

EUROTRAC-2  
(A EUREKA Environmental Project)

# CAPMAN

Coastal Air Pollution Meteorology and  
Air-Sea Nutrient Exchange

Annual Report 2000

**Subproject Coordinator**

Gerald Geernaert, National Environmental Research Institute, Roskilde (DK)

**Steering Committee**

Kevin Noone, University of Stockholm (SE)  
Serge Despiau, University of Toulon, La Garde (F)  
Søren Larsen, Risø National Laboratory, Roskilde (DK)  
Gerrit de Leeuw, TNO-FEL, Den Haag (NL)

*International Scientific Secretariat (ISS)*  
*GSF – National Research Center for Environment and Health*  
*Munich, Germany*  
*August 2001*

## Table of Contents

<b>I.</b>	<b>Report on the work of the subproject</b>	<b>1</b>
1.	Summary	1
2.	Aims of the period's work	2
2.1	Similarity theory, fetch dependent BL's, and the IBL	2
2.2	Heterogeneous processes and the fluxes of reactive gases and aerosols	3
2.3	Mapping the relative atmospheric contribution to coastal waters	4
2.4	Flow fields, air concentrations and deposition patterns in compliance studies	4
3.	Activities during the year	5
3.1	Kattegat Strait, between Denmark and Sweden	5
3.2	North Sea studies	5
3.3	Italian coastal studies	6
4.	Principal experimental or modelling results	6
4.1	Similarity theory, fetch dependent boundary layers, and the IBL	6
4.2	Towards improved parameterizations of nutrient fluxes	7
4.3	Mapping nutrient inputs to coastal waters	8
4.4	Italian coastal studies	9
5.	Main conclusions	9
5.1	Extending similarity theory and its applications	9
5.2	Heterogeneous processes and the air-sea fluxes of aerosols and reactive gases	9
5.3	Flow fields and deposition patterns	10
6.	Policy-related results	10
6.1	Regulatory models applied to coastlines	10
6.2	Impacts of atmospheric load on marine eutrophication	10
7.	Aims for the coming year	11
8.	Acknowledgements	12
<b>II.</b>	<b>List of authors and titles of reports and theses resulting from the subproject's work</b>	<b>13</b>
<b>III.</b>	<b>List of publications in the refereed literature from the subproject</b>	<b>13</b>
<b>IV.</b>	<b>Reports from the principal investigators</b>	<b>16</b>
	<b>ANNEX: Names and details of current principal investigators</b>	<b>61</b>

## **I. Report on the work of the subproject**

### **1. Summary**

Atmospheric deposition is a major input route for pollution to coastal seas around Europe. Depending on time of year, nutrients may increase biological activity of phytoplankton and toxic algae in the surface layer of the sea, causing changes in the composition of the benthic fauna and flora, and affect the oxygen consumption within both the water column and sediments. Data on atmospheric fluxes of nutrients, especially inorganic and organic nitrogen and phosphorus compounds in the gaseous and particulate phases, are rather limited and sometimes even conflicting. Therefore, quantifying the inputs of various nutrient compounds including fixed nitrogen and trace metals to coastal waters is needed. In particular, there is a need to understanding the processes regulating these inputs and to describe their impacts on the biogeochemistry of surface waters.

During recent years, much research work has been carried out and new questions raised in the field of atmospheric deposition. Trace elements such as Al, Si, Mn and Fe are documented to be increasingly important due to their biological roles. The bio-availability of these trace elements is therefore also included among the investigations carried out under CAPMAN. In addition, Cr, Ni, Cu, Zn, Cd, and Pb, all defined by the North Sea Conference as toxic components with high priority, are given full attention as well, with special focus on the source-transport-sink inventories of Europe's coastal seas.

The anthropogenic activity responsible for chemical deposition to marine ecosystems, as well as local and regional air pollution episodes, can in most part be traced back to emissions from industrial, agricultural, and commercial enterprises. Understanding the source-receptor relationships, and assessing the benefits of emission reductions on coastal environmental quality, require the construction and implementation of well functioning atmospheric chemistry and transport models. Models must be capable of discerning the necessary spatial resolutions for local authorities, and they must also be capable of separating regional versus long range transported emissions as causes of local adverse conditions (e.g., whether air pollution exceedances or marine eutrophication). As models are developed, the appropriate technology must be in place to provide testing and validation.

In order to support the needs of coastal zone managers and policymakers, and to extend the atmospheric sciences under the frame of EUROTRAC-2, the overall aim of CAPMAN is to improve the science base necessary to extend the quality of relevant data bases, and extend the applicability and performance of coastal models. This aim is supported by four objectives:

- (a) extending the understanding of atmospheric dynamics and chemical transformations in coastal circulations;
- (b) extending the understanding of aerosol mass closure and the role of organics;
- (c) extending the understanding of the physical and chemical processes governing the air-sea exchange of nutrients (and related parameters and compounds) over the coastal sea; and
- (d) extending our understanding of source-receptor relationships at various sites in coastal zones.

Research reported under CAPMAN during 2000 included advances in theoretical frameworks, experimental techniques, and modelling approaches which are needed to improve estimates of surface exchanges in coastal regions. The study regions reported for

2000 include the North Sea (De Leeuw), Kattegat Strait between Denmark and Sweden (Sørensen, Jickells, Sempreviva), Baltic Sea (Smedman), and the Italian coastal zones of the Mediterranean Sea (Finardi). Theoretical work focussed on generalizing the flux profile relations for all chemical compounds (Geernaert), exploring dynamical relationships between swell waves and the air-sea momentum flux (Smedman), and on examining the rate of air mass transformation associated with internal boundary layer growth (Dunkerley, et al.).

Experimental studies during 2000 focussed on a major campaign in the Kattegat between Denmark and Sweden (Sørensen, Jickells, Sempreviva), which combined measurements from coastal and island stations to model derived data bases for the region. Analysis of both past and new data sets gathered in the Mediterranean, Baltic, and North Seas were integrated into the analysis, in order to generalize the results to other regions. There were continued field campaigns on the island of Gotland (Smedman), in order to study the interaction between swell waves and both the Monin-Obukhov similarity theory as well as its applications. In addition, there were continued field measurements in the Belgian and Dutch parts of the North Sea, as part of a long term effort to explore the controls over air-water exchange of nutrients, micro-pollutants, and trace metals (Van Grieken).

Modelling of air-sea inputs placed focus on the development of heterogeneous chemistry submodules, exploring various dynamical modelling techniques able to address the complex conditions of coastal zones, and assessing a variety of air quality regulatory models for use in assessing impacts of industrial and agricultural emissions in coastal zones. In support of the major field study in the Kattegat during 2000, the American model COAMPS was integrated into the modelling efforts (Svensson and Tjernström). The COAMPS model was integrated into the modelling activities in order to extend the performance of model systems applicable to the Skagerrak and Kattegat region.

Specific modelling projects were designed to examine a variety of related coastal issues. The seasonal variation of nutrient deposition to coastal seas was explored for the Danish coastal waters, with a view towards identifying the relative importance of atmospheric deposition to adverse marine biological states (Hertel). In Italy, studies were conducted in order to determine the relative importance of long range transported pollution, with a view towards new emissions control policies for northern Italy involving industry (Finardi). Finally, internal boundary layer modelling was carried out for the west coast of Denmark, with a view towards improving the parameterizations associated with air mass modification during on-shore flow (Dunkerley, et al).

## **2. Aims of the period's work**

### *2.1 Similarity theory, fetch dependent boundary layers, and the IBL*

In order to construct parameterizations which accurately describe air-surface exchange, it is necessary that both meteorological and surface processes are formulated within a consistent mathematical framework. Monin-Obukhov similarity theory presently provides the working paradigm for flux parameterizations, which in turn is based on a set of assumptions, e.g., spatial homogeneity, steady state conditions, and a static surface. Unfortunately, the coastal ocean contains a rapidly evolving fetch-dependent wave field, internal boundary layers, sharp gradients of stratification and windspeed, and occasionally there are swell waves propagating on the surface. These and other coastal characteristics have posed problems for use of the existing similarity theory, thus resulting in unacceptably large uncertainty and lack of confidence in the presently available flux parameterizations.

There were three objectives governing research under CAPMAN during 2000:

- (a) revise the theory for fetch-dependent surface fluxes of momentum, heat, gases, and particles, by simplifying the complicated equations by incorporating real measurements over the North Sea (Geernaert);
- (b) document the systematic influences of swell on the fundamental equations of Monin-Obukhov similarity theory, and explore the dependence of the momentum and heat flux coefficients on the presence and direction of swell (Smedman, Rutgersson, Högström); and
- (c) further analysis of existing measurements collected on a sequence of masts extending inland from the coastline of Denmark, in order to produce improved parameterizations of internal boundary layer slope statistics with a view of building a better performing IBL model during advection and air mass transformations for onshore flow (Dukerley, Sempreviva, Larsen, Mikkelsen).

## 2.2 *Heterogeneous processes and the fluxes of reactive gases and aerosols*

Sea salt reactions have been demonstrated to strongly influence the air-sea exchange rates of nutrient compounds, namely ammonia, nitric acid, and nitrate (Sørensen). In addition, a simple model was constructed and published during 1999 which shows that sharp horizontal gradients over the coastal zone produce sharp spatial inhomogeneities which must take heterogeneous processes into account (Vignati). There were three objectives in 2000, to continue this work:

- (a) extend the performance of atmospheric transport and chemistry models, by updating the chemical schemes and model physics (Hertel, Schlünzen);
- (b) measure fluxes of ammonia and nitric acid using the relaxed eddy accumulation technique at several levels, in support of the first phase of the EU financed MEAD project, in the Kattegat (Sørensen), and relate these results to total nutrient deposition and biological productivity (Jickells); and
- (c) measure the fluxes of aerosols in coastal zones, with a view towards describing in detail the processes responsible for fluxes during coastal breaking conditions and processes associated with bubbles and spray (De Leeuw).

In support of the first objective, heterogeneous processes were to be further described in terms of improved parameterizations and submodels, for integration into operational mesoscale air pollution transport models. It was also expected that the models would be demonstrated in support of the CAPMAN field programs. During the 2000 period, the model ACDEP (Hertel) was run in its improved form (including a more advanced chemical scheme) for the Danish coastal waters. In addition, the METRAS/MECTM model of the University of Hamburg model was upgraded with more advanced chemistry submodules, and it was run for an early summer period for the North Sea, as a hindcast for 1998, to simulate complex atmospheric flows and deposition patterns over the coastal North Sea.

For the second objective, it was expected that the MEAD experimentalists would carry out a detailed field study of the interaction between air-sea fluxes and biological controls, as they apply in the Kattegat Strait between Sweden and Denmark. This was to involve direct dry flux estimates of ammonia and nitric acid, as well as wet deposited measurements of a host of

other nutrient compounds. The campaigns had an intensive period during early summer, and a less intense period extending over many months.

Concerning aerosols, there is a continuing objective to describe the processes associated with aerosol emissions in coastal zones. During recent years, the vertical sheet of aerosols emitted from coastal breakers as observed with lidars has added a new dimension to the problem of determining coastal aerosol budgets. The objective is therefore to consider these new observations of aerosols from breakers in the generalized sense, where future parameterizations of spray and bubble processes can be applied to both general wave breaking and to surf conditions.

### *2.3 Mapping the relative atmospheric contribution to coastal waters*

In most previous studies, the atmospheric load of nutrients to coastal waters has been treated as a secondary source relative to river input. In many regions, however, the improved technology applied to waste water has reduced the riverine contribution. An objective in 2000 was to test the hypothesis that late summer atmospheric deposition events in eutrophication prone regions may be critically important. The work in 2000 was to carry out a major field campaign during the spring through summer seasons, and to apply improved atmospheric deposition models for the entire Skaggerak and Kattegat domain. The use of models was to specifically include improved versions of the chemical submodels and emission inventories, so that trends and risk assessments may be performed with improved confidence (Hertel).

Because the flow field is highly complex over the Kattegat region, it was recognized that it was necessary to apply several sophisticated three dimensional Eulerian transport and air chemistry models to the domain, in order to determine the spatial variability of deposition associated with (1) coastline geometry and topography associated with the Kattegat, and (2) pattern of anthropogenic emissions. An objective in 2000 was therefore to assess the influence of offshore islands and coastline geometry as controls over the spatial variation of deposition (Svensson and Tjernström).

### *2.4 Flow fields, air concentrations and deposition patterns in compliance studies*

In many studies to assess compliance of various types of industries to air quality directives, modelling must be used to determine the types of environmental conditions associated with normal and exceedance events. In relatively homogeneous regions, Lagrangian transport models with simple physics and chemistry are used. In more complicated regions, e.g., coastal zones and mountainous terrain, much more sophisticated models may be required. There is therefore an objective to determine the level of model sophistication which is necessary, in order to be used by the regulatory agencies in coastal regions to test compliance (Finardi).

For the work in 2000, the specific objective was to compare a standard Lagrangian dispersion model with a more sophisticated three dimensional Eulerian transport and dispersion model. This comparison was anticipated to determine the differences in performance and accuracy, in particular for a variety of environmental conditions associated with exceedances and noncompliance.

### 3. Activities during the year

The activities during 2000 were dominated by regional studies.

#### 3.1 *Kattegat Strait, between Denmark and Sweden*

During 2000, the EU funded project MEAD (Marine Effects of Atmospheric Deposition) conducted its first major field campaign in the Kattegat Strait. Coastal stations were established at Bua (Swedish coastline), and additional stations were installed at Læsø (island in the Kattegat), and on the east coast of Jutland. The three stations provided meteorological data (wind vector, temperature, fluxes), and measurements of fluxes and/or concentrations of nutrient gases and aerosols were also provided by the three stations. During a roughly two week period during the summer season, sampling of marine biomass was also carried out by a ship during transects over the region.

To support the field measurements, two independent modelling efforts were carried out. Hertel (DK) used a more refined version of the ACDEP Lagrangian transport model, with improved chemistry, to map the wet and dry deposition of ammonia to the waters of the Kattegat region. The model framework included improved emission inventories, e.g., emissions from ships in the Strait were added. He also observed that there has been a decrease of riverine input to the domain in recent years, thus adding importance to the role which atmospheric deposition plays as a cause of episodes associated with poor water quality and eutrophication.

The second modelling activity (Tjernström and Svensson) applied to the Kattegat was to study the complex wind fields and chemical deposition patterns associated with irregular coastline geometry and the shadowing effects caused by the presence of islands (e.g., Læsø) in the Kattegat. Their approach was to use Eulerian models, and to explore the differences and advantages of both the Swedish meteorological model at MIUU/SU and the US Navy model COAMPS.

#### 3.2 *North Sea studies*

In recent years, the Belgian group (van Grieken) has made great strides in the development of sample preparation methods which require trace amounts of aerosol. This included a leaching system with a quantitative recovery of the species of interest, requiring extensive testing during field campaigns. During 2000, the methods developed by this group were used to gather a larger inventory of data over the North Sea, with a view towards calculating the fluxes of a variety of compounds, including Cl, NO<sub>2</sub>, NO<sub>3</sub>, SO<sub>4</sub>, F, Na, NH<sub>4</sub>, K, Mg<sub>2</sub>, and Ca<sub>2</sub>. There were four field campaigns, where the duration of each was of order one week.

As a follow-up to measurements carried out in 1998 and 1999 under the EU financed ANICE project, there was research on aerosol production, deposition processes, and model development. Analysis of the data proceeded into 2000. Further analysis was reported by De Leeuw (NL) based on a new set of measurements collected on the Meetpost Noordwijk (MPN) throughout 2000.

The aerosol research tasks, carried out by De Leeuw, made great efforts to entrain results from similar locations into their analysis. The EOPACE and the Irish based PARFORCE experiments from 1999 amassed enormous data sets on aerosols, optical properties, and supporting meteorology. EOPACE was carried out off the California coast, and PARFORCE

was conducted near Mace Head (Ireland). In both studies, the general aim was to improve the understanding of new particle formation and subsequently improve parameterizations. As in the North Sea studies, the results of the EOPACE and PARFORCE campaigns highlighted a large degree of spatial variability which could be resolved by a combination of lidar scans and time series analysis of fixed-site data sets. The results and conclusions of those studies began to emerge in 2000.

Further up the coast of the North Sea, i.e., on the Danish Jutland coastline, the earlier works of Sempreviva and Larsen to describe the evolution of the internal boundary layer were revisited and extended by an extended version of the original team, i.e., Dunkerley, Sempreviva, Larsen, and Mikkelsen. In their analysis of the data of the JYLEX project, IBL slopes were statistically analyzed within a dynamical framework, to resolve seasonal differences and/or influences of fetch and stability at different locations.

### 3.3 *Italian coastal studies*

The coastline of Italy is quite variable in terms of its coastline geometry and degree of complex topography. Superimposed on these natural features are local regions of high industrial activity and other regions of open untouched nature. In order to support the regulatory process, there has been a dramatic proliferation of modelling approaches, ranging from the simple more traditional Gaussian dispersion models to the more sophisticated three-dimensional meteorological and air chemistry models. Gaussian models are preferred for their ease of use, though it has become clear that some regions require a more sophisticated type of modelling approach to satisfy regulatory needs.

To address these concerns in 2000, Finardi, et al (Italy) began the process by first comparing the Lagrangian particle model SPRAY to the US-EPA regulatory model ISC3ST. The RAMS model, extended down to 1 km resolution based on grid nesting techniques, was used as the driver for the regulatory models. Finardi, et al., applied the models to two locations, i.e., the flat terrain characterising Fusina (near Venice), and more complex terrain characterising Vado Ligure.

## 4. **Principal experimental and modelling results**

### 4.1 *Similarity theory, fetch dependent boundary layers, and the IBL*

In a study carried out by Sempreviva, Frank, and Larsen, data collected during the JYLEX experiment were first analyzed in 1998 to explore the processes which account for spatial variability. This effort was based on four stations (32 m tall masts) which had been placed on a line extending directly inland from the coast, starting with the first mast at the coastline and the fourth mast located 30 km inland. Data analysis from these masts was designed to infer the rates of air mass modification, with special focus on adjustments of windspeed, temperature, and stratification. In 1999, the data were subsequently used to validate the performance of the University of Karlsruhe KAMM mesoscale model. Overall, the results of the validation exercise in 1999 demonstrated reasonably close agreement for modelled and measured windspeeds for neutral to unstable conditions. The model, however, tended to significantly underpredict the rate of temperature change, for fetches greater than 10 km.

In 2000, the JYLEX results were revisited. Dunkerley, et al., examined the two dimensional potential temperature profile over land downstream of the coastline, in order to explain variability in both the across and along shore directions. Dunkerley derived a first order



differential equation from a simplified form of the heat equation, using constant entrainment. Using JYLEX data, they were able to statistically infer the coefficients needed to produce a working IBL growth model, though for a restricted range of atmospheric stratifications.

Also in 2000, de Leeuw demonstrated with additional data that wave breaking in the surf zone produces a substantial portion of marine aerosol, even during light windspeeds. His calculations implied that the surf generated aerosol constitutes a major fraction of the coastal zone's aerosol production, i.e., extending from the coastline out to several tens of kilometers offshore.

In 2000, Geernaert demonstrated that the equation which relates the surface layer flux divergence of momentum and gases to spatially varying quantities can be simplified to a sole dependence on horizontal windspeed gradients, under most conditions. This simplification was based on observed spatial variabilities of wind and other surface data in the North Sea and Kattegat. He furthermore demonstrated that this simplification of the flux divergence equation allows a more operational way to wind power in coastal zones (where horizontal gradients are taken into account), and the finding of preliminary analysis shows substantial differences in wind power potential when compared to existing techniques.

In 2000, Smedman, Rutgersson, and Högrström added a serious complication to the application of Monin-Obukhov similarity theory. Their further analysis of meteorological and wave data from Gotland reinforced the view that swell waves have a dramatic influence on the drag coefficient and Stanton number (heat exchange coefficient). They demonstrated that the friction velocity is strongly reduced during swell conditions, and that this will act to most likely also affect the degree of atmospheric stratification via changes in the heat flux. Because the friction velocity affects all fluxes, deposition velocities (by similarity) will be reduced.

#### 4.2 *Towards improved parameterization of nutrient fluxes*

In order to improve parameterizations and estimates of nitrogen fluxes to the coastal ocean, there were efforts to incorporate and assess the role of chemical reactions in parameterizations which estimate dry deposition of reactive gases. Sørensen used the results of previous works to design an experiment to test hypotheses which involved direct flux estimates of ammonia and nitric acid, at several levels on a coastal mast. Measurements were carried out at Bua (Sweden) as part of the first intensive field campaign, during summer 2000.

Based on analysis of process interactions and supported by field data from the Kattegat, the team of Jickells, Baker, Cornell, Spokes, and Yeatman demonstrated that the deposition rate of fixed nitrogen depends critically on the size distribution of the component of interest in the aerosol. They further demonstrated that, in the coastal zone, air masses of marine and continental origin are rapidly altered, and that the alteration is derived primarily from the interactions of gas phase nitric acid and ammonia and fine mode aerosol salts in the air column. Because the size spectra are altered during this process, deposition rates will be altered as well.

Jickells, et al., also analyzed data from the west coast of Ireland, which were relevant to the MEAD air-sea chemistry studies. The Irish study showed that patterns of distribution of trace gases, such as methyl iodide, DMS, and isoprene are uniquely different, thus suggesting different biological and chemical mechanisms by which these gases are formed and released from the water column.

Data collected by the Belgian group (van Grieken, et al) showed the same type of marine and continental air mass mixing as was observed by Jickells et al. However, the Belgian group noted very high concentrations of ammonia during summer (over the North Sea). The seasonal patterns of concentrations and deposition reported in 2000 appear to corroborate previous findings.

The ANICE (EU funded 1997-2000) and PARFORCE projects reinforced the utility of lidar as a valuable tool in mapping aerosols over the coastal zone. These two projects, i.e., in the North Sea and off Mace Head, respectively, yielded valuable data derived from lidar. The data show a time history of aerosol sheets and plumes from breaking waves in the surf zone, thus adding key information on surface and boundary layer processes responsible for aerosol mixing in the coastal zone.

#### 4.3 *Mapping nutrient inputs to coastal waters*

In recent years, Hertel, et al., presented estimates of the nutrient load to various coastal regions of Denmark and adjacent countries. Model calculations indicated that the atmospheric deposition constitutes roughly 2/3 of the total nutrient load to fjords and bays. For the more open waters such as the Kattegat Strait, the atmospheric load is responsible for approximately 40% of the total nutrient content of the upper water column. During 2000, Hertel also reported that the proportion of riverine nutrient input is decreasing, and that the atmospheric deposition is most likely underestimated due to ship traffic data missing in the emission inventories. A further improvement to the Danish ACDEP system was produced in 2000. The improved version is based on use of ETA model input (from the THOR forecast system), which in turn produces substantially improved estimates of rainfall over the Danish waters than previous meteorological drivers. Another modification of ACDEP was also introduced in 2000, i.e., an improved treatment of chemistry and vertical diffusion and deposition. Model performance and validation exercises carried out during 2000 showed improved performance.

Hertel's study in 2000 also reinforced predictions made in 1999. At that time, he stated that the atmospheric load is generally the dominant transport pathway during the late summer and early autumn in northern Europe. Eutrophication events which occur during the late summer and early autumn in the Kattegat region are furthermore hypothesized to be the consequence of anomalously large atmospheric deposition events, either due to rainfall or a combination of rainfall and dry deposition. Unlike during spring and early summer, river input of nutrients to coastal marine regions is relatively unimportant. The MEAD project and its intensive field campaigns in 2000 and 2001 are designed to test this hypothesis (Jickells, Sørensen, Geernaert, Hertel, Sempreviva).

In order to extend the accuracy of nitrogen flux estimates using regional and basin scale models, Schlünzen, et. al., added heterogeneous chemistry to their model system, which in turn combines the METRAS meteorological model with both the MECTM chemical transport model and an aerosol model SEMA. The model system was applied to hindcasted episodes in 1998, over the North Sea, in support of the ANICE project. One finding of the studies was that model performance is highly sensitive to the roughness length on both land and sea, insofar that the difference of roughness length on the two sides of the coastline acts as a control on cross-shore acceleration, which in turn governs the deposition velocities.

During 2000, the meteorological complexity of the Kattegat region required that much greater emphasis be placed on the three dimensional structure of wind fields and deposition patterns

over the region. Tjernström and Svensson demonstrated that the 100 km wide region of the Kattegat will rarely exhibit conditions where the wind fields are undisturbed by the upwind coastline. They also demonstrated that turbulent transport is greatest over land (due to generally unstable conditions), and that there will be a general reduction of the momentum flux by nearly 50% over the coastal zone relative to open waters. They also demonstrated that sea breezes and low level jets play significant roles in the distribution of tracers over the region. Finally, the island of Læsø in the middle of the Kattegat was determined to have sufficient size to influence the evolution of the downstream boundary layer. Læsø was given responsibility for the formation of a momentum wake downstream, and flux patterns over the entire region are affected.

#### 4.4 *Italian coastal studies*

In the Italian regulatory community, one objective is to determine the level of sophistication of atmospheric dispersion models which are necessary to address compliance issues, i.e., involving the agricultural, industrial, and commercial emission sectors. Two sites were of focus during 2000, i.e., the relatively flat coastal region of Fusina, and the topographically more complex region of Vado Ligure. Model comparisons demonstrated that simple models are sufficient in the relatively flat coastal domain of the Venice region. However, this was not the case for Vado Ligure. For Vado Ligure, simple models produce different regions of high pollution concentration than what one obtains from more sophisticated models. In both cases, it was determined that RAMS correctly reproduces the sea breeze down to scales of one kilometer, and it was concluded that RAMS should continue as the meteorological driver for the regulatory models, regardless of the degree of topographic complexity.

### 5. **Main conclusions**

#### 5.1 *Extending similarity theory and its applications*

Based on an extension of Monin-Obukhov similarity theory and comparison to field observations from the ANICE project, Geernaert demonstrated that the flux divergence equation can be simplified by including only the horizontal windspeed gradient.

Smedman, et al., demonstrated that while Monin-Obukhov similarity theory works well in coastal and offshore zones during conditions of no swell, their data convincingly show that during conditions of swell (which occur more than 40% of the time at the Gotland site) that the drag coefficient and Stanton number are significantly reduced from their open ocean values.

Dunkerley et al produced a new two dimensional model which estimates the two dimensional growth of the thermal internal boundary layer, during onshore flow over relatively flat terrain. The model was developed and tested for a variety of fetches and stability conditions using towers on the west coast of Jutland (Denmark).

#### 5.2 *Heterogeneous processes and the air-sea fluxes of aerosols and reactive gases*

In order to test the hypothesis that ammonia and nitric acid will always exhibit a significant flux divergence, a first-of-its-kind field experiment was designed and carried out (by Sørensen) involving direct chemical flux systems (using the relaxed eddy accumulation or REA method) at more than one height on a mast. The REA flux systems were installed on a coastal research mast at Bua (Sweden) in the summer 2000.

In addition to quantifying the significance of atmospheric nitrogen deposition to phytoplankton life cycles, Jickells, et al., produced an improved description of the interactions between sea salt, nitric acid and ammonia. They also reported for the first time speciation, size distribution and composition of organic nitrogen in the remote marine atmosphere. In the studies leading to these conclusions, Jickells demonstrated and applied a newly developed technique to use nitrogen isotopes for tracing the sources and transformations of nitrogen in the coastal marine atmosphere.

The ACDEP model (Hertel) was improved, with a more advanced heterogeneous chemistry package, and the meteorological driver was improved with the incorporation of the ETA meteorological driver.

Surf aerosol (de Leeuw and Vignati) was demonstrated to have a significant influence on the total coastal zone aerosol budget. This finding was based, in large part, on the use of lidars to map the emissions and transport of aerosol sheets from wave breaking in the surf zone and subsequent transport of sheets and plumes into the boundary layer.

### *5.3 Flow fields and deposition patterns*

Based on application of two meteorological models, Tjernström and Svensson demonstrated that the main features of an offshore blowing air mass will be strongly influenced by the upwind coastline for a distance from the coastline exceeding 100 km. In this region, both the momentum and heat fluxes will exhibit dramatic changes at the coastline. It was furthermore shown that the island of Læsø in the Kattegat Strait introduces significant dynamical influences on the boundary layer structure in the region, i.e., with strong influence over sea breezes, air mass modification, low level jets, and surface deposition patterns over the region.

## **6. Policy-related results**

### *6.1 Regulatory models applied to coastlines*

It is not unusual to find that coastal zones contain a substantial fraction of a region's industries, agricultural and commercial activities. In order to regulate air quality, and in order to monitor compliance to regional and EU policies, it is essential that agencies use meteorological models which are capable of handling the topographic and coastal features of the domain. Finardi, et al., embarked on a study to assess the modelling requirements for Italy's northern coastal zones, by comparing the performance of two different models in two different coastal sites. In their 2000 activities, the two regions selected included: the relatively flat region of Fusina and the topographically complex region of Vado Ligure. The results indicated that simple models are sufficient in the flatter regions. On the contrary, it was also concluded that a higher level of sophistication was necessary in the hilly Vado Ligure region to obtain the desired level of model accuracy.

### *6.2 Impacts of atmospheric load on marine eutrophication*

During much of the summer and early autumn, many coastal zones of Europe experience excessive nutrient input, resulting in risks of marine eutrophication and/or oxygen depletion in its bottom waters. There has therefore been great emphasis in coastal zone policy development to identify control measures for nitrogen emissions to rivers, streams, and the atmosphere. In general, the perception has been that roughly 60% of the nutrient input to coastal waters originates from riverine pathways, and the rest is deposited from the

atmosphere. This perception is derived from monitoring studies, which average data over a number of years.

More recently, there has been a new research direction to build forecast systems, which operate on shorter time scales (order of days to weeks) able to predict adverse marine effects (e.g., eutrophication and/or oxygen depletion). During 1999, the ACDEP model (Hertel) was used to identify the ratio of atmospheric to total nutrient load to offshore regions during the course of the spring, summer and autumn seasons. It was found that during the period August-October, the majority of nutrients which enter the water column originate via the atmosphere, and deleterious states in the marine water column are suspected to be attributable primarily to atmospheric deposition. A major field program was initiated in 2000, financed by the EU (MEAD project) to explore the role which the atmosphere plays as a risk to marine water quality. In the meantime, the ACDEP model has been upgraded with both its meteorological driver and its chemical scheme, and there has furthermore been refinement of the emission inventories. It is anticipated that one of the results of MEAD (in 2002 and/or in 2003) will be a recommendation to adjust the time scales of future marine monitoring programs according to season and location, so that the data bases conform to the input needs of models designed to assess and/or forecast adverse states. The specific details of such a recommendation are awaiting further scientific analysis.

## **7. Aims for the coming year**

The similarity theory for air-sea fluxes, which takes into account quasi-homogeneous conditions, will be integrated into the meteorological model at the University of Stockholm, and subsequently tested against MEAD data in the Kattegat region. (Geernaert, Tjernström).

Further data collection concerning the influence of swell waves on Monin-Obukhov similarity theory will be carried out on the island Gotland. The data will be used to understand wave processes which are responsible for reduced drag and heat exchange coefficients. (Smedman, et al)

The second major field campaign under the MEAD project will be carried out in the Kattegat Strait. This campaign will be similar to that conducted during summer 2000, i.e., involving a coastal station in Jutland, Læsø, and Sweden, and use of a ship for carrying out transects over the domain. (Jickells, Spokes, Sørensen, Sempreviva).

Meteorological model development will be carried out in order to explore the roles of spatial variation, irregular coastline topography, and preconditioning in reference to observed algal blooms (Tjernström, Svensson). This will be parallel to further development of the ACDEP model, with eventual conversion to the higher resolution Eulerian REGINA model (to be implemented most likely in 2002) (Hertel).

Aerosol and gas concentration and flux data will be analyzed and interpreted from a variety of field experiments, i.e., the ANICE, BASYS, EOPACE, FETCH, ACSOE, MEAD experiments. This analysis will integrate available results regarding gas transfer and bubble interactions in the laboratory. In addition, bubble and aerosol measurements from the open sea will be analyzed in order to obtain detailed information on the sea spray source functions (de Leeuw, Sørensen, Jickells, Despiou).

Specifically for MEAD and hindcasting of ANICE events, the importance of aerosol formation in determining downstream deposition rates of both gases and aerosols will be

assessed using three-dimensional chemistry runs with the model system METRAS/SEMA (Schlünzen) and the ACDEP model system (Hertel).

The Belgian group is planning two campaigns in 2001, which focus on continued sampling of air-sea fluxes. (van Grieken).

For internal boundary layer studies, the analytic form of a two dimensional IBL growth rate equation will be extended to test a wider range of atmospheric stratifications. (Dunkerley, Sempreviva, Larsen, and Mikkelsen).

The comparison of regulatory models for use in different coastal sites of northern Italy will be extended in 2001, to consider photochemical pollution in coastal sites of southern Italy.

## **8. Acknowledgements**

The theoretical and experimental work on similarity theory, boundary layer parameterizations, and IBL air mass transformation carried out in Denmark were financed by the EU project MEAD and internal contributions from Risø National Laboratory and the National Environmental Research Institute. Swedish work on the influence of swell on surface fluxes was supported by the EU funded project PEP in BALTEX, and Swedish modelling work additionally acknowledges the US Naval Research Laboratory for use of their COAMPS model. The EU is also recognized via the ANICE project for providing funding for experimental research in connection to measurements on the Noordwijk Platform and the Weybourne site in the UK during the early part of 2001. Modelling carried out in both Germany and Denmark, was funded by the EU under the ANICE project, with additional support provided from the University of Hamburg and NERI Denmark. The Dutch participants also recognize financing from the Netherlands Ministry of Defence, European Commission, US Office of Naval Research, and TNO internal funding.

Belgian research on sampling techniques and field cruises into the North Sea were financed by the Belgian Prime Minister's Services – Office of Scientific, cultural, and technical affairs.

Jickells, et al., acknowledge the MEAD Project (EU financed) and NERC for supporting their contributions in 2000.

The research in the greater Ravenna region acknowledges Dr. Marco Cane and Dr. Valerio Strocchi, of ARPA-Sezione Ravenna, and Prof. O. Tubertini, University of Bologna, for valuable expert advice.

## II. List of authors and titles of reports and theses resulting from the subproject's work

- Brandt, J., J. H. Christensen, L. M. Frohn, R. Berkowicz, and F. Palmgren. The DMU-ATMI THOR air pollution forecast system - system description. Technical report from NERI no 321, National Environmental Research Institute, P.O. Box 358, Frederiksborgvej 399, DK-4000 Roskilde, Denmark, 60 p, 2000.
- Builtjes, P.J.H., H.M. ten Brink, G. de Leeuw, M. van Loon, C. Robles Gonzalez and M. Schaap, Aerosol Air Quality Satellite Data, Final Report 4.1/AP-06, 2001.
- De Leeuw, G., Coastal aerosols: occurrence and effects. EUROTRAC2, CAPMAN, annual report 1999, edited by G. Geernaert, pp. 55-60, 2000.
- De Leeuw, G., G.J. Kunz, M. Moerman, L.H. Cohen, K.H. Schlünzen, L. Klein, F. Müller, K. von Salzen, C.-J. Lenz, M. Schulz, S. Tamm, E. Plate, G. Geernaert, O. Hertel, E. Vignati, L. Frohn, B. Pedersen, B. Jensen, L.L. Sørensen, S. Lund, T. Jickells and L. Spokes (2000). Atmospheric Nitrogen inputs into the coastal ecosystem (ANICE) ENV4-CT97-0594. Second Annual Report (Feb 1, 1999- Jan 31, 2000). TNO Physics and Electronics Laboratory, Report FEL-00-C125, 2000b.
- Ellermann, T., O. Hertel, and Ambelas Skjøth, C. NOVA 2003, Atmospheric Deposition of Nitrogen 1999 (In Danish: NOVA 2003, Atmosfærisk deposition af kvælstof 1999). NERI, Technical Report, no. 332, 120 p, 2000.

## III. List of publications in the refereed literature from the subproject

- Baker, A.R., S.M. Turner, W.J. Broadgate, A. Thompson, G. McFiggans, O. Vesperini, P.D. Nightingale, P.S. Liss, and T.D. Jickells. Distribution and sea-air fluxes of biogenic trace gases in the eastern Atlantic Ocean, *Global Biogeochemical Cycles*, **14**, 871-886, 2000a.
- Baker, A.R., D. Thompson, M.L.A.M. Campos, S.J. Parry and T.D. Jickells. Iodine concentration and availability in atmospheric aerosol, *Atmospheric Environment*, **34**, 4331-4336, 2000b
- Cornell, S., K. Mace, S. Coeppicus, R. Duce, B. Huebert and T.D. Jickells. (in press). Organic nitrogen in Hawaiian rain and aerosol. *Journal of Geophysical Research*, 2000.
- De Leeuw, G., F.P. Neele, M. Hill, M.H. Smith and E. Vignati. Sea spray aerosol production by waves breaking in the surf zone. *J. Geophys. Res.*, 105 (D2), 29397-29409, 2000a.
- De Leeuw, G., L.H. Cohen, L.M. Frohn, G. Geernaert, O. Hertel, B. Jensen, T. Jickells, L. Klein, G. J. Kunz, S. Lund, M.M. Moerman, F. Müller, B. Pedersen, K. von Salzen, K. H. Schlünzen, M. Schulz, C. A. Skjøth, L.L. Sorensen, L. Spokes, S. Tamm and E. Vignati (2000). Atmospheric input of nitrogen into the North Sea: ANICE project overview. *Accepted for publication in Nearshore and Coastal Oceanography (Continental Shelf Research)*, ELOISE special issue, 2001a.
- De Leeuw, G., and L.H. Cohen. Bubble size distributions on the North Atlantic and the North Sea. *Gas Transfer and Water Surfaces*, edited by M.A. Donelan, W.M. Drennan, E.S. Salzman, and R. Wanninkhof, AGU, in press, 2001b.
- De Leeuw, G., G.J. Kunz and C. O'Dowd (2001). Micro-meteorological observations at the Mace Head mid-latitude coastal station. *Submitted for publication*, 2001c.
- Eyckmans, K., J. Zhang, J. de Hoog, P. Joos and R. Van Grieken. Leaching of nutrients and trace metals from aerosol samples; a comparison between a re-circulation and an ultrasound system. *International Journal of Environmental Analytical Chemistry*, submitted and accepted, 2000.
- Frohn, L.M., J.H. Christensen, J. Brandt and O. Hertel. Development of a high resolution air pollution model - The numerical approach. *Submitted for Journal of Computational Physics*, 2001.
- Geernaert, G. L., Flux profile relations under quasi-homogeneous conditions over the offshore coastal zone. *Boundary Layer Meteorology*, under revision, 2000.
- Hertel, O., Ambelas Skjøth, C., T. Ellermann, H. Skov and L. M. Frohn. Atmospheric Nitrogen Deposition to Danish Waters 1999. 10 pp. *Submitted for publication in Pure and Applied Chemistry*, 2001.
- Jensen, D.R., S.G. Gathman, C.R. Zeisse, C.P. McGrath, G. de Leeuw, M.H. Smith, P.A. Frederickson and K.L. Davidson (2001). Electrooptical propagation assessment in coastal environments (EOPACE): Overview and initial accomplishments. *Accepted for publication in Opt. Eng.*, 2001.

- Kleefeld, C., C.D. O'Dowd, S. O'Reilly, S. G. Jennings, P. Aalto, E. Becker, G. Kunz and G. de Leeuw. The relative contribution of sub and super micron particles to aerosol light scattering in the marine boundary layer (MBL). *Submitted for publication*, 2001.
- Kunz, G.J., and G. de Leeuw. LIDAR studies of spatial and temporal distributions of aerosols at Mace Head: influence of local sources. In: C. O'Dowd and K. Hämerli (Eds.) *New Particle Formation and Fate in the Coastal Environment*. Report Series in Aerosol Science, Finnish Association for Aerosol Research, pp. 48-54, 2000a.
- Kunz, G.J., and G. de Leeuw. Micrometeorological characterisation of the Mace Head field station during PARFORCE. In: C. O'Dowd and K. Hämerli (Eds.) *New Particle Formation and Fate in the Coastal Environment*. Report Series in Aerosol Science, Finnish Association for Aerosol Research, pp. 55-62, 2000b.
- Kunz, G.J., G. de Leeuw, C. O'Dowd and E. Becker, LIDAR studies of the atmospheric boundary layer and locally generated sea spray aerosol plumes at Mace head. *Submitted for publication*, 2001.
- Kusmierczyk-Michulec, J., M. Schulz, S. Ruellan, O. Krüger, E. Plate, R. Marks, G. de Leeuw and H. Cachier, Aerosol composition and related optical properties in the marine boundary layer over the Baltic Sea. *J. Aerosol Science*, *in press*, 2001a.
- Kusmierczyk-Michulec, J., G. de Leeuw and C. Robles Gonzalez, Empirical relationships between aerosol mass concentrations and Ångström parameters, *Submitted for publication*, 2001b.
- Leifer, I., G. de Leeuw and L.H. Cohen. Secondary bubble production from breaking waves: the bubble burst mechanism. *Geophys. Res. Letters*, Vol. 27 (24), p.p. 4077-4080, 2000a.
- Leifer, I., G. de Leeuw and L.H. Cohen (2000). Optical measurement of bubbles: system design. *Submitted for publication*, 2001b.
- Leifer, I., G. de Leeuw and L.H. Cohen. Calibrating optical bubble size by the displaced mass method. *Submitted for publication*, 2001c.
- Müller F., K.H. Schlünzen and M. Schatzmann. Test of numerical solvers for chemical reaction mechanisms in 3D air quality models. *Environmental Modelling Software*, **15**, 639-646, 2000.
- O'Dowd, C., K. Hämerli, J. Mäkelä, M. Väkeva, P. Aalto, G. de Leeuw, G. Kunz, E. Becker, H.-C. Hansson, E. Becker, A.G. Allen, R.M. Harrison, C. Kleefeld, M. Geever, S.G. Jennings and M. Kulmala, Coastal new particle formation: Environmental conditions and aerosol physico-chemical characteristics during nucleation bursts, *Submitted for publication*, 2001a.
- O'Dowd, C., K. Hämerli, J. Mäkelä, L. Pirjola, M. Kulmala, S.G. Jennings, H. Berresheim, H.-C. Hansson, G. de Leeuw, G.J. Kunz, A.G. Allen, C.N. Hewitt, A. Stroh, Y. Viisanen and T. Hoffmann, A dedicated study of new particle formation and fate in the coastal environment (PARFORCE): Overview of objectives and initial achievements, *Submitted for publication*, 2001b.
- Osán, J., J. de Hoog, A. Worobiec, C.-U. Ro, K.-Y. Oh, I. Szalóki and R. Van Grieken. Application of chemometric methods for classification of atmospheric particles based on thin-window EPMA data. *Analytica Chimica Acta*, submitted and accepted
- Osán, J. I. Szaloki, C.-U. Ro and R. Van Grieken. Light element analysis of individual microparticles using thin-window EPMA. *Mikrochimica Acta*, 132, 349-355, 2000
- Piazzola, J., A.M.J. van Eijk and G. de Leeuw. Extension of the NAVY aerosol model to coastal areas. *Opt. Eng.* 39, (6), 1620-1631, 2000.
- Robles-Gonzalez, C., J.P. Veefkind and G. de Leeuw. Mean aerosol optical depth over Europe in August 1997 derived from ATSR-2 data. *Geophys. Res. Lett.* 27, 955-959, 2000.
- Robles-Gonzalez, C., G. de Leeuw, P.J.H. Builtjes, M. van Loon and M. Schaap. Spatial variation of aerosol properties derived from satellite observations, *Submitted for publication*, 2001.
- Rutgersson, A., A. Smedman and U. Högström: The use of conventional stability parameters during swell. *J. Geophys. Res.*, Accepted, 2001.
- Spokes, L.J., S.G. Yeatman, S.E. Cornell and T.D. Jickells. Nitrogen deposition to the eastern Atlantic Ocean. The importance of south-easterly flow. *Tellus* **52B**, 37-49, 2000.
- Schlünzen, K.H., and L. Klein. (2000) Simulation of coastal atmospheric processes including aerosols. CAPMAN annual report 1999, International Scientific Secretariat, GSF-Forschungszentrum für Umwelt und Gesundheit GmbH, Munich, Germany, 63-67.



- Szaloki, I., J. Osan, C.-U. Ro and R. Van Grieken. Quantitative characterisation of individual aerosol particles by thin-window electron probe microanalysis combined with interactive simulation. *Spectrochimica Acta*, part B, **55**, 1017-1030, 2000
- Sørensen, L.L., G.L. Geernaert, G. de Leeuw, E. Plate and M. Schulze. Flux divergence for nitric acid in the marine atmospheric surface layer. *Under revision*, 2000.
- Van Grieken, R., K. Gysels, S. Hoornaert, P. Joos, J. Osan, I. Szaloki and A. Worobiec. Characterisation of individual aerosol particles for atmospheric and cultural heritage studies. *Water, Air and Soil Pollution*, **123**, 215-228, 2000
- Vignati, E., G. de Leeuw and R. Berkowicz. Modeling coastal aerosol transport and effects of surf-produced aerosols on processes in the marine atmospheric boundary layer, *Accepted for publication in JGR-Atmospheres*, 2001.
- Yeatman, S.G., L.J. Spokes and T.D. Jickells. Comparison of coarse-mode aerosol nitrate and ammonium at two polluted coastal sites. *Atmospheric Environment*, **35**, 1321-1335, 2001a.
- Yeatman, S.G., L.J. Spokes, P.F. Dennis and T.D. Jickells. Comparison of nitrogen isotopic composition at two polluted coastal sites. *Atmospheric Environment*, **35**, 1307-1320, 2001b.
- Yeatman, S.G., L.J. Spokes, P.F. Dennis and T.D. Jickells. Can the study of nitrogen isotopic composition in size-segregated aerosol nitrate and ammonium be used to investigate atmospheric processing mechanisms? *Atmospheric Environment*, **35**, 1337-1345, 2001c.

#### **IV. Reports from the principal investigators**

<u>Gerrit de Leeuw</u> <i>Aerosols in the Coastal Marine Boundary Layer: generation, transport and effects</i>	17
<u>Søren Larsen</u> , Faye Dunkerley, Torben Mikkelsen and Anne Maria Sempreviva <i>Modification of the Near Surface Temperature Field Inland from a Coastal Discontinuity</i>	23
<u>Sandro Finardi</u> , Gianni Tinarelli, Domenico Anfossi, Giuseppe Brusasca, Gabriele Carboni, Davide Sanavio, Silvia Trini Castelli and Anna Toppetti <i>A Comparison of Different Modelling Techniques to Evaluate Atmospheric Dispersion of Pollutants in Complex Coastal Sites</i>	26
<u>Gunilla Svensson</u> , Michael Tjernström and Mark Zagar <i>High-resolution Modelling of Atmospheric Dispersion and Turbulence Transport in the Coastal Marine Boundary Layer</i>	30
<u>Gary Geernaert</u> <i>Parameterization of Dry Deposition</i>	36
<u>Ole Hertel</u> , Carsten Ambelas Skjøth and Lise Marie Frohn <i>Mapping Nitrogen Loads of Coastal Marine Waters</i>	39
<u>Tim Jickells</u> , Alex Baker, Sarah Cornell, Lucinda Spokes and Stuart Yeatman <i>Air-Sea Exchange of Nitrogen and Iodine</i>	43
<u>Ann-Sofi Smedman</u> , Anna Rutgersson and Ulf Högström <i>The Effect of Swell on Air-Sea Exchange in the Baltic Sea</i>	47
<u>K. Heinke Schlünzen</u> and Laura Klein <i>Simulation of Coastal Atmospheric Processes including Aerosols</i>	52
<u>René Van Grieken</u> and Kurt Eyckmans <i>Air-Sea Exchange of Nutrients, Trace Metals and Organic Micro Pollutants at the North Sea</i>	57