GENERATION, TRANSPORT AND DEPOSITION OF MARINE AEROSOLS

A contribution to the EUROTRAC subproject ASE

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Summary
The work performed in 1990 in the framework of EUROTRAC and related research is summarized. This includes the analysis of the CLUSE data to derive the aerosol source functions specific for the CLUSE experiments in fresh and salt water, a study on the source function for the marine aerosol, and comparison of aerosol profiles in laboratory and field situations. Particle deposition on water surfaces was studied during the TWO-PIE experiments in June 1990 in Marseille.

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 experimental work in an air/water tunnel and numerical modeling. Additional data will be used from previous field experiments for the interpretation of the results to oceanic conditions. Specific goals for 1990 were:

- the determination of the source function in the CLUSE specific experimental configurations for fresh and salt water
- investigations on oceanic source functions for marine aerosols
- application of the Lagrangian CLUSE model[1] for aerosol transport over water waves
- participation in the TWO-PIE experiment on particle deposition velocities on broken water surfaces.

Activities during 1990

The analysis has been continued of the aerosol measurements during the 1988 CLUSE experiments in the Large Air-Sea Interaction Simulation tunnel of IMST in Luminy, Marseille (France) and the Petit-CLUSE 3 (October 1988) and 4 (April 1989) experiments in the Whitecap Simulation Tank at the Marine Sciences Institute of the University of Connecticut at Avery Point, Groton (CT, USA). During these experiments, a whitecap was simulated with an array of submerged aquarium frits. Source functions for jet droplets produced from this whitecap were determined with MgO-coated glass slides that were kept horizontally at different levels between 2 cm and 16 cm above the water surface. The upward fluxes of the freshly

produced jet droplets were determined from the size distributions of the droplets impacted on the bottom side of the glass slides. The surface flux was determined by extrapolation. The bubble fluxes, and thus also the droplet fluxes, from different frits varied due to, e.g., pore size. Therefore the surface flux measurements were made at several locations over the whitecap and averaged to obtain a representative mean value for the experimental configuration. The results from the MgO impaction method compare favorably with the estimates made by Edson[1]. The latter were based on assumptions involving the bubble spectra and data on bubble rise speed and bubble mediated production. The parameters were tuned to yield agreement of model profiles with experimental data in non-evaporating conditions. In view of these assumptions, the results are surprisingly good, see, e.g., Larsen et al.[2] Additional droplet flux measurements were made with an Optical Array Probe (Particle Measuring Systems, Boulder, CO) during the TWO-PIE experiments in June 1990 in Marseille (see below).

For interpretation of the laboratory experiments and the models resulting from these, over-sea and laboratory aerosol profiles have been compared.[3] Differences result from advection, mixing properties, production, removal and chemical composition. In the laboratory model fresh water aerosol is continuously produced over the whole length of the simulation tunnel. The droplets evaporate completely when relative humidity is lower than 100%. In the return flow channel they are removed by impaction on the fan, heat exchangers and other surfaces. Over the ocean the aerosol consists of a mixture of human-made, biological and natural aerosol of continental and marine origin, containing a large variety of hygroscopic and non-hygroscopic species which strongly influence the dependence on relative humidity. The aerosol concentrations result from the balance between local production and removal and the advection of aerosol produced elsewhere. The advected concentrations depend on the meteorological conditions that determine both the trajectory of the air mass and the fate of the aerosol. The meteorological conditions also determine the local production and removal of aerosol. In particular the production of sea spray is different from the laboratory situation. Sea spray is produced from air bubbles that are formed by breaking waves. The bubbles rise due to buoyancy and when they protrude the surface they produce jet drops and film drops.[4] Wave breaking is an intermittent process resulting in a whitecap. In contrast to the continuous laboratory whitecap, the whitecaps due to breaking waves evolve through several stages during which the bubble spectra and the bubble fluxes through the surface change also.[5] As a consequence, also the sea spray production rate varies with time. An estimate of the effect of intermittent and time-dependent production rates has been made, based on a compilation of published bubble size distributions. The data from acoustic measurements by Medwin and Breitz[6] give quantitative

information on the bubble concentrations at different times after wave breaking. Differences are more than one order of magnitude. The aerosol fluxes derived from these bubble data were compared with aerosol source function estimates published by Miller and Fairall[7] and Monahan et al.[8] The differences between the Miller and Fairall data and our results may be an indication for the production rate of spume droplets. The latter are produced by direct tearing from the crests at wind speeds over 9 m/s.

Initial computations have been made by Edson[9] on the influence of waves on the droplet profile. The results show that the wave rotor that was initially proposed by De Leeuw,[10] cannot be solely responsible for the maximum in the profile near wave-crest heights. However, if spatial distribution of the droplet source function is included, an elevated maximum is reproduced. It is not clear at this moment to which extent spume droplets contribute to this maximum. Spume droplets were not yet considered in the calculations.

An interesting difference between profiles measured over the ocean and the results from both the CLUSE experiments and the CLUSE numerical models is the droplet gradient. The CLUSE results show very strong decreases in the droplet concentrations with height,[1,9] whereas the field data show that the gradients for similar droplet sizes are usually much smaller.[10] Obviously this is due to the differences between laboratory and field conditions that were outlined above. In particular, we conjecture that the strongly different gradients are due to the advection of aerosol.[3] The advected aerosol is in effect a 'background' that adds to the profile due to the locally produced aerosol. In view of the long mixing times, the latter constitutes only a small fraction of the total aerosol concentration measured.

The series of CLUSE experiments was concluded with the workshop 'Modelling the fate and influence of marine spray', held 6-8 June 1990 at IMST, Laboratoire de Luminy, Marseille (France). Results from the CLUSE laboratory experiments and modeling efforts and from HEXOS field experiments were presented and discussed. They are described in the Workshop proceedings.[11]

The advected aerosol is particularly important as regards atmospheric input of pollutants into the North Sea. Large uncertainties exist concerning the deposition velocity of the sub-micron aerosol fraction on water surfaces, which contains industrial, agricultural and other man-made pollutants such as sulphuric and nitrogeneous compounds. The reduction of the uncertainties in these deposition velocities were the objective of the TWO-PIE experiments that were conducted in June 1990 in the Large Air-Sea Interaction Simulation Tunnel of IMST in Marseille. The experiments were aimed at the study of the influence of broken water surfaces and aerosol production rates, due to bubbles protruding the surface layer and thus disrupting the viscous sublayer. This effectively reduces

the resistence for deposition, resulting in an enhanced deposition velocity. Results from a feasibility experiment during the 1988 CLUSE experiments in Marseille indeed show higher deposition velocities in the presence of bursting bubbles as compared to a still water surface.[2] Based on our experience during the 1988 feasibility experiment, we modified the procedures for the June 1990 final experiment TWO-PIE. Tracer aerosol particles were generated and injected into the tunnel during a short time. Particle size distributions of the tracer aerosol were measured with several optical particle counters. Time series were continuously measured at one fixed level. Profiles were measured from close to the surface to the top of the tunnel boundary layer. Since about four profiles were measured during each run, these profile measurements yield additional time-serial information at a range of levels. The measurements were made both with and without bursting bubbles, at three different bubble rates, each of these at three different humidities from 70% to 100% and at four different wind speeds from 1.5 m/s to 9 m/s. The time-serial measurements directly yield the decay rates as a function of particle size, after correction for the background aerosol. The profile method yields the deposition velocities from the gradients of the particle concentrations. From comparison of the decay rates for different bubble rates, the influence of bubble bursting can be determined since the experimental conditions were otherwise unchanged.

Results from these experiments are not yet available. Preliminary analysis shows that, as expected, the removal of the tracer aerosol increases as wind speed increases. To determine the influence of the bubble rate, detailed analyses need to be made. This is the principle effort planned for next year, provided that funding will become available.

Principal results

The research summarized above yielded the following results:

-measurements of droplet source functions with MgO-coated glass slides are feasible in the typical CLUSE laboratory situation where droplets are only produced from whitecaps simulated by using aquarium aerators; care must be taken however with the calibration of the ratio of the droplet impact diameter to the true droplet diameter

-the mean droplet source functions for the CLUSE specific laboratory conditions have been experimentally determined for fresh and salt water; these data will later be combined with the simultaneously measured bubble spectra

-over-ocean jet droplet source functions have been estimated from data available from the literature on oceanic bubble spectra, bubble rise times, and bubble/droplet relations

-effects of spume droplets on the source function still need to be determined

-comparison of aerosol profiles shows that the concentration gradients in the CLUSE experimental and numerical laboratory models, which neglect advection effects, are much stronger than over the ocean
-laboratory data have been collected to determine the influence of bubbles and spray on the deposition velocity of sub-micron particles on water surfaces.

Main conclusions

The work described above was made as part of a collaboration between several institutes (see list of authors). Conclusions reached from the experimental and numerical studies have been published in various papers. Comprehensive overviews of the present state-of-the-art of our research can be found in the recently published proceedings of the workshop on 'Modeling the fate and influence of marine spray'.[11] The results from the experimental efforts of the TNO Physics and Electronics Laboratory were used for model testing and as inputs to the models, e.g., the aerosol source function. Conclusions from these analyses concern the experimental methods to measure aerosols, the aerosol source functions and the shape of the profiles:
-impaction and optical methods yield similar results provided that the proper calibrations are used
-calibration of MgO impaction slides involves the thickness of the MgO layer
-Rotorod rotating impaction samplers can be used to determine the droplet size distributions provided that the droplet concentrations and the sample times can be matched such that coincidence of the droplets is negligible
-droplet surface fluxes can be measured in the CLUSE laboratory situation with MgO-coated glass slides; results compare favorably with data derived by other methods
-the aerosol surface flux over the ocean is time-dependent due to the intermittent nature of wave breaking
-profiles are not solely due to the balance between production and removal, advection effects must be considered as well to understand the gradient of the profile
-the observed shape of the profile, and in particular the maximum near wave-crest height,[10] has started many discussions which thus far have not been conclusive.[9,12,13,14]

Aims for the coming year

The aims for the coming year are:
-to finish the analysis of the CLUSE and petit-CLUSE 3 and 4 data concerning aerosol profiles, the relation between bubble spectra and aerosol source functions in fresh and salt water, the influence of waves on aerosol profiles and

the spectra of temperature, humidity and air flow.
-the analysis of the TWO-PIE deposition data
-to determine over-ocean aerosol source functions from the analysis of field-experimental data on aerosol profiles

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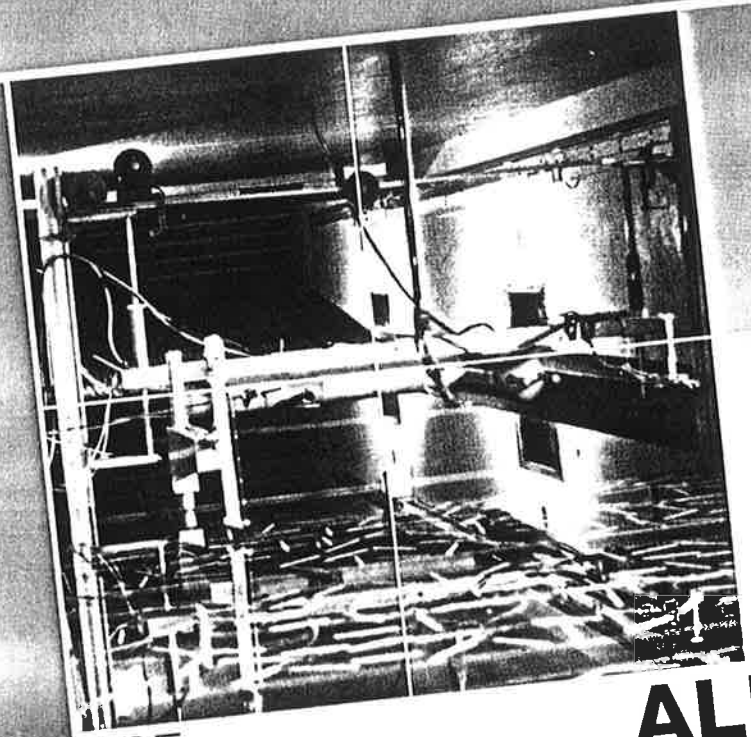

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