Aerosols in the Coastal Marine Boundary Layer: generation, transport and effects

A contribution to subproject CAPMAN

Gerrit de Leeuw

TNO Physics and Electronics Laboratory
P.O. Box 96864, 2509 JG The Hague, The Netherlands
Phone: +31 70 374 0462; FAX: +31 70 374 0654; email: deleeuw@fel.tno.nl

Summary

Results are presented from research in the coastal atmospheric boundary layer, contributing to the EUROTRAC2 subproject CAPMAN, conducted in the period February 2000 - February 2001. The work in this period focussed on the analysis and interpretation of the results from field experiments (cf. the previous CAPMAN annual report [De Leeuw, 2000] for a brief overview). Atmospheric inputs of nitrogen compounds in the coastal environment, new particle formation, the production of sea spray aerosol from waves breaking in the surf zone and from breaking wind waves, and their dispersal throughout the marine boundary layer are studied using a variety of methods.

Aim of the research

The primary mission of the atmospheric research group at the TNO Physics and Electronics Laboratory (TNO-FEL) is to understand and describe processes in the atmospheric boundary layer affecting the propagation of electro-magnetic radiation, in particular at electro-optic wavelengths. Such processes include scattering and absorption of radiation by aerosols and molecules, refraction due to gradients of temperature and humidity, and effects of turbulence such as scintillation. The studies include a variety of processes connected with air-sea exchange and atmospheric transport, which affect air and water quality and climate. Scales considered range from micro-scale for some of the primary processes, to regional and global studies of aerosols using satellite remote sensing. Coastal areas are special because of the sharp transition in sources, sinks and physical properties at the coastline.

Activities during the year

ANICE. The aim of the ANICE (Atmospheric Nitrogen Inputs into the Coastal Ecosystem) project is to improve transport-chemistry models that estimate nitrogen deposition to the sea. To achieve this, experimental and modelling work is being conducted which aims to improve understanding of the processes involved in the chemical transformation, transport and deposition of atmospheric nitrogen compounds, in particular in the coastal zone. ANICE is an EU project co-ordinated by TNO-FEL and involves NERI and Risø (both DK), the Institute of Inorganic and Applied Chemistry and the Meteorological Institute of the University of Hamburg (Ge), and the University of East Anglia (UK). The initial results from the project were summarised in an overview article [De Leeuw et al., 2001].

The activities of TNO-FEL in 2000 included the further analysis of the experimental data from Meetpost Noordwijk, i.e. aerosol particle size distributions and profiles, bubble size distributions, lidar measurements, and CO_2 fluxes. Lidar measurements were analysed to provide either the height of the boundary layer or cloud base. A statistical analysis was made to derive relations between aerosol concentrations and meteorological parameters. Particle size distribution profiles, for particles larger than about 13 μm in diameter measured with Rotorods [De Leeuw, 1986], were analysed together with bubble spectra to derive information on the bubble-mediated production of sea spray aerosol. This required the re-analysis of the bubble spectra with a new system. In this study, the production of sea spray from bursting bubbles was very simply formulated, and also transport phenomena were not accounted for. New work on these subjects is initiated for 2001. The initial results from the ANICE analyses are presented in more detail in De Leeuw et al. [2000b]. The bubble measuring system used in this work is described in Leifer et al. [2001b], it's calibration in Leifer et al. [2001c] and results obtained with these systems in Leifer et al. [2001c] and De Leeuw and Cohen [2001].

The surf has been determined to be a major contributor to sea spray aerosol at short fetches. Lidar measurements during the ANICE experiments showed that surf-produced sea spray plumes, in off-shore winds, could be discerned over at least 5 km. These phenomena were further evaluated using the Coastal Aerosol Transport model CAT [Vignati, 1999]. In addition, calculations were made on the influence of surf-produced aerosols on the HNO_3 concentrations and profiles at short fetches. The results are published in Vignati et al. [2001].

Surf-produced sea spray aerosol. An important factor in coastal areas is sea spray aerosol produced by waves breaking in the surf zone. Sea spray aerosol sustains specific vegetation in coastal areas, is an aggressive salt causing damage to, e.g., structures and cultural heritage, takes part in heterogeneous processes, and influences atmospheric scattering. Experimental data on the production of sea spray aerosol in the surf zone obtained during the EOPACE projects in California in 1996 and 1997 and an empirical source function are described in De Leeuw et al. [2001]. Quantitative experimental results on the dispersion of surf-produced aerosol were obtained during PARFORCE experiments at Mace Head in 1998 and 1999 [Kunz and De Leeuw, 2000a; Kunz et al., 2001].

Production of sea spray aerosol by breaking wind waves. The most commonly used sea spray aerosol source functions are those from Monahan et al. [19xx] and from Smith et al. [1993, 1998]. The Andreas [1998] source function was derived from Smith et al. [1993]. Comparison with experimental data, both the surf source function derived by De Leeuw et al. [2001a] and data presented in O'Dowd et al. [1997], show that the Smith et al. source function works well for the larger particles it was derived for (>2-3 μm radius), whereas Monahan et al does a good job for smaller particles. However, this same comparison shows that for the smallest particles (<0.1-0.2 μm radius) deviations occur. Therefore simulations were made using CAT to derive a source function that reproduces the complete spectrum presented in O'Dowd et al. [1997]. The result is presented in Vignati et al. [2001]. Work on the bubble-mediated production of sea spray aerosol, from simultaneous measurements of bubble and aerosol spectra during ANICE was described above. New work on this subject has been initiated.

PARFORCE. New particle formation at the coast is studied in the PARFORCE project. TNO-FEL participated with lidar measurements on the structure of the boundary layer and aerosol plumes, as well as with micrometeorological measurements. The experimental work was undertaken during campaigns in September 1998 and in June 1999. Results presented in Kunz and De Leeuw [2000] and Kunz et al. [2001] show that lidar is a powerful tool to visualise plume structures and the evolution of the boundary layer. The micrometeorological measurements were analysed in an attempt to derive relations between the various variances and fluxes and the formation of new particles. Although many occurrences were found in which both the particle concentrations (3 nm and larger) and micrometeorological quantities suddenly changed, the results were not conclusive [De Leeuw et al., 2001]. An extensive micrometeorological characterisation of the Mace Head site has resulted from this work. The PARFORCE work has been finished with the preparation of a series of publications for a PARFORCE special issue in JGR-Atmospheres.

Porquerolles. During the winter of 2000/2001, TNO-FEL participated in experiments at Porquerolles (France) organised by the University of Toulon, with continuous aerosol measurements. Optical particle counters were installed at the Porquerolles site by TNO-FEL, maintenance and data collection were taken care of by the University of Toulon.

Satellite remote sensing of aerosol. Aerosol optical depth (AOD) retrieved from the Along Track Scanning Radiometer 2 (ATSR-2) on the ESA ERS-2 satellite were compared with concentrations of sulphate and nitrate concentrations calculated with the LOTOS model [Builtjes xx]. Thus information could be obtained on the contribution of these aerosol types on the total AOD [Robles-Gonzalez et al., 2001]. Results apply both over land and over the sea. Further, the AOD was assimilated in the LOTOS model, leading to a first and preliminary estimate of the PM2.5 concentration fields over Europe. Ångström parameters obtained from sun photometer measurements over the Baltic during the BASYS experiment were used to derive relations between this parameter and aerosol mass fractions for several aerosol types [Kusmierczyk–Michulec et al., 2001a]. The relations were applied to ATSR-2 data over land to derive fractional concentrations of black carbon. The results compare favourably with available concentrations measured at ground level [Kusmierczyk–Michulec et al., 2001b].

Principal results

The ANICE measurements over the North Sea and the PARFORCE measurements at Mace Head have demonstrated the use of lidar to semi-continuously monitor the atmospheric boundary layer. Thus the evolution and the vertical structure, including mixing phenomena (turbulent and convective) and the transport of plumes produced locally in the surf zone, and at higher wind speed by breaking wind waves, can be studied. This yields useful information for the analysis of chemical and physical phenomena involving the concentrations of gases and aerosols.

The Coastal Aerosol Transport model CAT has been further developed in co-operation with other CAPMAN participants [Vignati et al., 2001]. CAT required an empirical sea spray source function, which was derived from experimental data. CAT was used to evaluate the evolution of aerosol plumes produced in the surf zone, and the subsequent influence of the surf produced aerosol on the total sea spray concentrations at short fetches, the mixing ratio between sea spray and continental aerosol as function of fetch, and the effect of surf-produced sea spray aerosol on HNO_3 concentrations and profiles.

The PARFORCE experiments have resulted in a micro-meteorological characterisation of the Mace Head site. Variations of micro-meteorological quantities appear to occur simultaneously with the onset or ending of nucleation events [De Leeuw et al., 2001].

Satellites offer a powerful tool to retrieve information on the spatial variations of certain aerosol properties, on regional to global scales.

In the reporting period, 4 publications appeared in refereed journals which report on work contributing to CAPMAN, 5 others are in press, 10 have been submitted. Also 4 reports were published. (see section 8).

Main Conclusions

ANICE experimental results summarised in De Leeuw et al. [2000], indicate that the concentrations of gaseous nitrogen compounds (HNO_3 and NH_3) decrease rapidly with increasing fetch resulting in a reduction to ‘background’ levels when the air mass is transported across the North Sea, over a distance of only about 200 km. The gases are highly soluble and are therefore either directly deposited to the surface or taken up by aerosols where they are accommodated through chemical reactions.

The aerosol dry deposition flux is in part due to different physical processes and depends on particle size, and thus the dry deposition velocities for gases and particulate nitrogen compounds are different. Moreover, the direct gas fluxes are determined by the partial pressure difference of the gaseous species in the water and in the air directly above the water, both of which have been observed during the ANICE experiments to vary strongly in both space and time. In model calculations such variations, especially those in the sea, are usually not taken into account. Neglecting spatial variations may lead to significant overestimation of dry deposition of NH_3 [De Leeuw et al., 2000].

Model calculations with CAT [Vignati et al., 2001] show that surf-produced aerosol has a large influence on the sea spray concentrations in the coastal marine atmospheric boundary layer, and thus on the relative contributions of continental and marine aerosol. Furthermore, sea spray aerosol is produced in the surf zone in very high concentrations that immediately at the coast line are available for heterogeneous chemical reaction, and thus influences, e.g., atmospheric input of nitrogen in coastal waters. Accounting for surf-produced sea spray aerosol, the large influence of the reaction between nitric acid and sea spray on both the concentrations and the gradients of HNO_3, over a fetch of only 25 km has been demonstrated. This leads to the conclusion that surf produced aerosol cannot be ignored and needs to be accounted for both in modelling and in experimental work.

LIDAR techniques provide an efficient way to determine BL structure and evolution with high spatial and temporal resolution, as well local generation and evolution of aerosol plumes [Kunz et al., 2001].

The micrometeorological characterisation of the Mace Head Atmospheric Research station (Ireland) [De Leeuw et al., 2001] shows significantly different characteristics for different wind directions, reflecting the terrain influence on the roughness length and related quantities. The effect of surface roughness further causes a tidal effect on, e.g., the drag coefficients in on-shore wind. The sloping terrain results in a tilt of the air flow, and thus has a strong effect on the direction of the derived fluxes. The results further suggest the occurrence of an internal boundary layer in on-shore wind directions. Diurnal variations are visible in parameters such as the air temperature and relative humidity and their respective variances, and the associated heat and water vapour fluxes. The diurnal patterns are disturbed by larger scale meteorological processes, such as the passage of frontal systems. The influence of the (micro-) meteorological situation on other atmospheric processes is illustrated for new particle formation.

Aim for the coming year

In 2001 the ANICE work will be finalised with a final report, summarising and integrating the work by the participating institutes (see section 2). Bubble and aerosol measurements at open sea will be analysed to obtain detailed information on the sea spray source functions. The analysis of EOPACE data from California and North Carolina is expected to be completed in 2001. A start will be made with the analysis of the Porquerolles data. New experimental work is in preparation. Together with the University of Stockholm work is undertaken on the production of sea spray aerosol based on laboratory measurements of bubbles and aerosols. Also direct aerosol flux measurements are planned as part of this cooperation, during a field experiment near Hawaii in August/September 2001. Lidar, aerosol and bubble measurements will be made from FLIP. Several publications are foreseen.

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