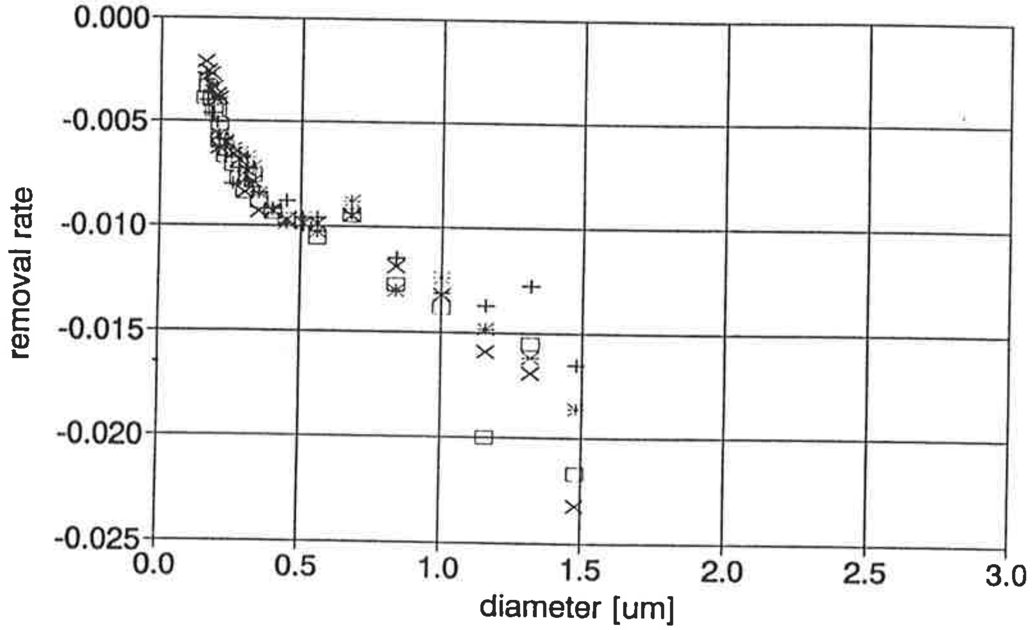


Figure 1. Rate of removal of tracer aerosol particles versus particle size. The measurements were made at a reference wind speed of 3 m/s, a relative humidity of 90%, and at four different bubble fluxes: + all bubblers in operation, * with 2/3 of the bubblers, □ with 1/3 of the bubblers, x no bubbles.

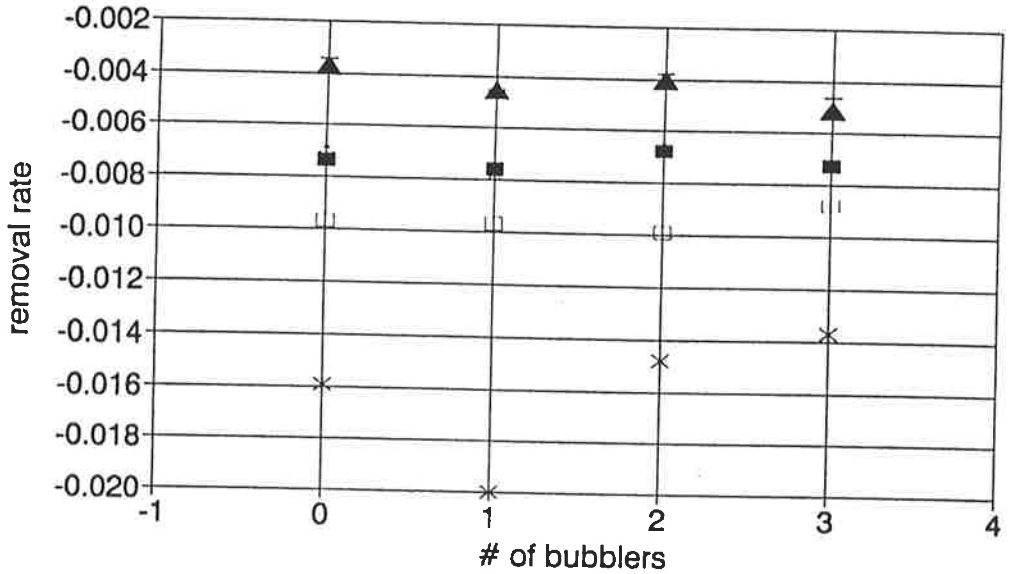


Figure 2. Tracer aerosol removal rate versus bubble flux (relative units), at a wind speed of 3 m/s and a relative humidity of 90%. Particle sizes are 0.19 μm (+), 0.20 μm (▲), 0.28 μm (*), 0.31 μm (■), 0.45 μm (□) and 1.16 μm (x).

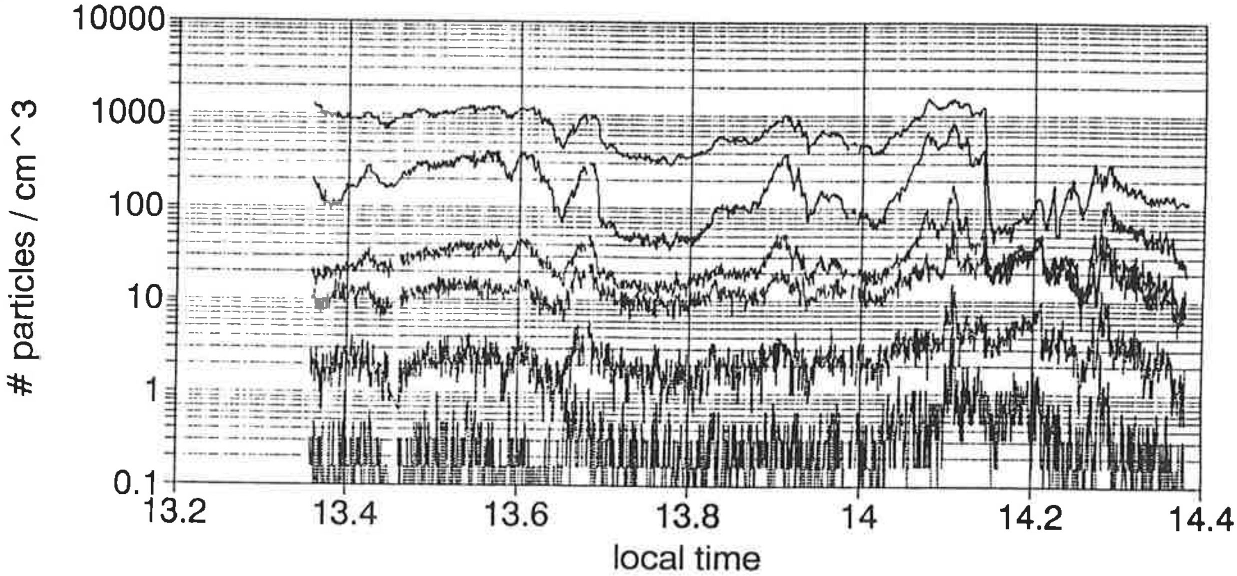
EUROTRAC ASE experiment. The EUROTRAC Air-Sea Exchange experiment was conducted from the Forschungs Platform Nordsee (FPN), 1-22 September, 1992, in a cooperation between seven institutes from Germany (Univ. Hamburg, MPI, Mainz), Belgium (Univ. Antwerp, Univ. Brussels, Univ. Gent), Denmark (Riso) and The Netherlands (TNO Physics and Electronics

Laboratory). The main objective of the TNO Physics and Electronics Laboratory during this experiment was the extension of our data base on the height dependence of particle size distributions in the marine atmospheric surface layer, for a wide variety of conditions. This data base is used for studies of the factors that are important for the dynamics of the aerosol in the marine boundary layer. Results from the statistical analyses will be used to improve the physical descriptions of the aerosol properties, including production, dispersal and removal (deposition). The final goal is the development of a comprehensive physical model that is generally applicable.

The emphasis during the EUROTRAC ASE experiment was on the smaller particles which were measured with optical particle counters, in addition to the giant particles for which a relatively large data base is now available for the North Sea and the North Atlantic. The FPN offered the opportunity to combine, for the first time, both techniques. To this end, optical particle counters of TNO (a CSASP-200 from Particle Measuring Systems, Boulder, CO, USA), size range 0.2-20 μm in diameter) and University of Hamburg were mounted in a cage which was hooked to the 20-meter crane at the SW corner of the platform [Schulz and Dannecker, 1990]. The height dependence of the particle size distributions was measured at a distance of about 15 m upwind from the platform, at levels between 2 and 30 m above mean sea level. These aerosol spectra were extended to larger diameters (100 μm maximum) by simultaneous Rotorod [De Leeuw, 1986] measurements from the cage at the same heights. Additional measurements were made with the Rotorods mounted on a wave-following buoy system at levels between 0.35 and 3 m and at fixed heights between 1 and 5 m. Relative humidity and air temperature were also recorded from the cage. During these profiles, Riso recorded particle size distributions at a fixed height of about 27 m, to monitor changes in the ambient aerosol concentrations, which would possibly also influence the profile during the course of its measurement. Prior to and during the experiment, particle counter comparison experiments were conducted.

In between the profile measurements, the cage was put at a fixed height of 4-6 m above the sea surface, where 1-min averaged particle size distributions were continuously recorded. These short intervals were chosen to study the response of the aerosol concentrations to changes in meteorological conditions. An example is presented in Figure 3, where time traces are shown for particles in several size ranges, measured during 24 hours (13 Sep, 08:35 - 14 Sep, 09:10). The data, which were averaged over 1-min intervals, illustrate the scales involved in the change of aerosol concentrations and their response to changes in ambient conditions.

An example is the sudden change in the concentration of the smallest particles in the early morning of 14 September (around 03:30 am). A similar change is also observed for larger particles, although less obvious as particle size increases. This change is due to the passage of a cold front, and the accompanying change in wind direction from S to WNW (see Figure 3b for meteorological parameters). Between 3 and 4 am the rain stopped and the relative humidity dropped significantly. Also the temperature decreased from 14.3^o to 11.2^o, i.e. a change of about 3^o resulting in very unstable thermal stratification (sea water temperature was 17.5^o). At the same time we observed that the concentrations of the large particles increased significantly. This is ascribed to the increase in wind speed from 10 m/s to 18 m/s over a period of only 7 hours. Also the change in thermal stability could have been an important factor causing both an increase in the whitecap ratio [Monahan and O'Muircheartaigh, 1986] resulting in a higher production rate, and more efficient transport of the freshly produced particles. In addition, also confused seas, due to a wave front travelling through a cold front, may have contributed to the observed effects. Further, the wash-out by rain, and the occurrence of small rain drops, may have caused the observed variations in the particle concentrations. Visibility was observed to vary with the occurrence of rain and showers and seems to be related to the variations in aerosol concentrations, as expected.



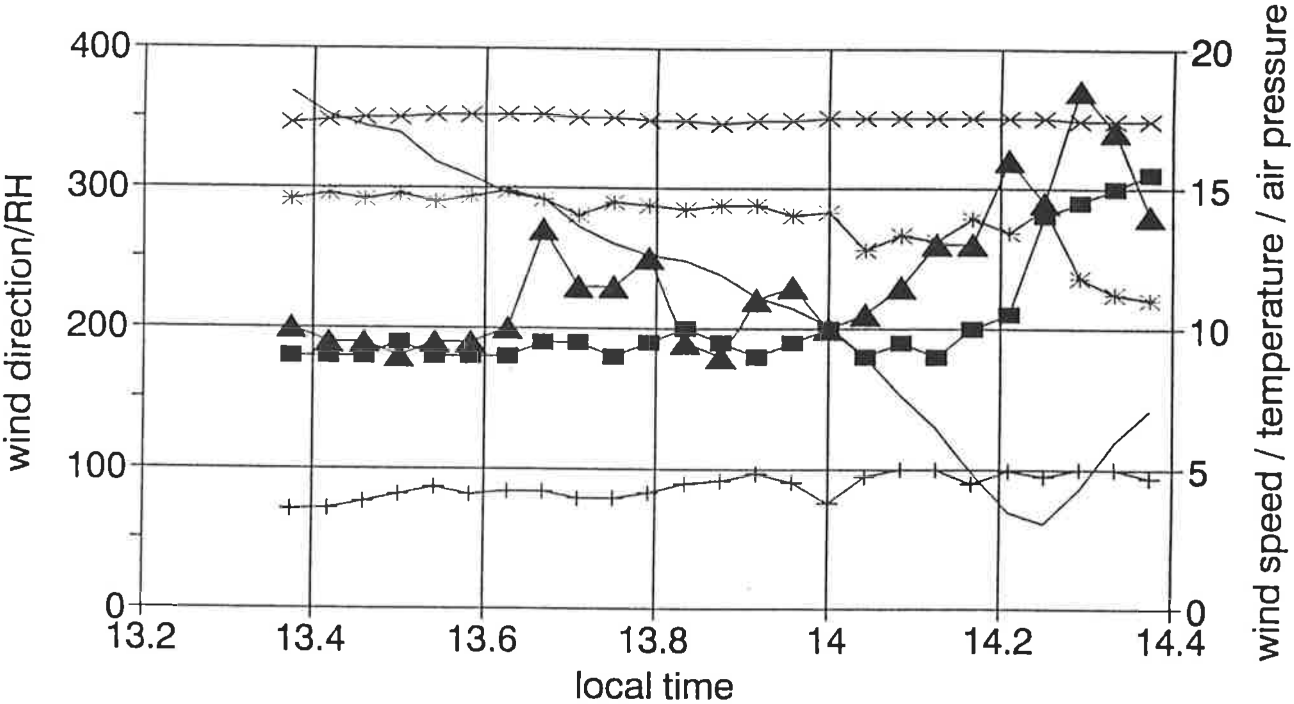


Figure 3. Top: time series of aerosol concentrations averaged in the diameter intervals of 0.2-0.3 μm, 0.3-0.5 μm, 0.5-1.0 μm, 1.0-2.0 μm, 2.0-5.0 μm and >5 μm (top to bottom traces). Time averages are over 1 minute. The data shown are an indication for the time scales involved in the change of aerosol concentrations in response to changes in ambient conditions.
Bottom: time series of hourly observations of meteorological parameters: ■ wind direction [deg. from North]; ▲ wind speed [m/s]; x water temperature [deg. C]; * air temperature [deg. C]; + relative humidity [%]; - air pressure [hPa; - 1000].

The detailed analysis of the aerosol data is among the goals for 1993.

The profile measurements were made between 9 and 20 September. In the afternoon of 16 September the TNO optical particle counter failed, probably due to a wave washing over the cage when the wind rapidly picked up to a storm. Repair was not possible, but fortunately data are available from the optical particle counters of the Univ. of Hamburg group.

In the coming period, the data will be analyzed. Correlations will be made between the aerosol concentrations and meteorological parameters, the height dependence of the concentrations will be investigated and a physical model will be further developed and tested with the data available from both the FPN experiment and previous experiments at the North Sea. These efforts will include a critical evaluation of the aerosol source function and studies on aerosol deposition and on the relative contributions to the total concentrations of aerosols of continental and marine origin. Air mass trajectories and weather maps are available.

Principal Results

The principle results from the activities summarized above are:

- the optimization of the CLUSE model for fresh-water droplets over a flat water surface has been completed
- algorithms for the evaporation of salt-water droplets and for the description of the air flow over waves have been introduced in the CLUSE model for application over the open ocean (SEACLUSE)
- SEACLUSE is now ready for testing and for validation with experimental data
- an analytical model has been developed that describes the temporal behaviour of a bubble protruding the water surface in terms of a simple capillary wave model
- the final results from the TWO-PIE deposition experiments lead to the conclusion that the removal of the tracer aerosol is independent of the whitecap ratio (related to the bubble flux at the air-sea interface, giving the total surface area of the laminar sublayer that is destroyed by bubbles protruding the air-sea interface). For the final interpretation, other factors, such as the additional vertical air flow caused by the bubbles and the scavenging by sea-spray droplets, must be taken into account
- the data base for the height dependence of aerosol size distributions in the marine atmospheric boundary layer has been expanded with particles smaller than 10 μm in diameter, which are of significance for deposition studies
- the data base for the larger particles (D>10 μm) has been extended to a wider range of conditions. In particular, the effect of continental contributions can be better quantified because chemical composition was determined by other participants, as well as effects of water depth, fetch and water temperature on the concentrations (cf. De Leeuw and Kunz [1992]).

Main Conclusions

The SEACLUSE model, i.e. the extended CLUSE model for application in open-ocean conditions, has been formulated and is almost ready for testing and evaluation. The experimental aerosol profile data from the HEXOS experiments [De Leeuw, 1990] will be used for this purpose. The data base has been extended during the EUROTRAC ASE experiment from FPN in September 1992. An important contribution from this experiment is the height dependence of small (D<10 μm) particles and the combination with chemical composition. Cooperations to include atmospheric chemistry have been established. This will lead to a comprehensive picture of the aerosol dynamics above water surfaces. Concentrated efforts have been made on the analysis of the TWO-PIE particle deposition data. This analysis has not yet been completed. Therefore, definite conclusions are not available at this stage. An analytical model for bubble-mediated droplet production has been formulated which shows that a parent-bubble/daughter-droplet relationship can be derived from a simple capillary wave model which predicts the correct ejection velocities.

Aims for the Coming Year

The activities in 1993 will focus on:

- the conclusion of the analysis of the TWO-PIE deposition data and the interpretation of the results,
- the critical evaluation of the oceanic source function for sea-spray aerosol,
- test and evaluation of the SEACLUSE numerical model with field data, and the application of the model to estimate the effects of sea spray aerosol on the total water vapor and latent heat fluxes,
- integration of the HEXIST/CLUSE/PIE modeling efforts with atmospheric chemistry models to study wet phase chemistry and scavenging processes, and the influence of sea-salt aerosol on the transformation of N-compounds,
- analysis of the data from the EUROTRAC ASE experiment in September 1992,
- using the results from the source function and deposition analyses, a start will be made with the evaluation of the relative contributions of aerosol of continental and marine origin to the concentrations in coastal regions.

In addition, we will participate in experiments at the North Sea in the Fall of 1993, aimed at studies of bubbles and their influence on air-sea exchange of gases (CO_2 and water vapour) and aerosols (September) and at marine aerosol properties (October/November). Air-sea exchange studies near the ocean margins (OMEX) will be prepared.

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Part 3

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