7.2 FACTORS DETERMINING PARTICLE DYNAMICS OVER THE AIR-SEA INTERFACE.

A contribution to the EUROTRAC subproject ASE, topic 5

Gerrit de Leeuw¹, Soren E. Larsen² and Patrice G. Mestayer³

1. TNO Physics and Electronics Laboratory, P.O. Box 96864, 2509 JG The Hague, The Netherlands
2. Department of Meteorology, Riso National Laboratory, Roskilde, Denmark
3. Equipe Dynamique de l'Atmosphere Habitee, Laboratoire de Mecanique des Fluides, Ecole Centrale de Nantes, Nantes, France

Summary

The work performed in 1992 in the framework of the EUROTRAC subproject ASE was mainly focused on modeling and on the finalization of the analysis of laboratory data on the influence of bubbles on the deposition velocity on water surfaces. The modeling efforts concentrated on the extension of the modified CLUSE numerical model [Rouault et al., 1991; De Leeuw et al., 1992] to over-ocean conditions (SEACLUSE [Van Eijk et al., 1993]) and to the evolution of small continental aerosol particles (CLUSA [Mestayer, 1993]). The modifications in the SEACLUSE-code include the ejection process, the influence of waves on the air flow and the (partial) evaporation of salt-water droplets. In the CLUSA model the emphasis is on the growth of 0.05-5 μm particles due to condensation of water vapor and the deposition of these particles to the water surface. The experimental part of the programme includes laboratory experiments on the deposition velocity of sub-micron tracer aerosol particles on water surfaces in the presence of a simulated whitecap and associated sea spray aerosol (project TWO-PIE). The aim for 1992 was to finalize the analysis of these experimental data. The cooperation with the Johannes Gutenberg Universitaet in Mainz, Germany, and Chalmers University of Technology in Stockholm, Sweden, to integrate our modeling efforts with atmospheric chemistry models to study two-phase chemistry and scavenging processes, has been intensified. TNO and Riso participated in the EUROTRAC ASE field experiment that was conducted from the Forschungs Plattform Nordsee (FPN) in September 1992.

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.

Bubbles protruding the water surface produce film and jet droplets, while at high wind speeds sea-spray droplets are also produced by direct tearing from the wave crests. The study of the production mechanism and the formulation of an oceanic source function are among our objectives. At the same time the deposition velocity of sub-micron aerosol particles may be enhanced because the bubbles disrupt the laminar sub-layer which offers the largest resistance against the deposition of these particles. This mechanism has been investigated during the 1990 TWO-PIE experiments in the Large Air-Sea Interaction Simulation tunnel at Laboratoire de Luminy (Marseille, France), by using tracer aerosols for which the removal was measured in the presence of variable bubble source intensities [De Leeuw et al., 1991].

During the 1988 CLUSE experiments in the same tunnel [Mestayer et al., 1988], the effects of the evaporation of droplets produced by the bubbles on overall sea-air transfer of water vapour and latent heat have been studied. The CLUSE experiments have resulted in a Lagrangian [Edson, 1989] and a Eulerian [Rouault, 1989; Rouault et al., 1991] model for fresh water droplets over a flat water surface and their effects on humidity and temperature profiles. For extension to oceanic conditions, the influence of waves on the air flow has to be considered and fresh water droplets have to be replaced by sea-spray droplets which can only partly evaporate.

In addition, wet-phase chemistry may occur on the droplets, which modifies the air chemistry. The study of these processes is performed in an international and multi-disciplinary cooperation between different institutes.

Activities During the Year

The activities in 1992 were a continuation of the work described in De Leeuw et al. [1992]. They were mainly concentrated on the analysis of the TWO-PIE experimental data and on numerical modeling. These activities are described in more detail below. In addition, the cooperations have been established between the partners in ASE subtopic 5 (Riso, ECN, TNO) and Johannes Gutenberg Universitaet in Mainz, Germany, and Chalmers University of Technology in Stockholm, Sweden, to include the influence on atmospheric chemistry of the droplets generated at the air-sea interface and dispersed throughout the marine boundary layer. The analytical description of the jet droplet production from a rising air bubble has been completed [Dekker and De Leeuw, 1993].

Experimental data, required for evaluation of the models, were obtained during the EUROTRAC ASE field experiment that was conducted from FPN in September 1992 (see De Leeuw et al., this issue).

TWO-PIE analysis. The analysis and interpretation of the TWO-PIE experiments in May/June 1990 in the Large Air-Sea Interaction Simulation Tunnel of IMST Laboratoire de Luminy (Marseille, France), see De Leeuw et al. [1991] for a description, has almost been completed. The objective of these experiments was to study the influence of whitecaps (bubbles) and associated sea spray on the deposition velocities of tracer aerosol particles [Larsen et al., 1990a, b]. To this end, tracer aerosol particles were released and their deposition was determined from the decrease of the concentrations with time (see also De Leeuw et al., 1993, this issue), and from profiles of particle concentrations. The influence of the whitecap coverage was determined by repeating these experiments at four different bubbler rates while keeping all other parameters (wind speed, relative humidity, air and water temperature) constant.

Most of the cases have now been analyzed. The final results show no systematic variation of the deposition velocity with the bubble flux. This is illustrated in Figure 1 for $0.283 \mu\text{m}$ tracer aerosol particles. The deposition velocities were determined from the decrease in the concentration in successively measured profiles, at wind speeds of 6 m/s and 9 m/s. The bubble fluxes were varied by operating no bubblers, 1/3 of the bubblers, 2/3 of the bubblers or all bubblers.

The absence of an effect caused by the bubble flux is further illustrated in Figure 2, where the deposition velocities for $0.483 \mu\text{m}$ particles has been plotted as function of wind speed and bubble flux. The deposition velocity clearly increases with wind speed, as expected, but is independent of bubble flux.

Figure 1. Deposition velocities of 0.283 μm tracer aerosol particles in the IMST tunnel for wind speeds of 6 m/s and 9 m/s, plotted as function of bubble flux. The deposition velocities were determined by the removal of the aerosol particles as function of time.

Figure 2. Deposition velocities of 0.483 μm tracer aerosol particles in the IMST tunnel as function of wind speed and bubble flux.

The conclusion that the deposition velocity is independent of the bubble flux is in contrast with our earlier impression that the deposition velocity does increase with the bubbler rate [De Leeuw et al., 1991; 1992], which was caused by an erroneous background correction. All high-humidity cases have now been analyzed, the analyses of the lower-humidity cases will be completed in 1993. The interpretation of the results and the preparation of a final report will conclude the TWO-PIE efforts.

Numerical modeling. The work on the Eulerian CLUSE model [Rouault et al., 1991] has been continued. The optimisation of the code for the original model for fresh-water droplets and flat surfaces has been completed.

SEACLUSE. The development of the SEACLUSE model that applies to open-ocean conditions required two major changes to the original model:

1. The introduction of an algorithm for the evaporation of salt water [Mestayer, 1990]. In contrast with fresh water, a salt water droplet cannot evaporate completely, but only to an extent determined by the salt concentration and the ambient relative humidity. Therefore, the model must keep track of the salt concentration of the droplets.
2. The introduction of a module to describe the air flow over the waves. The module calculates the field of horizontal and vertical winds over the wave surface (represented by Stokes waves). An example is shown in Figure 3. The wind field is then used to calculate droplet trajectories that yield flight times, final fall velocities and initial profiles for the droplet concentrations. This information, together with the ejection velocities, is stored in look-up tables for use during the actual model run.

Figure 3. Modelled wind field over the wave surface. The arrows represent both the direction and the velocity of the wind in this 2-D representation. Clearly, the flow is reversed in the wave trough resulting in an eddy. The wave surface is represented by a Stokes wave (Figure from Van Eijk et al. [1993]).

These two major changes, as well as various minor changes, have been introduced to yield the first version of the model for open-ocean conditions. In its present state, SEACLUSE is nearly ready for testing and validation with experimental data.

CLUSA. The description of the evolution of continental aerosols over water surfaces requires a description of the air flow in a coastal atmospheric boundary layer. Particles are considered in the 0.05-5 μm radius range. The profiles of wind speed and turbulent viscosity in the coastal zone must be described as function of the 10-meter wind speed and the fetch, over a developing wave field. The upper limit boundary condition is that of an infinite reservoir of aerosols in hygro-thermal equilibrium, simulating a long-range transport and mixing from the continent. The source functions representing the aerosol transformations due to condensation of water vapor are derived from the cloud nucleation model of Pruppacher and Klett [1978]. The emphasis is on modeling the role of viscosity (i.e. the Brownian motion) on the fine aerosols in the immediate vicinity of the water surface [Mestayer, 1993].

In the future we aim for the integration of the CLUSA and SEACLUSE models and the introduction of chemical processes into the model. For the validation of the models, experimental data are available from the HEXOS experiments [De Leeuw, 1990]. The data set has been extended with data from the EUROTRAC ASE experiment from FPN in September 1992. TNO and Riso participated in this experiment to measure particle size distribution profiles for particles in the range from 0.065 μm to 100 μm (see De Leeuw et al., this issue).

Principal Results

The principle results from the coordinated efforts 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); some numerical problem has been encountered here which has to be solved before the model is ready for testing with experimental data
- CLUSA has been coded and tested for the vertical profiles of wind speed and turbulent viscosity
- a lower boundary condition has to be found for the removal at the surface of particles of intermediate range
- 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

Main Conclusions

The SEACLUSE model, i.e. the extended CLUSE model for application in open-ocean conditions [Van Eijk et al., 1993], has been formulated and is now ready for testing and evaluation. The experimental aerosol profile data from

the HEXOS experiments [De Leeuw, 1990] will be used for this purpose. For the CLUSA model a satisfactory boundary condition has to be found for the deposition of particles of intermediate sizes. Cooperations to include atmospheric chemistry have been established. This will lead to a comprehensive picture of the aerosol dynamics above water surfaces and the influence of aerosol on physical and chemical processes in the marine atmosphere. 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,
- the formulation of an appropriate boundary condition in the CLUSA model for the deposition of intermediate particles
- integration of the HEXIST/CLUSE/PIE modeling efforts with atmospheric chemistry models to allow for wet phase chemistry and scavenging processes,
- 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; field experimental data from the FPN92 and other ASE experiments will be used.

Acknowledgements

The work performed in 1991 was supported by the Commission of the European Communities (EG DG XII contract STEP-CT90-0047). The TNO efforts were further supported by the Netherlands Ministry of Defence (Contract A92KM615) and the US Office of Naval Research (ONR, Grant N00014-91-J-1948). ECN received additional support from the French joint CNRS/Ministry of Environment committee for EUROTRAC (grant Environnement 92N82/0019). The work performed by Riso was further supported in the framework of the Marine Research Programme in Denmark. Part of the numerical modeling was made while Lex van Eijk held a position as Enseignant Invité at the Ecole Centrale de Nantes. Part of the TWO-PIE efforts by Soren Larsen was made while he held a position as Professeur Invité at the Ecole Centrale de Nantes.

References

- H.J. Dekker and G. de Leeuw, *J. Geophys. Res.* 98 (C6), 10223-10232 (1993).
- G. de Leeuw, *Tellus* 42B, 342-354 (1990).
- G. de Leeuw, S.E. Larsen, P.G. Mestayer, J.B. Edson and D.E. Spiel, Deposition, dynamics and influence of particles in the marine atmospheric boundary layer, *TNO Physics and Electronic Laboratory, Report*, 80 pp. (1991).

- G. de Leeuw, S.E. Larsen, P.G. Mestayer, A.M.J. van Eijk, H. Dekker, A. Zoubiri, P. Hummelshoj and N.O. Jensen, *EUROTRAC annual report 1991*, part 3, section II: ASE, 61-67 (1992).
- G. de Leeuw, A.M.J. van Eijk and H. Dekker, In: *EUROTRAC Annual Report 1992*, part 3 (ASE), this issue (1993).
- J.B. Edson, Ph. D. Thesis, The Pennsylvania State University, University Park, PA, USA, 141 pp., August 1989.
- S.E. Larsen, J.B. Edson, P.G. Mestayer, C.W. Fairall and G. de Leeuw. In: P. Borrell et al. (Eds.), *Proc. of EUROTRAC Symposium '90*. Academic Publ., The Hague, pp. 71-75 (1990a).
- S.E. Larsen, J.B. Edson, C.W. Fairall, G. de Leeuw and P.G. Mestayer. In: P.G. Mestayer, E.C. Monahan and P.A. Beetham (Eds.), *Modeling the fate and influence of marine spray. Proc. of a workshop held 6-8 June 1990, Luminy, Marseille, France*, pp. 155-167 (1990b).
- P.G. Mestayer, In: P.G. Mestayer, E.C. Monahan and P.A. Beetham (Eds.), *Modeling the fate and influence of marine spray. Proc. of a workshop held 6-8 June 1990, Luminy, Marseille, France*, pp. 65-76 (1990).
- P.G. Mestayer, C. Lefauconnier, M. Rouault, S. Larsen, C.W. Fairall, J.B. Edson, D.E. Spiel, K.L. Davidson, E.C. Monahan, D. Woolf, G. de Leeuw, H. Gucinski, K.B. Katsaros and J. DeCosmo. In: W.A. Oost, S.D. Smith and K.B. Katsaros (Eds.) *Proc. NATO Advanced Workshop: Humidity Exchange Over the Sea main experiment (HEXMAX) analysis and interpretation (De Bilt/Epe, The Netherlands, April 25-29, 1988)*, Techn. Rep. Dept. Atmos Sci, AK-40, Univ. Washington, Seattle (WA), pp. 154-163 (1988).
- P.G. Mestayer, In: *EUROTRAC Annual Report 1992*, part 3 (ASE), this issue (1993).
- H.R. Pruppacher and J.D. Klett, *Microphysics of Clouds and Precipitation*, D. Reidel Publ. and Co., Dordrecht, Netherlands, 714 pp. (1978).
- M.P. Rouault, *Modelisation numerique d'une couche limite unidimensionnelle stationnaire d'embruns, These de Doctorat, Univ. d'Aix-Marseille 2, Marseille, France, 1989* (in French).
- M.P. Rouault, P.G. Mestayer and R. Schiestel, *J. Geophys. Res.* C96, 7181-7200 (1991).
- A.M.J. van Eijk, P.G. Mestayer and G. de Leeuw, *Conversion of the CLUSE model for application over open ocean, TNO Physics and Electronics laboratory, report FEL-93-A305, (1993), in print.*

EUROTRAC
(A EUREKA Environmental Project)

Annual Report 1992

Part 3

ASE

Air-Sea Exchange

Subproject Steering Group

Søren E. Larsen (Roskilde) - coordinator
Willy Baeyens (Brussels)
Sauveur Belviso (Gif sur Yvette)
Patrick Buat-Ménard (Gif sur Yvette)
Jean-Louis Collin (Paris)
Olivier F. X. Donard (Talence)
Roy M. Harrison (Colchester)
Gerrit de Leeuw (The Hague)
Peter S. Liss (Norwich)
Spyridon Rapsomanikis (Mainz)
Michael Schulz (Hamburg) - deputy

EUROTRAC
International Scientific Secretariat
Garmisch-Partenkirchen
December 1993