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(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)

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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 compunds 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 implemention 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 Skaggerak 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_2, NO_3, SO_4, F, Na, NH_4, K, Mg_2, and Ca_2. 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 re-visited 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 Högström 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 coastsl 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 signficant 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 intput 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, Despiau).

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

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IV. Reports from the principal investigators

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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Modification of the Near-Surface Temperature Field Inland from a Coastal Discontinuity

A contribution to subproject CAPMAN

S.E. Larsen, F. Dunkerley, T. Mikkelsen and A.M. Sempreviva

Riso National Laboratory, Dept. of Meteorology and Wind Energy
P.O. Box 49, DK-4000, Roskilde, Dänemark

Summary

In 2000 analysis of the JYLEX data set has been used to verify a model linking the growth of the coastal internal boundary layer (CIBL) to near-surface temperature field. This model will in turn be used as input to a simple linear model for the modification to the mean flow across the coastline.

Aim of Research

In order to predict the modification of the wind field at a coastal discontinuity a description of the CIBL is required. The CIBL height (λ) is not an easily measured parameter and, although a number of models for this have been compared with experimental data (e.g. Gryning and Batchvarova 1996, Kallstrand and Smedman 1997, Melas and Kambezidis 1992), there is no particular consensus on the most appropriate model to use. Experiments relate to different stability, roughness and terrain conditions and models require varying complexity of input.

The linear approach to wind flow modelling means that the effect on the mean wind field of changes in temperature stratification, roughness and terrain at the coastline can be considered separately. Results are generated rapidly and the aim is to run the model with minimal inputs. Both $\lambda(x)$ and $\theta(x,z)$, the potential temperature profile, are required. Hence if these parameters can be related, so that the description of CIBL can be obtained from land based temperature and wind speed measurements only, then this may provide a simple and robust method of providing information to the wind flow model.

Activity During 2000

We wish to establish a link between an assumed potential temperature profile over land downstream of the coastline (in 2 dimensions)

$$T_{land}(x,z) = T_{sea}e^{-z/\lambda_1} + \theta(x)e^{-z/\lambda(x)} \tag{1}$$

and an assumed profile for the CIBL denoted by

$$\lambda(x) = ax^b \tag{2}$$

where T_{sea} and $T_{land}(x,0)$ are the near surface temperatures over sea and land respectively and a and b are constants. From the literature, b is typically in the range *0.5* to *1.0*, whereas a varies more widely. Using the simplified form of the heat equation

$$U\frac{\partial\theta'}{\partial x} = -\frac{1}{\rho c_p}\frac{\partial H}{\partial z} \tag{3}$$

where H(z) is the heat flux and U(z) is the upstream mean wind, and assuming constant entrainment $H_\lambda = -AH_0$ at $z=\lambda$, we obtain

$$\lambda \frac{\partial \theta}{\partial x} + \theta \frac{\partial \lambda}{\partial x} = \frac{g H_0 (1+A)}{\rho c_p \theta_0 U} \tag{4}$$

Substituting for λ from (2), equation (4) becomes a 1st order ordinary differential equation in $T_{land}(x)$ which can be solved to give

$$\theta(x) = \frac{H_0 (1+A)}{a \rho c_p U} x^{1-b} \tag{5}$$

To verify that this model is a reasonable approximation we should be able, by analysis of suitable measured values of $T_{land}(x,0) - T_{sea}$, to find constant values of *a* and *b*, which are consistent with the literature.

The JYLEX data set (Sempreviva et al., 1992) has been used for the analysis. Temperature and velocity data were collected during a two year period from measurements at four meteorological masts placed inland from the west coast of Jutland in Denmark. Previous analysis has concentrated on data averaged over season or wind sector. In this new work the individual 10 minute data records are used. The results are presented for stable or neutral conditions over the sea and unstable conditions over land for winds from 225° to 304°. Data from masts 1 to 3 only are used and because of the form of (5) the data set is restricted to $T_3 - T_{sea} > T_2 - T_{sea} > T_1 - T_{sea} > 0$. Heat flux data was obtained from sonic anemometer measurements. The near surface temperatures are approximated by measurements at 2m.

Table 1 contains exponents *b* and coefficients $d'(=(1+A)/a\rho c_p)$ for nine 10° wind sectors obtained by fitting a power law curve to $(T_{land}(x,0) - T_{sea})U_{sea} / H_0$ as a function of downstream distance from the coast. *Figure 1* shows the same function plotted for the 265°-274° sector only and the result for all sectors with summer data only is presented in Figure 2.

Table 1 Power law exponents for curve fitting

sectors	$(T_{land}(x,0) - T_{sea})U_{sea} / H_0$		
	b	d´	No of points
225-234	0.72	0.017	135
235-244	0.74	0.014	130
245-254	0.74	0.014	152
255-264	0.71	0.008	74
265-274	0.71	0.009	86
275-284	0.67	0.010	107
285-294	0.65	0.008	207
295-304	0.64	0.007	178

We can note from the results that, despite the large spread in the experimental data, the power law exponents (*b*) are consistent across changes in wind sector and season and in reasonable agreement with the literature. Differences between the sectors can be explained by the changes in fetch length from the coast to the mast 1 which affects the calculation of T_{sea} and U_{sea}. The coefficients (*d'*) for the different sectors also show good agreement. We might expect more variability in this parameter because of the relatively wide range of fluctuations in the heatflux time series calculated from the sonic data. In fact $d' = 0.01$ corresponds to $a = 0.1$ which again is reasonably consistent with the literature. (e.g. Bergstrom et al., 1988).

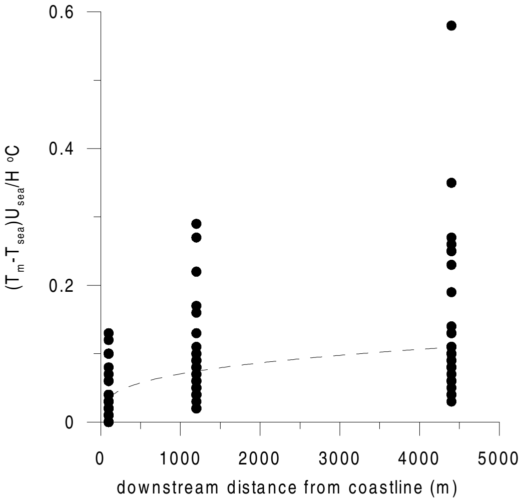

Figure 1: $(T_{land}(x,0) - T_{sea})U_{sea} / H_0$ as a function of downstream distance (x) for winds from 265° to 274°. (dashed line represents best power law fit to data.)

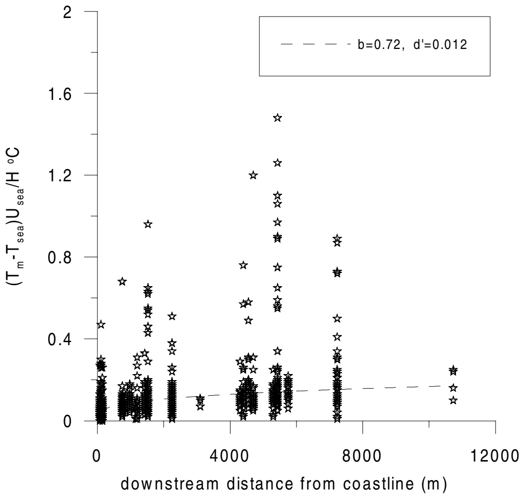

Figure 2: $(T_{land}(x,0) - T_{sea})U_{sea} / H_0$ as a function of downstream distance (x) for winds from 225° to 304° - summer data only. (dashed line represents best power law fit to data.) 645 points used.

The next aim is to expand the analysis of the JYLEX data set to include other combinations of stability conditions. The application of the same process to other appropriate data sets is also being considered.

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A Comparison of Different Modelling Techniques to Evaluate Atmospheric Dispersion of Pollutants in Complex Coastal Sites

A contribution to subproject CAPMAN

Sandro Finardi, Gianni Tinarelli, Domenico Anfossi, Giuseppe Brusasca, Gabriele Carboni, Davide Sanavio, Silvia Trini Castelli and Anna Toppetti

ARIANET, via Elvezia 42, 20052 Monza (MI), Italia

Summary

The performances of the Lagrangian particle model SPRAY (Tinarelli et al., 1994) and the US EPA regulatory model ISC3ST (US) have been intercompared in two applications dealing with industrial emissions at coastal sites. The two considered models have been applied to reconstruct climatological ground level concentrations around the thermal power plants of Fusina, nearby Venice, and Vado Ligure. The former site is characterised by flat terrain and shows the climatological features of the north-western Adriatic Sea coasts, with local scale circulations like sea/land breezes having a limited statistical frequency. The latter site is located on the Ligurian coast and it is characterised by very complex terrain and a climatology dominated by the superposition of land/sea breezes and slope flows. In Venice, the two models give similar results for both long term average concentrations and daily average percentiles. Relevant differences have been observed only for very high (over 98th) hourly average percentiles and maximum values. On the contrary, at the more complex Vado Ligure site the two modelling systems produced different results that are hardly comparable. The reliability of the downscaling of synoptic weather data to the local scale to drive air pollutant dispersion models in complex coastal conditions, has been evaluated through the application of a mesoscale prognostic model. Grid nesting technique has been used to enhance space resolution from the meteorological analysis grid size (0.5 deg.) to the target resolution of 1 km. The prognostic non-hydrostatic model RAMS (Pielke et al., 1992) has been used as the meteorological driver for the Lagrangian particle model SPRAY. The results of earlier modelling applications on the site of Vado Ligure offered the possibility to compare diagnostic and prognostic meteorological codes. This comparison has been extended to the pollutant concentration produced by the same dispersion model driven by the two different meteorological approach. The overall results obtained by the two modelling systems seem to be comparable.

Aim of the research

In this phase of the project we wish to compare the results obtained through the application of a modelling system made by a mass-consistent diagnostic meteorological model coupled with a Lagrangian particle model and a steady state Gaussian model. Even if it is well known that steady state models are not suited to reproduce dispersion phenomena in complex terrain, they are still frequently employed in the frame of air quality impact assessment studies. It is therefore relevant to compare the concentration fields obtained by the different modelling techniques in such a complex condition. A different item of our project regarded the possibility to reconstruct local flow fields directly from large scale meteorological information. This potentiality is very interesting for practical applications because the diagnostic reconstruction of local scale flow needs to be based on local meteorological measurements (ground stations and vertical profiles) that are normally hardly available, and the execution of very expensive and time consuming field campaigns. The comparison of diagnostic and prognostic meteorological model results can set in evidence the advantages and limitations of the two modelling approaches.

Activities during the year

The performances of the Gaussian model ISC3ST and of the modelling system composed by the mass-consistent meteorological model MINERVE (Aria Tech., 1995) and by the Lagrangian particle model SPRAY have been compared in two practical applications concerning industrial emissions at coastal sites. The first test case regarded the site of Fusina. The place is characterised by flat terrain and local wind measurements that generally show a limited horizontal variation and vertical shear of the winds. The two models have been applied to compute ground level concentrations due to the emissions of two thermal power plants located a few kilometers apart. The simulations covered a period of one year. Hourly average concentrations have been post-processed to compute long term statistics. The concentration fields corresponding to the Italian and European standards and guidelines have been intercompared. The second test case concerned the reconstruction of ground level concentrations due to the emissions of a thermal power plant sited in Vado Ligure. On site field campaigns provided a relevant amount of data for model evaluation. During a previous study, short term episodes have been reconstructed through the application of the cited MINERVE+SPRAY modelling system. Hourly ground level concentration fields have been then used to build seasonal and yearly averages, applying a method based on the statistic of the weather type associated to each episode. These results have been used as a basis to compare concentrations obtained by the Gaussian model ISC3ST, that has been applied to simulate the whole year that includes the previously analysed episodes. In order to verify the possibility to reconstruct local flow directly from large scale meteorological information, a selected summer episode (23-25/07/1997) has been simulated applying the non-hydrostatic prognostic model RAMS. A two way nesting technique has been employed to describe both the synoptic driving flow and local scale phenomena. Three nested grids have been defined, with horizontal resolution of 16, 4 and 1 km respectively, for a total space extension of about 1100, 200 and 53 km. RAMS has been initialised using the ECMWF 0.5 degrees resolution analysis fields, synoptic and local observations. ECMWF analyses have been used to drive boundary conditions with a 6 hour time resolution. The Lagrangian particle model SPRAY has been driven by the mean wind fields generated by both MINERVE and RAMS meteorological processors. Diagnostically simulated turbulence fields have been defined using a built-in parameterisation scheme based on the Monin-Obukhov similarity theory and the evaluation of the surface energy budget. In the prognostic case, turbulence fields have been generated by the interface code MIRS (Trini Castelli and Anfossi, 1997), that uses the information given by the closure parameterisation scheme employed by RAMS. In particular the velocity variances $\sigma_i^2\,(i = 1,2,3)$ were computed either from Hanna (HS) scheme (1982) or from RAMS according to the Mellor and Yamada (MY) closure scheme (1982).

Principal results

In the coastal site of Fusina, characterised by a rather simple wind climatology, the results of MINERVE+SPRAY and ISC3 models are comparable, at least for the more relevant features of the concentration fields. Important differences are observed only for hourly percentiles higher that 98^{th}. A quite different result is obtained for the site of Vado Ligure, where terrain and flow complexity originated large differences in the concentration fields produced by the models. For seasonal and yearly average (*Figure 1*) concentrations the two models indicated areas of impact located over different portions of the computational domain. Moreover the average concentration fields produced by ISC3 do not reproduce the wind rose characteristics. ISC3 showed a tendency to overestimate concentrations, producing maximum values that are not observed by any station of the air quality control network. Concerning the evaluation of the possibility to downscale synoptic weather information to the local scale, RAMS correctly reproduces the breeze cycle features close to the ground, with a general tendency to overestimate wind speeds near the surface. There are instead some problems to reproduce all the spatial and temporal details shown by the measurements. The main discrepancies are located in the layer between 750 m and 1500 m. Figure 2 shows horizontal wind cross-sections produced by MINERVE and RAMS in the layer closer to the surface during nocturnal land breeze conditions. The wind pattern is similar in the region around the power plant location, while in the northern, western and south-western part of the domain the two wind fields substantially differ. With regard to dispersion simulations, the two modelling systems show results of comparable quality. The RAMS+SPRAY modelling system seems to be more reliable than the diagnostic modelling system in regions not covered by meteorological measurements, far from the emission. As an example, *Figure 3* depicts the SO_2 g.l.c. trends recorded at the Bocca d'Orso chemical station, located 8.7 km to the north-west of the emission site, at a height of 530 m asl.

Main conclusions

The evaluation of the impact on air quality of industrial emissions has to verify the attainment of air quality standards that usually state limits on yearly and seasonal statistics of pollutant. In coastal areas, where non-stationary meteorological conditions can be frequent, different modelling tools can give very different results, influencing the compliance with air quality standards. Steady state and 3D modelling techniques give similar results only in places characterised by "simple" wind climatology, e.g. sites characterised by weak space variations of wind speed and direction. In complex coastal sites 3D models are needed to correctly reproduce at least the major features of pollutant dispersion. The comparison of diagnostic and prognostic meteorological models used as a driver to Lagrangian particle dispersion models showed results of comparable quality. These results confirmed the possibility to use meteorological fields obtained from the prognostic downscaling of large scale weather data to reconstruct the local scale flow where extensive observations are not available. This approach can be an alternative to the use of diagnostic models based on an expensive measuring network.

Aim for the coming year

The activities concerning the coastal sites of Fusina and Vado Ligure and financed by ENEL have been completed during the last year. The possibility to start a new project concerning photochemical pollution in southern Italy coastal sites is being discussed.

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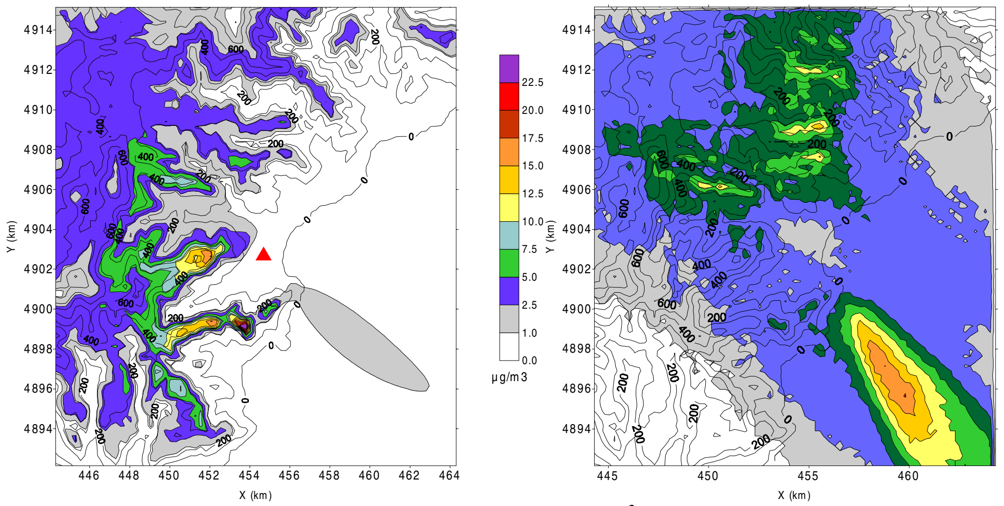

Figure 1. Ground level yearly average concentration of SO_2 ($\mu g/m^3$): (left) ISC3ST, (right) SPRAY driven by MINERVE. The red triangle indicates the emission location.

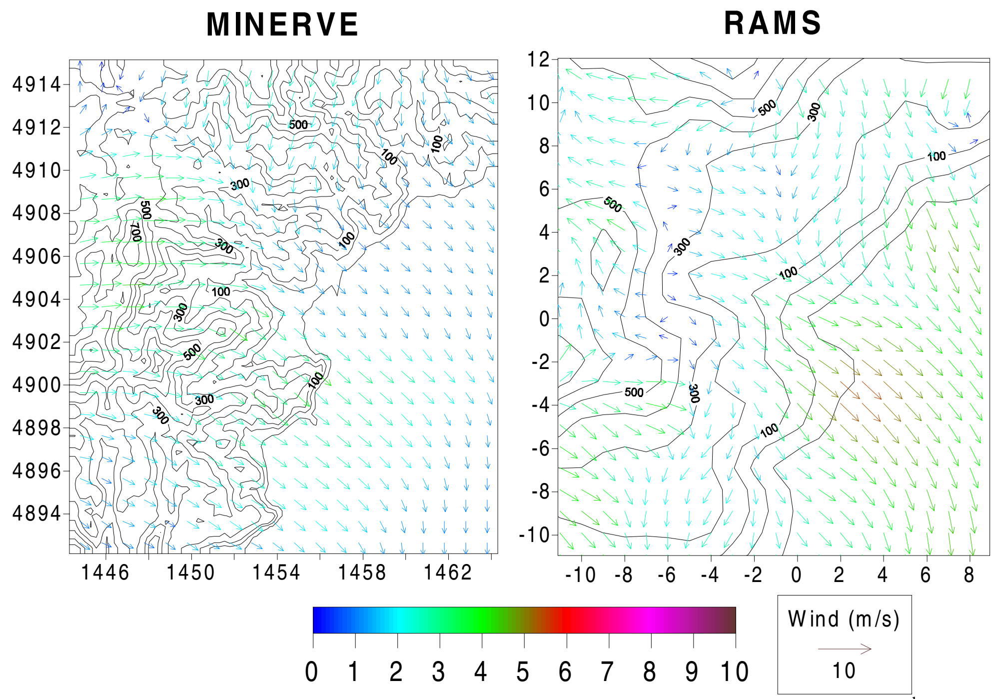

Figure 2. Near ground wind field computed by MINERVE (left) and RAMS (right). July 23rd 1997 23:00 lst.

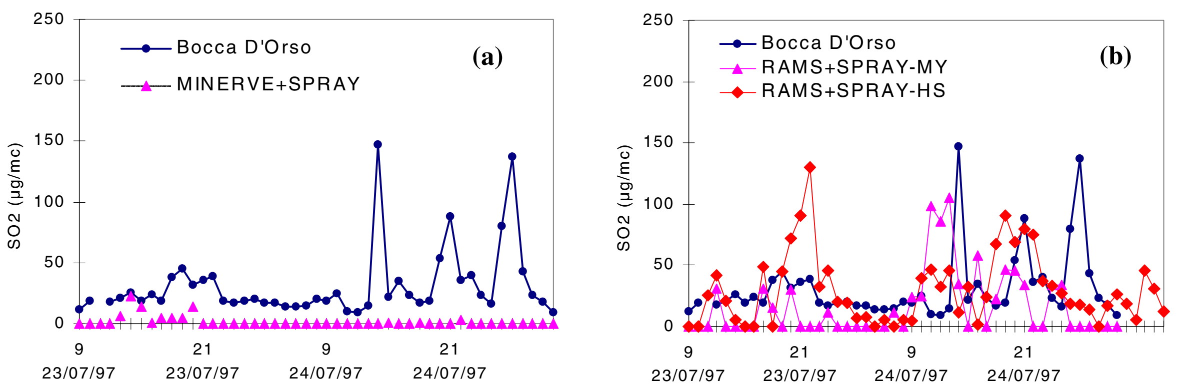

Figure 3. Comparison between computed and measured g.l.c. time series at the Bocca d'Orso station. a) MINERVE+SPRAY b) RAMS+SPRAY

High-Resolution Modeling of Atmospheric Dispersion and Turbulence Transport in the Coastal Marine Boundary Layer

A contribution to subproject CAPMAN

Gunilla Svensson, Michael Tjernström and Mark Zagar

Department of Meteorology, Stockholm University
S-106 91 Stockholm, Sweden

Summary

Due to the highly variable conditions in the marine boundary layer close to a coastline, accounting for the atmospheric deposition of nitrous compounds to the coastal ocean requires high resolution modeling of both the dispersion processes in the atmosphere and the deposition. For the deposition, no global theory exist for deposition in the case of heterogeneous meteorological conditions, although a simplified theory was suggested by Geernaert (2000, personal communication). The aim of this project is partly to study the dispersion of atmospheric gases in highly complex conditions, through high-resolution modeling, and to device a parameterization of the atmospheric deposition for use in coarser scale models.

In this paper, we report on the initial attempts in these fields. High-resolution modeling of the dispersion of a passive tracer for the Kattegat using the US Navy Coupled Ocean/Atmosphere Mesoscale Prediction System - (COAMPS) model (Hodur 1997) illustrates the complexity of the flow in the strait between Sweden and Denmark. For summer conditions, the low-level flow in the strait favors a northerly or southerly flow even for background flow across the strait - a channeling effect due to the often stably stratified boundary layer over the water. Dispersion of a tracer that was continuously released at the surface over northern Jutland illustrates the fate of pollutants for a single day. The plume meanders strongly but remains unmixed and reaches the Swedish coast at different locations for different times of the day.

Highly simplified simulations with a very high-resolution model were utilized to study the variability in the turbulent fluxes close to the coast. Near-shore variations in surface momentum flux of the order of 50% compared to that far off shore were found. It was also found that the distance off-shore at which the surface fluxes approached the free undisturbed conditions are of the order of ~100 km, thus the Kattegat is not wide enough to ever be treated as an open ocean – coastal effects will influence deposition all over this water body.

Aim of the research

This study is part of the MEAD project. The over-all aim of this project is to model the conditions that are favorable for harmful algae blooms in the Kattegat sea between the west coast of Sweden and Denmark. The aims of the current project are:

1. Study dry deposition in horizontally inhomogeneous conditions, using field experiment data and high-resolution meteorological models.
2. Improve the understanding of the meteorological processes that govern extreme events of dry deposition of nutrients into the coastal marine ecosystem.
3. Develop dry deposition parameterizations suitable for inhomogeneous coastal environments in an operational meteorological model.
4. Parameterize wet deposition over the study area.

Activities during this year

This year has been devoted to address the first objective. The main tools utilized in this research are two atmospheric models, a high-resolution mesoscale research model (the MIUU model, Tjernström 1987; Enger 1990) and an operational modeling system, the COAMPS model (Hodur 1997). The latter also has the option of being used in research mode and includes an advection and dispersion algorithm for passive tracers. The situations simulated so far are idealized cases, i.e. controlled experiments.

The research model is a mesoscale model that may be used for idealized cases or for real cases characterized by relatively stationary meteorological conditions on the larger scale. This model is easy to run in one to three dimensions and for this first study, we have limited the calculations to two dimensions. The model is set up so that half of the model domain is ocean and the other half is land; one coastal zone is thus considered. The model was run for this setup for a number of background wind speeds and directions, with different temperature differences between the sea and the land and for different initial stability of the atmosphere. If the background wind speed is weak enough, sea-breeze circulations will arise. The main objective in this study is to see how the various background conditions change the turbulent transport to the ocean surface in the coastal zone. The flow is influenced by the change in surface roughness and temperature when crossing the coastline. Consequently, the turbulent transport, which controls the deposition, is influenced.

The COAMPS model can be run either operationally or for idealized cases, given a vertical sounding and the SST-fields as input. It also includes an option for advection and dispersion of a passive tracer, which was utilized here. A number of idealized sunny summer cases (August) were run, using climatological SST and low background wind speed from three different directions. The model was run with one nest, the outer domain covering most of Denmark and parts of southern Sweden and the inner domain covering the Danish and Swedish coasts with the Kattegat strait in between. The simulations were performed with an area source of an inert tracer at the surface, covering the northern tip of Jutland. These simulations reveal the complexity in the three-dimensional flow patterns that evolve over the area. It is clear that the ideal picture of a linear transport from Denmark over the water to Sweden is inadequate even for westerly winds.

Principal results

Figure 1 shows the momentum flux in the lowest km of idealized flows from the MIUU model, with winds from the land towards the sea, i.e. easterly background wind. The left panel shows a case with strong (10 ms^{-1}) flow; a turbulent boundary layer up to 500 m is seen over land. This boundary layer collapses when the air is advected over the colder water. The surface temperature difference here is 6 K between the land and the sea. *Figure 2* shows the momentum flux close to the ground normalized with the value over the open water far offshore. The turbulent momentum transport is much larger over land than over the water, due to the differences in stability. A more interesting aspect in this figure is that there is a reduction of the momentum transport by more than 50% in the coastal zone compared to the offshore value. Also interesting to notice is that this effect can be seen as far from the coast as 120 km. Since the strait between Sweden and Denmark is about 100 km wide, this means that undisturbed conditions over the water in Kattegat cannot be expected; the flow in the region will always be modified by the coasts.

In the right panel in *Figure 1*, the background wind direction is the same but the speed is reduced to 4 ms^{-1}. In this case a sea-breeze circulation develops, which changes the flow, and the momentum flux is reduced over land. Just at the coastline, however, there is a zone with enhanced fluxes. The reduction in the momentum flux over the water is in this case almost constant in a 40-km zone from the coast (see *Figure 2*, blue line).

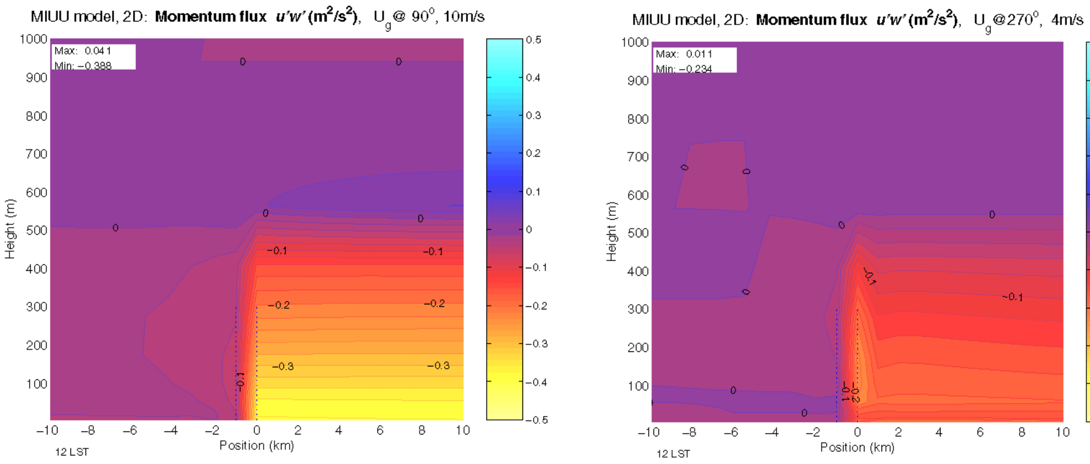

Figure 1. Vertical cross-section of the momentum flux for offshore flow at 10 ms^{-1} (left, compare the blue line in Figure 2) and 4 ms^{-1} (left, compare the red line in Figure 2).

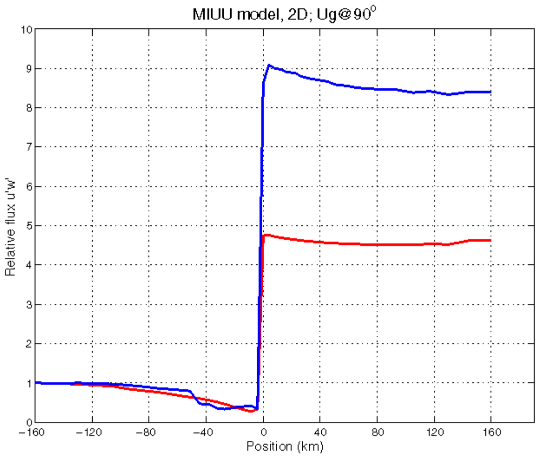

Figure 2. Normalized (by its offshore value for each case) momentum flux for two idealized simulations where the background wind was offshore at 10 (red) and 4 (blue) ms^{-1} (see Figure 1)

A conclusion from this idealized study is that although the turbulence parameterization in the mesoscale model assumes horizontal homogeneity close to the surface at each grid-point, the effects of the sudden changes at the coastline are still substantial. The momentum flux is used as a proxy for deposition here, representing the turbulent transport, as the deposition flux itself is related to the momentum flux. In addition, the amount of matter deposited to the surface is highly dependent on the concentrations in the air.

Figure 3 shows a sequence of horizontal cross sections for the inner grid of a COAMPS simulation using climatological SST, about 16°C while the inland temperatures reach about 25°C at maximum with a realistic diurnal cycle. The background wind is here from NW at a speed of 4 ms^{-1}. The arrows in these panels show the wind field close to the ground and the colors represent the concentrations of the inert tracer, emitted over land in the upper left corner with a constant emission rate. The panels show results from 6 am to 9 pm local time (note that the scale on the colorbar varies).

High concentrations (at 6 am and 9 pm) indicate that the tracer is confined to a relatively shallow layer due to stably stratified conditions, initially over both land and sea. During the day, the plume of the tracer can be followed out over the water in Kattegat. The effect of the island, Læsø, where a local convective boundary layer develops during the day, can clearly be seen in the concentration field at noon. Here the tracer is mixed through a deeper layer, thus the low-level concentrations are reduced. The transport during the day is mainly towards the SE, but the wind field shows large complexity with a sea breeze developing along the Danish coast.

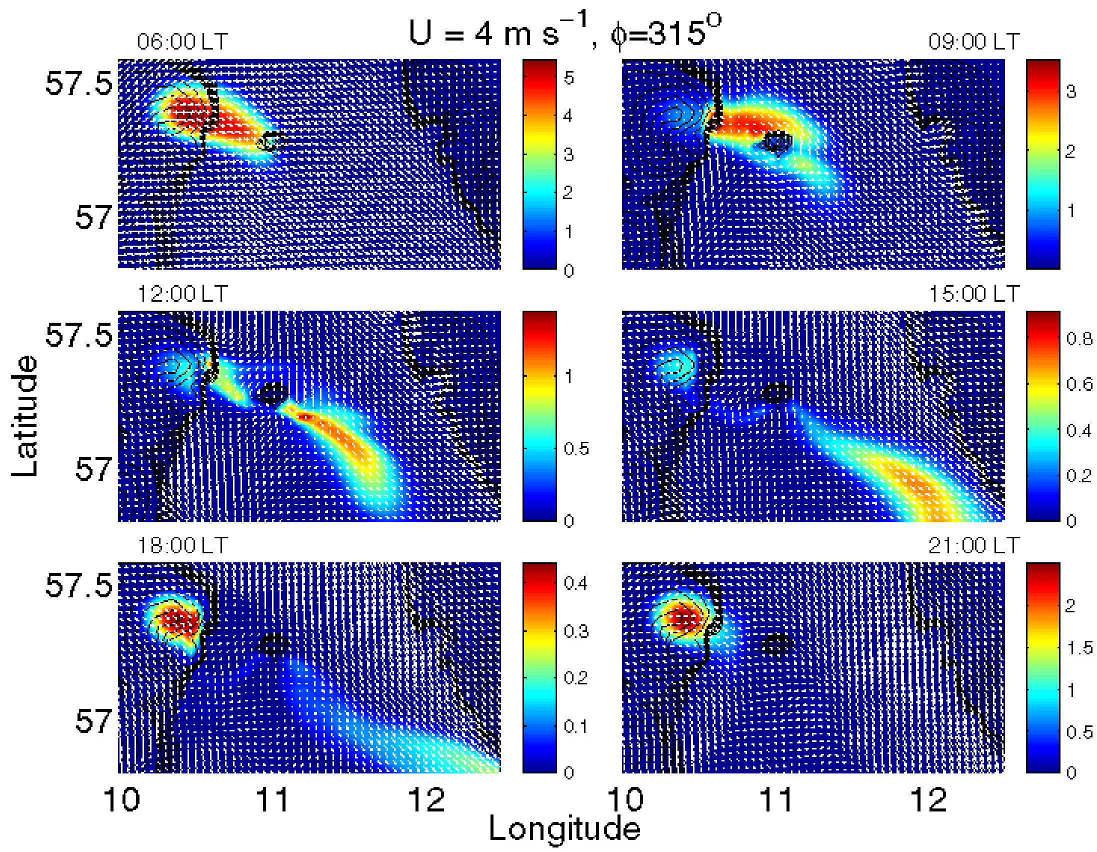

Figure 3. Horizontal cross-section across the Kattegat of wind speed (arrows) and tracer concentration (color scale) for an inert tracer continuously released at a constant rate at an area source over the northern tip of Jutland.

If the background wind is instead from the East (*Figure 4*), the plume of the tracer is instead trapped in a flow along the Danish coast by a local circulation. Some of the matter is then rapidly transported towards the south presumably in a coastal low-level jet, a local wind maximum close to the ground. No tracer can be found at the Swedish coast in this case.

Main conclusions

The simplified mesoscale simulations with just one straight coastline illustrates three things. Firstly, the turbulent fluxes at the coastline immediately on the offshore side are affected by the step-change in surface conditions. In this particular case, there was a reduction in the momentum flux by a factor of 20. Secondly, while comparing the near-coast turbulent fluxes by those far offshore representing open water conditions, there was a factor of 2 difference. The coastal fluxes gradually adjusted to the open-water conditions but the distance over which this occurs is of the order of 100 km, thus in a strait like the Kattegat the whole area will be influenced by the coastlines regardless of wind direction. Finally, if the background offshore flow is sufficiently weak a local breeze circulation will develop that further perturbs the fluxes. In this case, there will also be a mesoscale transport in that there is a substantial vertical wind in the sea-breeze front. The latter transport is not included in coarser models.

The COAMPS simulations show that the wind field over this region, with the two coastlines and the island of Læsø, is quite complex and that mesoscale phenomena such as sea breezes and low level jets are common. There appears from this limited experiment to be a tendency for the low-level flow over the Kattegat to be either northerly or southerly, trapped in the strait. The tracer emitted from one side cannot be expected to be found in a simple plume over the water. The concentration close to the surface, i.e. the material that is subject to deposition, varies quite substantial with time and space over both land and sea, due to the different stability in the atmosphere at different locations. Complex deposition patterns are thus expected to be found in the region.

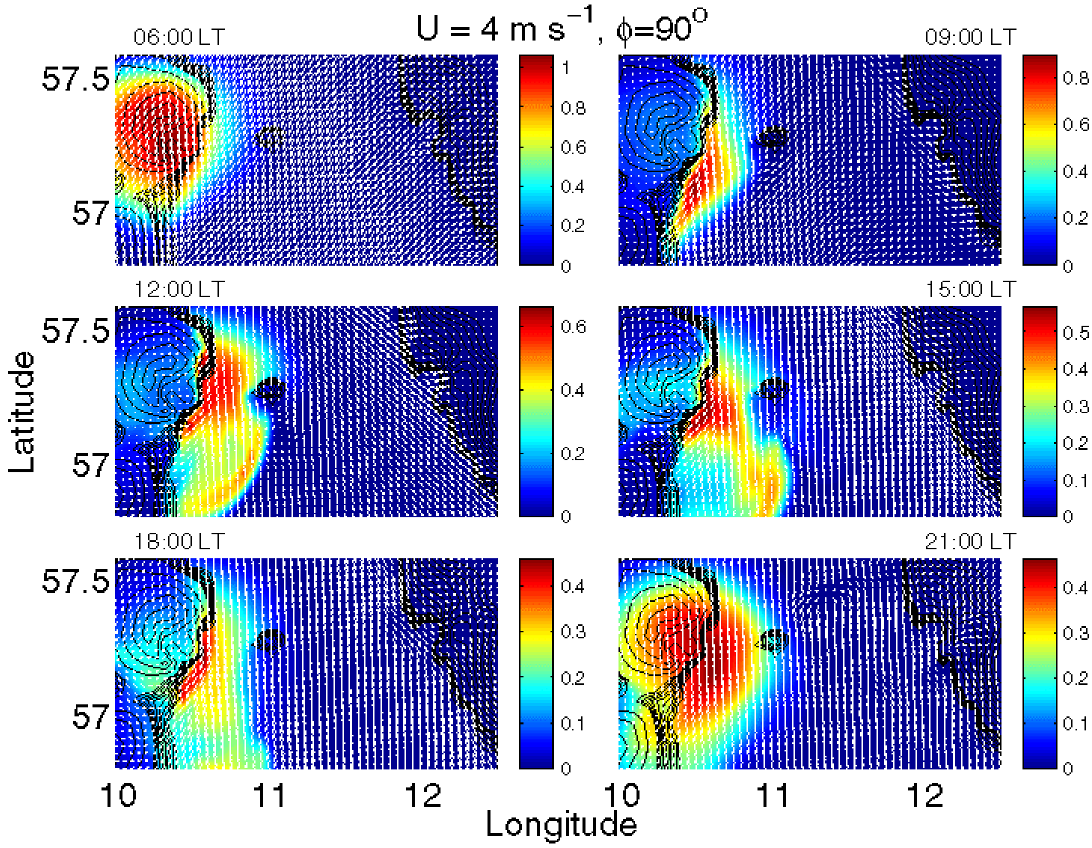

Figure 4. Same as Figure 3, but for easterly flow.

Aim for the coming year

The work with two-dimensional idealized simulations of one coastline is ongoing. A large number of cases will be run with varied input parameters (wind speed and direction, stability and land-sea temperature difference) to create a model database. This will be analyzed with respect to momentum fluxes in the coastal zone and its relation to the various input parameters. Tracer calculations will be implemented in the research model along with a deposition parameterization. At first this needs to be formulated in a standard way, using a deposition velocity related to the momentum flux.

Furthermore, we plan to investigate how much the assumption of homogeneity in the turbulence parameterization influences the results. A new boundary condition for the turbulence, without the assumption of homogeneity for the prognostic turbulent kinetic energy equation will be tested. New formulations for the boundary condition for the other prognostic variables (e.g. wind, temperature, humidity, tracer etc.) then have to be developed. The MEAD project includes two intensive field campaigns, one in 2000 and one in 2001. We will select appropriate episodes from these and run the MIUU mesoscale model so that the new parameterizations of the boundary conditions can be tested in controlled situation.

From the retrospective study of algae blooms, which is also part of the MEAD project, we will run detailed high-resolution simulations over several days for episodes with observed algae blooms, using the COAMPS model. The aim is to attempt to identify the atmospheric preconditioning for these blooms.

Acknowledgements

This project is part of the *Marine Effects of Atmospheric Deposition* (MEAD), sponsored by the European Union under contract number EVK3-CT-1999-00014. The authors are grateful to all the participants in MEAD for valuable comments and discussions. The authors are also grateful to Dr. Rich Hodur and Dr. Stephen Burk at the US Naval Research Laboratory for helping us get access to the COAMPS model.

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Parameterizations of dry deposition

A contribution to subproject CAPMAN

Gary Geernaert

National Environmental Research Institute, DK-4000 Roskilde, Denmark

Aims and Rationale

The overall aim of the work is to extend the theory for turbulent fluxes of momentum, heat, and gases, in order to extend parameterizations for conditions of quasi-homogeneous conditions.

Activities During the Year

During 1999, generalized equations for the flux profile relations were derived. For the momentum budget under quasi-homogeneous conditions, it was found that the vertical windspeed profile could be expressed as:

$$\partial U/\partial z = u^*/kz\,(Ø_m - R - S + W) \tag{1}$$

where $R = z/z_o\,\partial z_o/\partial z$; $S = ß\,(z/L)^2\,\partial L/\partial z$; and $W = kzU^2/2u^{*3}\,\partial U/\partial x$. The value of ß is typically 3 for unstable conditions; and approximately 5 for stable flow. The quantity $Ø_m$ is the profile stability function, the quantities R, S, and W are respectively corrections to the windspeed profile caused by horizontal gradients of roughness, atmospheric stability, and windspeed. In (1), $Ø_m$ may be considered to be the only "local" parameter, while all others are spatially varying. It was also found that, under most conditions, equation (1) reduces to a more practical form, i.e.:

$$\partial U/\partial z = u^*/kz\,(Ø_m - R + W) \tag{2}$$

It was also found that equation (2) could be integrated, in order to estimate the fetch dependent drag coefficient variability over the coastal ocean. The value of the drag coefficient then becomes a function of fetch and horizontally varying quantities, such as windspeed and roughness. The real drag coefficient for short fetch conditions is smaller than what is typically used in most modelling studies, i.e., caused by a flux divergence within the surface layer. For simplicity, one may say that the smaller value of the drag coefficient is associated with the deviation from the classical assumption that the surface layer is a constant flux layer.

Back in 1999, a similar equation was derived for chemical compounds, which applies to those which are reactive and those which are relatively nonreactive. A general expression was derived:

$$\partial c/\partial z = (\langle w'c'\rangle/u^*kz)(\phi_c(z/L) - W - V) - (S\Delta c/\langle w'c'\rangle) - \gamma\partial c_o/\partial x + (\Delta c/2UC_D)\partial U/\partial x \tag{3}$$

where:

$$W = \gamma\, z/z_c\, \partial z_c/\partial x \qquad V = (ku^*Uz(c-c_o))/\langle w'c'\rangle^2\ \partial c/\partial x \tag{4}$$

and S is a source or sink of the chemical during transport.

Note that if the conditions are horizontally homogeneous, and if the chemical is slowly reacting, equations (2) and (4) reduce to the more popular form, i.e.:

$$\partial g/\partial z = (\langle w'g' \rangle / u^* kz) \ \phi g(z/L) \quad (5)$$

where g represents either concentration c or windspeed U.

During 2000, the utility of these equations was explored in more detail. Comparison to data obtained from measurements carried out during the MEAD field campaigns in 2000 (in the Kattegat between Denmark and Sweden) showed that the coastal ocean exhibits far more windspeed variability than what has been generally assumed. Based on model calculations, the terms containing windspeed variability were observed to be substantially larger than terms containing roughness length variability. This led to a further simplification, such that roughness length gradients in the coastal zone could be assumed to be relatively unimportant as a control over the flux divergence. This, however, must not be confused with the value of the roughness length as a parameter as a control over the drag coefficient, during pseudo-constant flux layer conditions.

The vertical windspeed profile was explored during 2000, in terms of its variation in the offshore domain, with a view towards improved estimates of wind power production from offshore wind mills. Most models of offshore wind power potential are based on the use of a constant flux layer regardless of distance from the coastline. When one introduces a horizontal gradient of windspeed, the shape of the vertical wind profile will change, thus changing the mean windspeed as a function of height as well as the gradient across the blades. During 2000, only simple formulations of $\partial U/\partial z$ were constructed, and alterations of the windspeed profile were very sensitive to the strength of $\partial U/\partial x$ as a function of x. This implies that estimates of offshore wind power potential will rely strongly on the use of high resolution boundary layer models applied to the domain, and it will further require that the assumption of a constant flux layer (in the surface layer) is relaxed.

Plans for the next year

The terms of equations (1) and (3) will be examined in more detail, in reference to more realistic estimates of $\partial U/\partial x$ in domains controlled by complicated coastal geometries and variations in upwind roughness. Where possible, the model calculations will be compared to data gathered in the field campaigns carried out in the Kattegat Straits, under the MEAD project. Data from the MEAD project are already available from 2000, and additional data will be provided after the field campaigns in early summer 2001.

Summary

A general form of the flux profile equation was derived for both windspeed and chemicals, which considers quasi-homogensous conditions. During 2000, it was found that the integration of these equations for practical application rely on a careful and accurate treatment of the horizontal windspeed gradients. The use of these equations was also explored for the offshore energy sector, insofar that estimates of wind power potential rely on accurate estimates of the vertical windspeed gradient in the offshore region. These gradients, in turn, are sensitive to the flux divergence present within the surface layer.

Acknowledgements

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Mapping Nitrogen Loads of Coastal Marine Waters

A contribution to subproject CAPMAN

Ole Hertel[1], Carsten Ambelas Skjøth and Lise Marie Frohn

National Environmental Research Institute (NERI), Department of Atmospheric Environment (ATMI)
P. O. Box 358, Frederiksborgvej 399, 4000 Roskilde, Denmark

[1] Corresponding author: e-mail: Ole.Hertel@dmu.dk,
Tel. +45 46301148, Fax. +56 46301214

Summary

The ACDEP model has for the last eight years been used as an operational tool for calculations of atmospheric nitrogen depositions to Danish marine waters. Furthermore the model has been applied for the North Sea and the Baltic Sea. The results have shown the importance of atmospheric nitrogen depositions compared with other loads from run-off and point sources. An example is that for Danish marine waters the calculations for 1999 have shown that on a yearly basis the atmosphere contributed with a nitrogen input of similar size as the river run-off. The high atmospheric loads in 1999 were partly due to high frequency of precipitation events leading to significant wet deposition of especially aerosol bound nitrogen compounds. During the same period the Danish wastewater treatments have improved, leading to decreased contributions from river run-off.

There are considerable uncertainties in currently used operational models for calculation of atmospheric nitrogen deposition. These uncertainties concern input data as well as parameterisations of the physical and chemical processes. Large uncertainties are known to be associated with the temporal and spatial distributions of emissions. Concerning the parameterisations of the various processes in the model, main focus is currently devoted to the treatment of aerosols. Detailed field studies of chemical composition in size distributions are crucial for the development and test of new parameterisations of aerosol processes in the next generation of operational atmospheric transport-chemistry models.

Aim of the research

The aim of the project is to improve current atmospheric transport-chemistry models developed for assessment of loads and impacts of nitrogen deposition to coastal marine waters. Furthermore to apply the developed models for calculations to specific coastal waters and try to link atmospheric nitrogen loads to observed effects on the marine ecosystems. The main part of the work has been devoted to improving the performance of the Danish variable scale Lagrangian model ACDEP (Atmospheric Chemistry and Deposition) model (Hertel et al., 1995). The ACDEP-model is applied for calculations of nitrogen depositions to Danish marine waters, and in a number of research projects for similar calculations for the North Sea and the Baltic Sea. A part of the work is devoted to improvements of the description of aerosol processes in the ACDEP-model. The long-term goal of the project is the development of REGINA (REGIonal high resolutioN Air pollution model) a Eulerian nested grid model (Frohn et al., 2001), which in turn will substitute the Lagrangian model. REGINA will to some extent be built on best parts from various transport-chemistry models at NERI-ATMI.

Procedures for sensitivity studies, validation and evaluation of the model will be outlined as a part of the project, together with some recommendations for experimental studies needed for improving the performance of current models.

Activities during the year

A new version of the ACDEP-model was developed in 2000 and applied within the Danish Background Programme. This new version uses meteorological input from the Eta model operated as a part of the NERI pollution forecasting system THOR (Brandt et al., 2000). Another modification concerned the treatment of chemistry and vertical diffusion/deposition, which in the new version is solved using the same numerical scheme. A first evaluation of the new version of the model indicated a considerable improvement of the model performance.

Calculation grids with a resolution of 30 km x 30 km have been established for the North Sea area, the Baltic Sea and the Adriatic. A web site with calculation grids, and results presented in figures and tables has been established at: www.dmu.dk/AtmosphericEnvironment/ACDEP. This site will be continuously updated with new results when available.

Principal results

Calculations are performed on a routine basis within the Danish Background Monitoring programme (Ellermann et al., 2000; Hertel et al., 2001). The model was updated in connection with the change to a new set of meteorological input data from the Eta-model running under the THOR system and the new numerical handling of chemistry, vertical diffusion and deposition in the same scheme.

Calculations of nitrogen depositions to the Baltic have been performed for 1999 with the new version of ACDEP. Another set of calculations has been performed for 1999 for the North Sea. These calculations are conducted as a part of the EU programme ANICE. The calculated annual atmospheric nitrogen depositions to these two waters are shown in *Figure 1*.

Main conclusions

Atmospheric nitrogen deposition is becoming increasingly important for the nitrogen budgets of the coastal marine waters, but the impact on algae blooming is still only poorly determined. The algae blooming may be linked to events with high atmospheric loads mainly in connection with rain episodes. In order to resolve such events with transport-chemistry models considerable improvements are needed. There will be a need for high quality input data concerning meteorology and emission inventories at higher spatial and especially temporal resolution compared to the data sets currently available.

Furthermore, the current treatment of aerosol processes needs to be improved. For this purpose there is a great need for experimental data concerning chemical composition of aerosols in different size fractions. Such data are currently only available for short time periods and with highly limited time resolution – typically 24 hours or even longer sampling times.

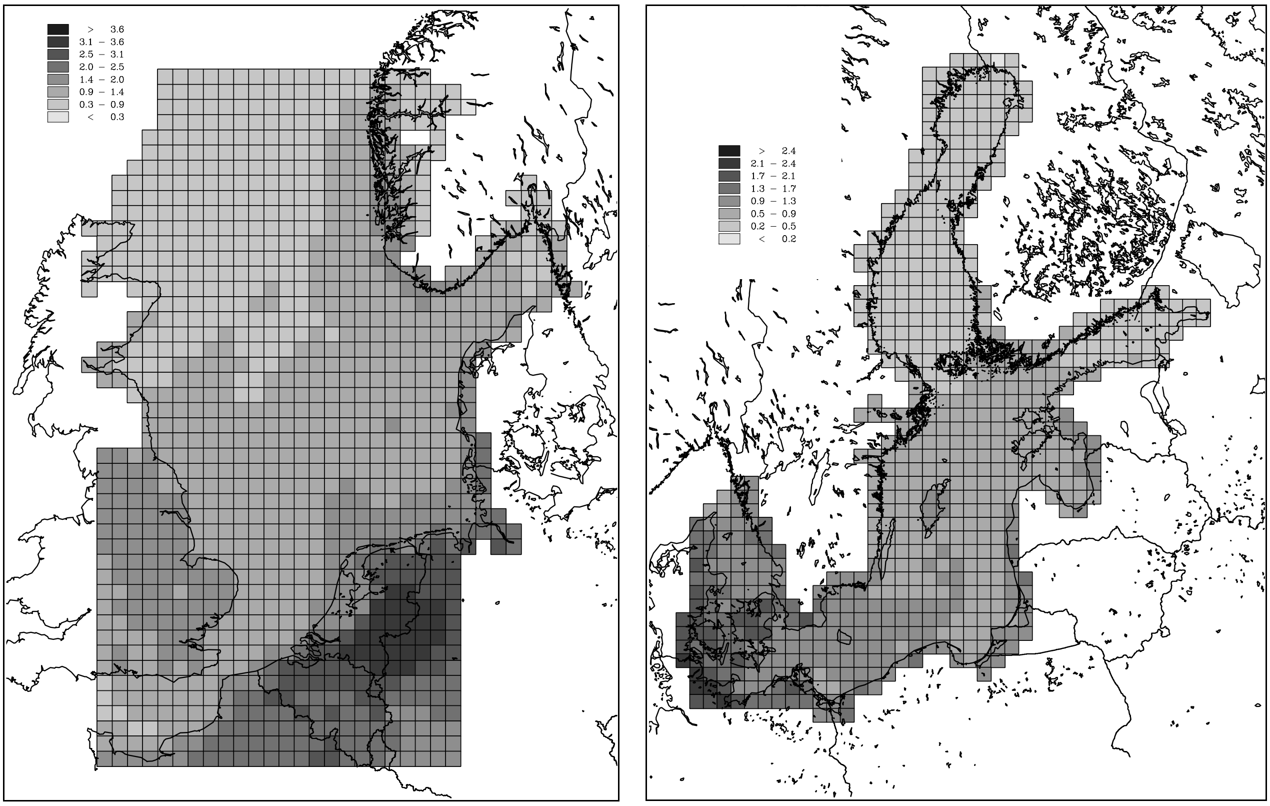

Figure 1. Calculated annual nitrogen deposition (ktonnes N/km^2) to the North Sea (left figure) and to the Baltic Sea in 1999 (right figure).

Aim for the coming year

One of the main aims for the year 2001 will be the development of an operational version of ACDEP for (semi) automatic calculations of nitrogen depositions to the Danish marine waters and the Baltic Sea. In the same way as for the previous years, calculations of nitrogen depositions to Danish marine waters will be carried out within the Danish Background Monitoring Programme. Similarly, calculations for the Baltic Sea will be carried out as part of a project for the Nordic Council of Ministers.

The developed aerosol module will be fully implemented in ACDEP, and the model will be tested versus measurements from Denmark and from EMEP monitoring stations over Europe.

As a part of a MSc thesis work, a nested grid version of the forecasting model Eta will be developed and applied with a high resolution for an area surrounding Denmark. The main aim is to achieve a more precise description of the meteorology for this domain such as coastal effects, wind fields and distribution of precipitation amounts.

Calculations of nitrogen depositions to the Adriatic Sea have been proposed. In case funding is obtained, ACDEP will be applied for these calculations.

The mathematical framework of the new REGINA model is in place and tested (see Frohn et al., 2001). Currently the model is being validated with measurements from the EMEP measurements stations network.

Acknowledgements

The work under the Danish Background Monitoring Programme is carried out in close co-operation with Thomas Ellermann at NERI. In the ANICE project, Elisabetta Vignati, currently at Joint Research Centre ISPRA, ITALY developed the aerosol module for implementation in ACDEP. The Nordic Council of Ministers funded the application of ACDEP for the Baltic Sea. Jørgen Brandt and Jesper Christensen are acknowledged for providing meteorological data from the Eta-model.

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Air-Sea Exchange of Nitrogen and Iodine

A Contribution to subproject CAPMAN

Tim Jickells, Alex Baker, Sarah Cornell, Lucinda Spokes and Stuart Yeatman.

School of Environmental Sciences,University of East Anglia,
Norwich, NR4 7TJ, U.K

Summary

Most of the research described here aims to improve our understanding of the transport, deposition and impact of atmospheric nitrogen on marine ecosystems. Recent highlights of our work include:

1. an improved description of the interactions between sea salt, nitric acid and ammonia in the coastal marine atmosphere,
2. the first report of the speciation, size distribution and composition of organic nitrogen in the remote marine atmosphere,
3. the demonstration of the utility of nitrogen isotopes for tracing the sources and transformations of nitrogen in the coastal marine atmosphere,
4. the quantification of the significant contribution of atmospheric nitrogen deposition to the nitrogen requirements of coastal phytoplankton.

In addition we have a significant programme of research into iodine cycling in the coastal marine environment. Recently we have shown the importance of marine macroalgae as a source of gas phase methyl iodide to the coastal marine atmosphere and also described the speciation of aerosol iodine.

Aims of the Research

The overall aim of the research is to better understand atmospheric transport and deposition processes in the marine environment and its effect on marine ecosytems.

Activities During the Year

The main field campaigns have been involved with the EU funded FP5 project MEAD (Marine Effects of Atmospheric Deposition) in the Kattegat. In addition we have been analysing samples and data from previous campaigns, particularly those of the EU FP4 project ANICE (Atmospheric Nitrogen Inputs to the Coastal Ecosystem) and the UK NERC ACSOE project (Atmospheric Chemistry Studies in the Oceanic Environment).

Principal Results

Nitrogen

The deposition rate of fixed nitrogen depends critically on the size distribution of the component of interest in the aerosol. In the coastal zone, air masses of marine and continental origin interact. We have shown that during such interactions the size distribution of continentally derived nitrate and ammonium are significantly altered. This arises from interactions of gas phase nitric acid and ammonia and fine mode aerosol salts containing ammonium and nitrate with coarse mode seasalt. We have documented that in general 40-100% of the nitrate is found in the coarse mode (<1.0 μm in diameter) and under polluted continental conditions 19-51% of the ammonium is as coarse mode aerosol at sites around the North European coast (*Figure 1*). This is the first such systematic documentation of this size shift for ammonium. We have gone on to show that these reactions significantly enhance deposition rates of these nitrogen species, and that such deposition is a quantitatively significant nitrogen source for the phytoplankton living in the coastal sea waters of this region under nutrient depleted summer conditions (Spokes *et al.*, 2000; Yeatman *et al.*, 2001a, de Leeuw *et al.*, in press). We also show that such deposition is dominated by the relatively rare highly polluted episodes.

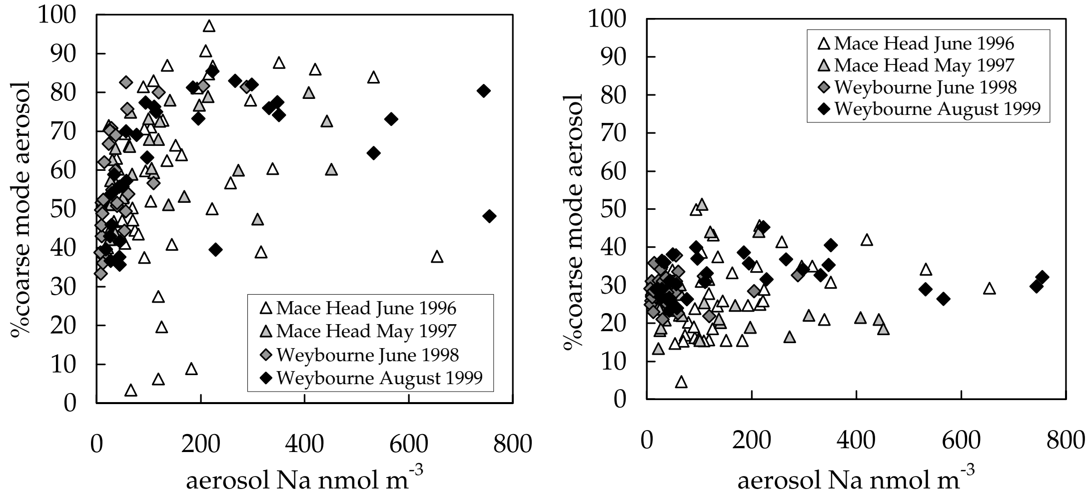

Figure 1. Percentage coarse mode nitrate (left hand plot) and ammonium (right) in aerosols collected at Mace Head on the west coast of Ireland and at Weybourne on the North Norfolk coast of the U.K. as a function of aerosol sodium concentration. At both sites, and at the Meetpost Noordwijk 9 km off the Dutch Coast, coarse mode nitrate and ammonium are a significant fraction of the inorganic nitrogen aerosol load. Under less polluted conditions, such as those seen on the southwest coast of Sweden, little coarse mode ammonium is seen.

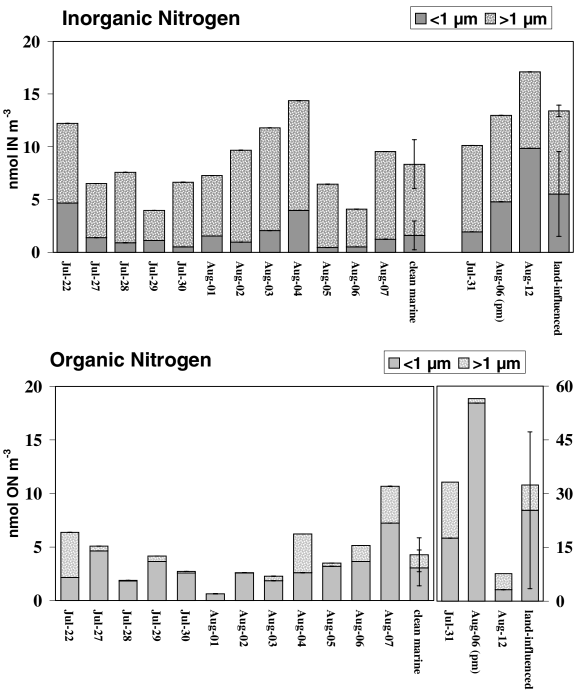

Figure 2. Size segregated aerosol from Oahu, Hawaii, shows the tendency for inorganic and organic nitrogen to be found on different sized particles, with most of the organic nitrogen on fine mode aerosol. The 3 days when local island air was sampled are shown on the right-hand side of each plot, separately from the clean, trade wind aerosol.

We have previously shown that organic nitrogen is an important component of aerosols and rainwater, and we have now shown that organic nitrogen is significant in clean marine air sampled on the coast of Hawaii. This organic nitrogen is primarily associated with fine mode aerosol, less than 1.0 µm in diameter (*Figure 2*, Cornell *et al.*, in press). Urea appears to be an important component of this organic nitrogen, possibly derived from long range transport from Asia where it is used extensively as a fertiliser. Since the organic nitrogen appears to remain associated with fine mode aerosol, while the inorganic nitrogen is transferred to the coarse mode, the atmospheric residence time of organic nitrogen will be greater than that of inorganic nitrogen and hence more liable to undergo long range atmospheric transport.

Nitrogen has two stable isotopes ^{14}N and ^{15}N, the former being the most abundant. Isotopic fractionation of the nitrogen relative to the ratio in atmospheric N_2 can occur during chemical reactions such as combustion processes. Measurement of isotopic ratios in aerosols can, in theory, allow different sources of aerosol fixed nitrogen to be differentiated, as has been done previously for sulphur. We have conducted the first large scale systematic study of nitrogen isotopes in aerosols and documented that there are indeed systematic differences in isotopic composition for both nitrate and ammonium between locations and between air masses of different origin at the same location (*Table 1*, Yeatman *et al.*, 2001 b,c).

Table 1. Isotopic Composition of Aerosol Nitrate Measured at Weybourne on the North Norfolk Coast of the U.K. classified by air-parcel trajectory into different source regions.

	Average Nitrate Concentration ($nmol\ m^{-3}$)	$\delta^{15}N$ $^{o}/_{oo}$
Northern UK	41	$+15 \pm 3$
Southern UK	82	$+10 \pm 3$

Since these are the first such studies reported, the processes controlling the isotopic fractionation are still not fully determined, but the methodology opens the way to a new way by which sources can be apportioned. This use of stable isotope ratios promises to be a valuable tool in air pollution and eutrophication management.

Iodine

We have been involved in a large study of trace gas emissions from waters offshore of the west coast of Ireland. The study showed that the patterns of distribution of various important trace gases such as methyl iodide, dimethylsulphide and isoprene are all different, suggesting different mechanisms by which these gases are formed and released from the water column, although all are connected to phytoplankton activity. This means that is it not possible to scale the emissions of one gas from another with confidence. The pattern of methyl iodide concentrations in the water imply that macroalgae growing in the coastal waters are a major source (*Figure 3*). Other organohalogen gases are also released from these macroalgae and the resulting release of iodine gases can produce significant quantities of IO radicals with important consequences for atmospheric oxidation reactions in the coastal atmosphere (Baker *et al.*, 2000a, Carpenter *et al.*, 1999).

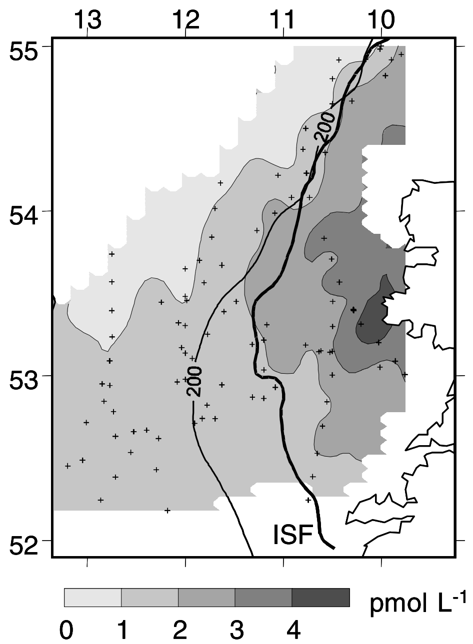

Figure 3. Concentration of methyl iodide in surface waters off the west coast of Ireland, May 1997.

Atmospheric iodine chemistry is complex and poorly understood, but extensive recycling of iodine from the aerosol phase has been proposed to take place. Such recycling assumes the presence of iodide in the aerosol phase. We have conducted a study of the speciation of aerosol iodine in the coastal marine environment. We find that about half of the inorganic iodine is present as iodate and the remainder as iodide. In addition we identified for the first time a significant component that we believe to be high molecular weight and low volatility organic iodine (*Figure 4*, Baker *et al.*, 2000b).

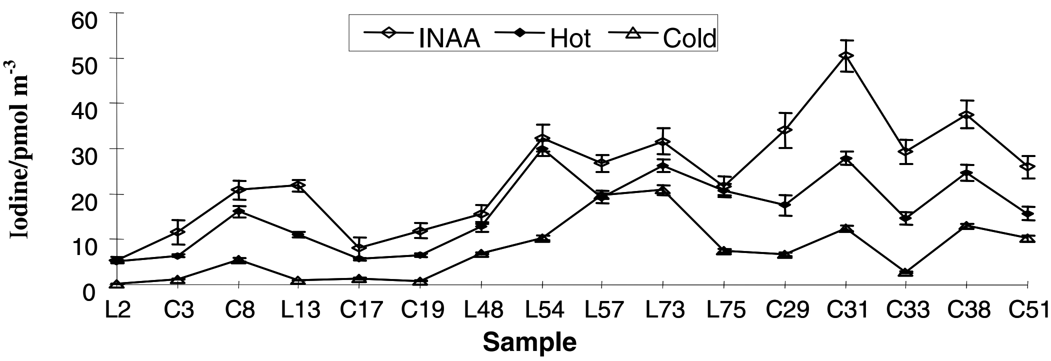

Figure 4. Total aerosol iodine (INAA) and inorganic iodine extracted into aqueous solution at 95°C (Hot) and 20°C (Cold). 'Hot' concentrations are generally higher than 'Cold', possibly because boiling destroys organo-iodine compounds in the aqueous extracts. Based on solubility considerations, non-extracted iodine (Total minus Hot) is also probably organic. (see Baker *et al*, 2000b).

Acknowledgements

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The Effect of Swell on Air-Sea Exchange in the Baltic Sea

A contribution to subproject CAPMAN

Ann-Sofi Smedman[1], Anna Rutgersson[1,2] and Ulf Högström[1]

[1]*Department of Earth Sciences, Meteorology, Uppsala University, Uppsala, Sweden*
[2]*Swedish Meteorological and Hydrological Institute (SMHI), Norrköping, Sweden*

Introduction

Swell are waves travelling faster than the wind. In the open ocean, swell is more or less omnipresent and often omnidirectional. It originates in areas with strong winds but can travel over thousands of kilometers with very little attenuation. Thus, swell observed at a given position in the open ocean at a given occasion can have any direction quite independently of the local wind direction.

Up till recently, the study of swell has received much less attention than wind waves, and its properties are hence less well known. In recent years several studies have, however, highlighted intriguing features of the effect of swell on the turbulent exchange of momentum, sensible heat and water vapor at the air-sea interface, *Smedman et al*. (1994, 1999), *Rutgersson et al*. (2001), *Drennan et al.* (1999), *Grachev and Fairall* (2000).

The Baltic Sea differs from the open ocean in an important respect, the size being small in relation to the size of typical synoptic disturbances which may give rise to swell. This means that usually there is not more than one, well-defined source of swell present at a given time in the Baltic Sea, and that the direction of swell at a certain site is likely to be more or less unidirectional.

Data from several years of measurements of concurrent atmospheric data and wave data from the site Östergarnsholm east of Gotland (see Section 2) have been used for the studies presented here. It is found that swell occurs as often as 40% of the time at this site. Very often the swell comes from a southerly direction. As winds from this sector are common, a situation with swell and wind having roughly the same direction is typical for this site. The studies presented here represent such, rather well defined conditions.

Site and measurements

Östergarnsholm is a low island with no trees, situated about 4 km east of Gotland. On the southernmost tip of the island, a 30m tower has been erected, with its base only about 1 m above mean water level. The tower is instrumented with Solent sonic anemometers at 9, 16 and 25 m above the ground and slow response, ‘profile’ sensors for wind and temperature at 5 levels. Wave height and direction is obtained from a Waverider buoy (run and owned by the Finnish Institute for Marine Research) anchored about 4 km to the South-south-east of the tower at a water depth of 36m. Measurements started in 1995 and have run semi-continuously since then.

The measurements on the tower represent open sea conditions with very long fetch (> 100 km) for the sector from north-east over South to south-west. As shown from the ‘flux footprint’ calculation in *Smedman et al*. (1999), the measurements on the tower are likely not to be influenced by limited water depth outside the island.

Effects of swell on the momentum flux

In numerical models the exchange of momentum at the surface of the sea is usually expressed in terms of the drag coefficient C_D, defined as

$$C_D = u_*^2 / u_{10}^2 \qquad (1),$$

where u_* is the friction velocity and u_{10} the wind speed at 10m height. *Figure 1a* shows, for *unstable conditions*, C_D plotted against the stability parameter *z/L*, where z is height above the water surface and *L* is the Monin-Obukhov length:

$$L = -\frac{u_*^3 T_0}{kg\overline{w'\theta_v'}} \qquad (2),$$

with T_0 mean temperature of the surface layer (K), k = von Karman's constant, g = acceleration of gravity and $\overline{w'\theta_v'}$ the buoyancy flux. The filled symbols represent measurements during swell, open symbols non-swell. The full line is the corresponding standard formulation used in most models. It is seen that the momentum flux observed with swell is generally lower than expected. As shown by *Smedman et al.* (1994), *Grachev and Fairall* (2000) and others, the momentum flux during wind-following swell is sometimes even directed upwards from the ocean instead of, as usually observed, downwards.

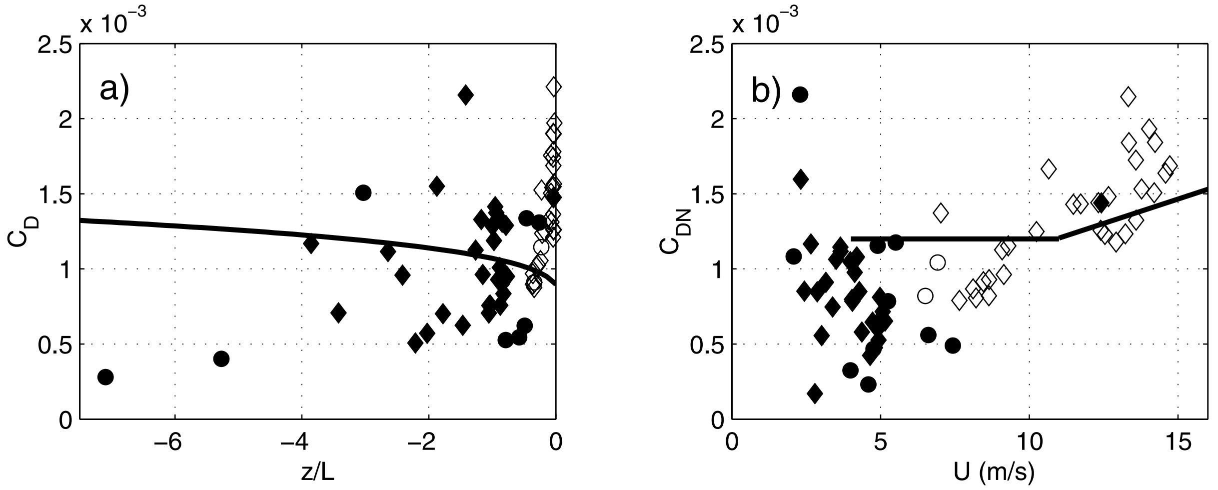

Figure 1. Drag coefficient during unstable conditions, a) C_D as a function of *z/L*; the line was calculated from standard expressions; b) neutral drag coefficient calculated with standard ϕ_m -functions, plotted as a function of 10 m wind speed; the solid line is based on *Large and P*ond (1981). From *Rutgersson et al.* (2001).

In *Figure 1b* C_D has been reduced to its value at neutrality (*z/L* = 0), denoted C_{DN}, with the aid of standard Monin-Obukhov expressions (*Högström*, 1996) and plotted against u_{10}. The full line is derived from *Large and Pond* (1981). Also in this graph it is clear that the swell data are lower than expected. This is, however, not surprising, considering that the dimensionless wind gradient expressions $\phi_m(z/L) = \frac{kz}{u_*}\frac{\partial u}{\partial z}$ observed during swell (not shown here) differ considerably from what is otherwise observed and used in reducing C_D to C_{DN}. A characteristic observed feature during swell in unstable conditions is the occurrence of a low level (< 10 m) wind maximum – a wave-driven wind which gives negative values for

ϕ_m within an appreciable part of the surface layer, *Smedman et al.*(1999). Monin-Obukhov similarity is thus not valid during these conditions.

In *stable conditions*, i.e. when *z/L* > 0, the observed ϕ_m -curves agree much better with what is otherwise observed (not shown here), but nevertheless, C_D is only half its value during non-swell conditions. *Rutgersson et al.* (2001) observe such conditions for u_{10} as high as 8 m s^{-1}.

Exchange of sensible heat and water vapor

The turbulent exchange of sensible heat, *H* is derived in numerical models with a bulk formulation similar to that for the exchange of momentum:

$$H = \rho \overline{w'\theta'} = \rho C_H u_{10}(\theta_{10} - \theta_s) \qquad (3),$$

where ρ is air density, C_H the bulk exchange co-efficient, θ_{10} potential temperature at 10 m and θ_s surface temperature.

Figure 2 presents data for C_H plotted against *z/L* for *unstable* conditions (*z/L* < 0). The measurements show that C_H is only about 10 % lower in the mean than expected from standard expressions (the thin line) during swell. Note, that swell occurs over a very wide stability range, -7 < *z/L* < 0, whereas non-swell conditions, represented by the thick line, is obtained only for *z/L* > -0.5. As noted in Section 3, the friction velocity is strongly reduced during swell conditions and, as seen from the definition of the Monin-Obukhov length, Eq. (2), this will result in a strong reduction in –*L* and correspondingly large values of –*z/L* even for a comparatively small heat flux.

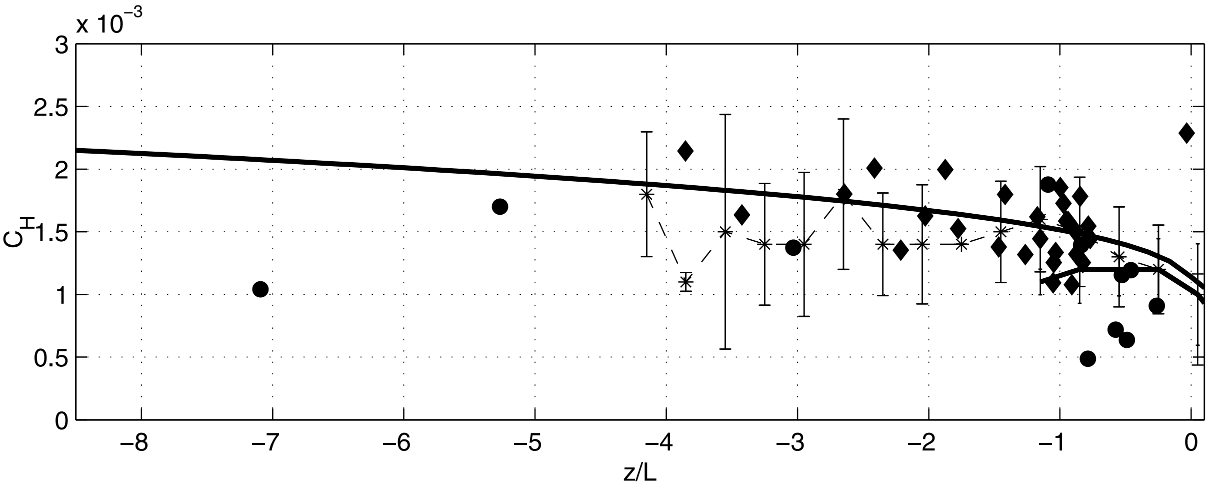

Figure 2. C_H plotted as a function of *z/L* for unstable conditions. Filled symbols represent measurements during swell conditions. The thin solid line has been derived with C_{HN} = 1.1· 10^{-3} and standard ϕ_h - functions. Dashed lines connects averages over *z/L* intervals using all available swell data for 1995 – 1998 in the range –4.5 < *z/L* < 0. Thick solid line is average of all data from the same time period *without* swell. From *Rutgersson et al.* (2001).

Figure 3 shows C_H for *stable conditions* as a function *z/L*. Again the thin line has been derived from standard expressions. The observational swell data are found to be approximately constant ≈ 0.5·10^{-3} over the stability-interval 0 < *z/L* < 1.5, decreasing probably for higher *z/L*. Also the data for non-swell conditions, the short thick line, indicates that C_H ≈ 0.5·10^{-3} for near-neutral stable conditions, rather than about 1.1·10^{-3} as traditionally thought. This lower value was also found by *Davidson* (1974) during swell and by *Oost et al* (2000). Thus, there is almost a discontinuity of C_H at $z/L = 0$. This is probably caused by a feedback effect: stable stratification reduces turbulence, which reduces the waves, which reduces friction further etc. This interpretation is in agreement with the finding reported in Section 3. During stable conditions, C_D is found to be only about half of what is usually assumed in spite of the fact that ϕ_m agrees with what is found over land, indicating that the roughness length is being reduced over sea in stable air by this feedback effect.

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Figure 3. C_H for stable conditions plotted against z/L. Notations as in Figure 2. From *Rutgersson et al.* (2001).

The results obtained for the flux of sensible heat appears to be equally valid for the corresponding exchange of water vapor, but the experimental data are still relatively scarce.

Conclusions

Analysis of several years worth of measurements at Östergarnsholm shows that swell occurs as often as 40% of the time in the Baltic Sea. As the source of swell is in most cases located in a well-defined geographical area of the Baltic, uni-directional swell is often found. At Östergarnsholm, the swell has often approximately the same direction as the local wind. The results given here all represent such conditions.

The exchange of momentum at the water surface is strongly influenced by swell. This results in a reduced bulk exchange coefficient, C_D. Often a wind maximum is found below 10 m (a wave-driven wind), invalidating Monin-Obukhov similarity.

The exchange of sensible heat and water vapor is also affected by swell but not as much as in the case of momentum. At slightly and moderately stable conditions, the bulk coefficient C_H is found to be virtually constant at $0.5 \cdot 10^{-3}$, also during non-swell.

Acknowledgement

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Simulation of Coastal Atmospheric Processes including Aerosols

A contribution to subproject CAPMAN

K. Heinke Schlünzen and L. Klein

Meteorologisches Institut, ZMK, Universität Hamburg, Bundesstr. 55, 20146 Hamburg, Germany

Summary

The dependence of the close–to-coast deposition on the meteorological situation and the relevance of atmospheric processes was studied with the aid of the atmospheric mesoscale transport and fluid model METRAS (Schlünzen, 1990; Schlünzen et al., 1996) and the chemistry transport model MECTM (Müller et al., 2000). The study region was the south-western North Sea, the simulations were performed for the 1998 measurement campaign of the ANICE (Atmospheric Nitrogen Inputs into the Coastal Ecosystem) experiment.

Aim of the Research

The mesoscale model system METRAS/MECTM is applied to simulate coastal atmospheric phenomena including the nitrogen transport and the composition of coastal aerosol.

METRAS is a multi-layer dispersion model which can generate three-dimensional meteorology fields. This model is coupled with the Chemical Transport Model (MECTM) which includes a gas phase chemistry (Stockwell et al., 1997) and the aerosol model SEMA (von Salzen and Schlünzen, 1999a, 1999b, 1999c). SEMA combines a thermodynamic equilibrium approach with a kinetic approach to predict the condensation and evaporation of gases at the surface of the aerosol.

With the model system scenario studies were performed to calculate the atmospheric nitrogen input to coastal waters in a spatial resolution of 8 km and a time resolution of minutes for the ANICE experimental days. At lateral boundaries and to initialise the model, results of larger scale models were used for the meteorology. The model results were compared with available routine data and with the measurements from the ANICE project for the box-model aerosol runs and the METRAS runs.

Activities During the Year

One of the most important components of any regional air quality model is its chemical mechanism. A transport-transformation model must include a gas phase chemical mechanism that incorporates all significant reactions, but must also be simple by comparison with the very complex chemistry of the real atmosphere. The most well known reactions in a photo-chemical model are the chemistry of ozone, nitrogen, and sulphur-containing species. Through a chain of photolytical reactions ozone reacts with H_20 to produce the hydroxy radical (HO) which in turn reacts with both inorganic and organic species. The reactions of HO with SO_2 and with NO_2 are the major gas phase sources of H_2SO_4 and HNO_3, which is a very important loss process for NO_x, and both play a role in the formation of coastal aerosols.

NH_3, after N_2 and N_2O is the most abundant compound of nitrogen in the atmosphere (Seinfeld, 1998). NH_3 can react with OH, but as it is highly soluble and reactive with atmo-spheric acids, that is the preferable route for the removal of the gas phase from the atmo-sphere. NH_3 and SO_2 are rapidly taken up by aerosols and are therefore relevant when in-vestigating deposition in coastal areas.

The METRAS/MECTM system solves:

- Equation of 3D motion in a surface following co-ordinate system,
- Conservation of energy,
- Budget equation for water (gas, cloud, rain), and
- Budget equation for the tracer gases including emissions, transport, transformations and deposition.

The MECTM uses the Regional Acid Deposition Model (RADM2) gas phase chemical mechanism as described in Stockwell et al., 1990. The RADM2 uses a reactivity lumped molecular approach and includes the above mentioned gas phase reactions as well as alkenes and ketones. Alkenes (ie. Ethene, propene) are important constituents of the polluted and rural troposphere. They are very reactive species that have relatively high rate constants for reaction with HO radical, and they react with ozone to produce aldehydes. Ketones represent a significant sink of carbon since they are less reactive and are transported over long distances.

In the MECTM calculations are made with consideration to gaseous emissions, vertical and horizontal dispersion, chemical transformations, removal of species due to deposition. For solving the stiff chemical equation system the hybrid solver (Müller et al., 2000) was selected for use in the chemistry model so that it would be compatible with the aerosol modules used in later runs. In this solver, the iteration procedure is continued until the convergence criterion is satisfied for all species. The time step used in the chemistry is 220 - 260 s (3-4min).

Anthropogenic emissions for both point and area sources were obtained from the University of Stuttgart, Institute for Energy Economics and the Rational Use of Energy. These include emissions from industry, residential, and a mobile emissions inventory (including running losses, cold-starts, etc) as well as agriculture (NH_3 emissions). The data was aggregated to the METRAS grid and converted to the units that would be necessary for the gas phase chemistry reactions.

The biogenic emission factors are calculated in MECTM by a program which uses the land use classification data and the emission factors from McKeen et al. (1991). A biogenic emission inventory data file was created with the factors stated in kilogram per model grid cell per hour.

In order to accurately model and predict the effects of air pollution, photo-dissociation reaction rate estimates must be made. The simulation accuracy of the entire chemical system is highly dependant upon the accuracy of the photolysis rates, which are the primary source of radicals in the atmosphere. The current approach taken for setting photolysis rates in the METRAS/MECTM was based on the System for Transfer of Atmospheric Radiation (STAR) model developed by Ruggaber (1994). It included two stages of processing: (1) a table of clear-sky photolysis rates was calculated for 21 different gases using a pre-processor which calculates the rates for each photolysis reaction and each hour based on meteorological information extracted from the METRAS runs; and (2) photolysis rates were interpolated from the pre-generated table by the MECTM based on grid cell location and model time. These rates are then matched to the RADM2 mechanism in the chemical transformations during the chemistry run.

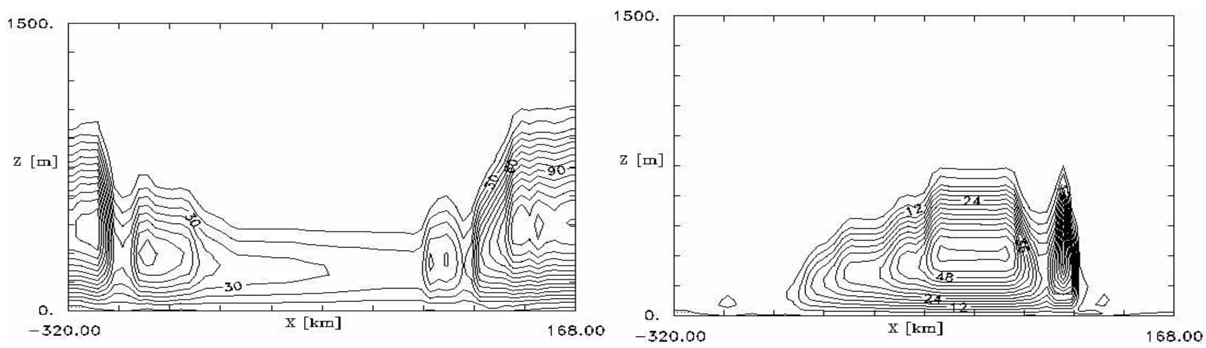
The results of the model runs were validated against observations using a series of programs developed for that purpose. These programs interpolated the METRAS results to the locations of the observation stations and then various statistical methods were applied. The results of this analysis have been discussed in the the previous CAPMAN annual report (Schlünzen et al., 2000). In section 4 the results of the deposition study are discussed.

Principal Results and Main Conclusions

There are four main factors governing dry deposition in the atmosphere:

- concentration in the atmosphere,
- atmospheric turbulence,
- chemical properties of the depositing species, and
- nature of the surface/receptor.

The turbulence near the ground affects the deposition rate to the surface, the solubility and chemical reactivity at the surface may influence the uptake, and surfaces may be non-reactive for gas absorption/adsorption. They may be smooth enough for particles to bounce off, or as in the case of vegetation, may be highly susceptible to deposition. In the model, consideration is given to atmospheric transport processes as well as to removal mechanisms and the physical and physiochemical properties of gases.

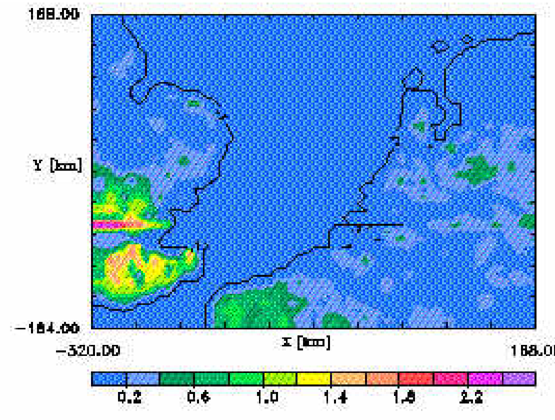


a) Daytime, 14:00, June16, 1998. b) Night-time, 23:00, June 16, 1998.

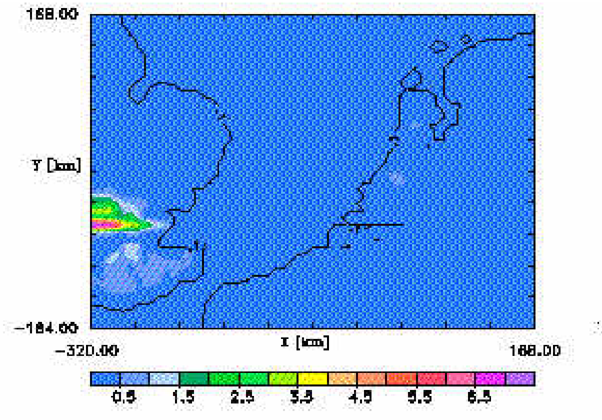
Fig 1. Vertical cross-section to 1500m height of vertical exchange coefficients during the day (a) and during the night (b). The North Sea lies in the middle and is bounded by England and the Netherlands. (not to scale)

The MECTM/METRAS system was run for the period of June 16-20, 1998. The 3-dimensional model allowed for simulating complex atmospheric flows. Dispersion and deposition processes in the coastal region are made complex by the flow changes resulting from the abrupt difference in surface characteristics between land and water which can best be simulated by using a 3-D model system. One example of the differences which strongly affect the atmospheric flow is the roughness length. The sea surface roughness is at least several orders of magnitude smaller than the agricultural land in the region so the mechanically generated turbulence is lower. There is also a difference in thermal convection which is partly due to land being warmer than the North Sea so there is more vertical motion in the atmosphere above. These inconsistencies between land and sea contributes to more vertical mixing of pollutants over land than over water. This was well simulated by the MECTM (*Fig 1*). During the day there is intense vertical mixing over land which compensates the higher emissions coming from urban areas like London, and agricultural areas like those located in the Netherlands.

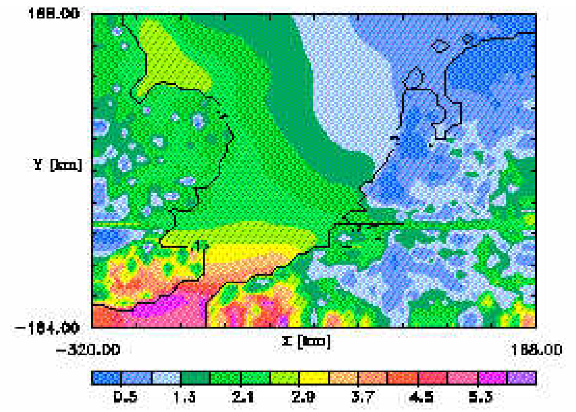
Deposition values for hourly and daily amounts were calculated. *Fig. 2* shows the daily deposition values for various nitrogen compounds. The highest concentration modelled was clearly N-NO, but the greatest amount deposited to the waterway is N-HNO_3 due to its solubility in water. The high values of N-NH_3 deposited over land are largely a result of the emissions from the agricultural regions in the modelled area.



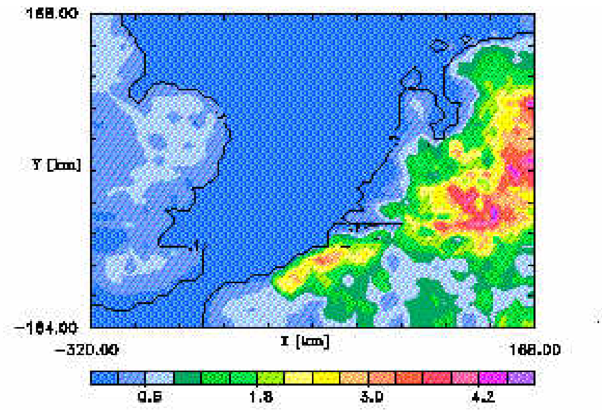
a) N-NO_2 daily deposition, June 16.



b) N-NO daily deposition, June 16.



c) N-HNO_3 daily deposition, June 16.



d) N-NH_3 daily deposition, June 16.

Fig 2. Daily deposition values for various nitrogen compounds, June 16, 1998 (mol/m^2/day).

Deposition differs over water and over land due to the different surface resistances that are present. The ANICE model area comprised mainly sea surface and agricultural land use and the effect of the different surfaces on deposition can be seen. The less soluble species have greater deposition over land (i.e. N-NO_2), while the more soluble species such as N-HNO_3 deposited over the water as well as land. The greatest deposition occurred downwind from the London urban area as a results of the high concentration of emissions in that region at 16 June, 1998.

Aim for the Coming Year

The aims for the coming year are to continue the analysis of the effect of chemical reactions on deposition amounts and patterns. This includes investigating the effect of both wet and dry aerosols on deposition.

Acknowledgements

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Air-Sea Exchange of Nutrients, Trace Metals and Organic Micro Pollutants at the North Sea

A contribution to subproject AEROSOL/CAPMAN

René Van Grieken and Kurt Eyckmans

University of Antwerp, Universiteitsplein 1, 2610 Wilrijk, Belgium

Summary

As part of a project concerning the air-water exchange of nutrients and inorganic micropollutants to the North Sea, the atmospheric deposition of nutrients and trace metals via various forms and pathways was investigated. This was done by intensive sampling campaigns on the research vessel Belgica, combined with a sampling campaign over a period of one year at a sampling point at Knokke-Heist, near the Dutch border, and for a period of three months at Adinkerke, near the French border. Samples were taken for nutrients, trace elements, rain water and gases.

Aim of the period's work

The sample treatment and analysis of the different kinds of samples was already optimised. In this period the main goal was to achieve a large dataset over a sampling period of more than one year.

Activities during the year

Most of the samples, taken in 2000, are analysed.

Due to heavy weather during the Belgica campaigns (ns 9/00 from 27/3 until 31/3 (terminated on 29/3 due to storm) and ns 24/00 from 2/10 until 6/10), almost all the samples were contaminated by sea spray. The campaign at the sampling point at Knokke-Heist in Het Zwin (at a distance of 500 m from the sea and at a distance of 2 km of the Dutch border) was terminated succesfully at the end of May. This sampling campaign had a duration of almost one year and the samples were taken on a weekly basis. Intensive sampling periods of one week were also organised in order to investigate the daily variations.

In the beginning of June, the sampling equipment was removed to Adinkerke, a sampling station near the French border at a distance of 5 km to the sea. There, the pollution of the atmospherical particles over the French-Flemish North Sea coast was investigated, with special attention to the borderline transports and the impact on the environment. The sampling was terminated at the end of July and restarted for the month of December.

Almost all the analysis have been carried out and these data will be used, in combination with meteorological data, for the calculation of fluxes.

Principal results

4.1 Belgica sampling campaigns

The first sampling campaign on board of the research vessel Belgica during the ns 9/00 (from March 27 to 31, 2000) had to be aborted on the 29th of March due to stormy weather. All the filter samples were ruined due to seaspray blown over the ship. There were also two samples taken, using a May impactor. All the different stages were leached and the results of one of these samples is depicted in *Figure 1.*

The second sampling campaign on the Belgica (ns 24/00 from October 2-6) was also not a big success. There was a wind speed of 8-9 Bft and only a fraction of the sampling could be carried out.

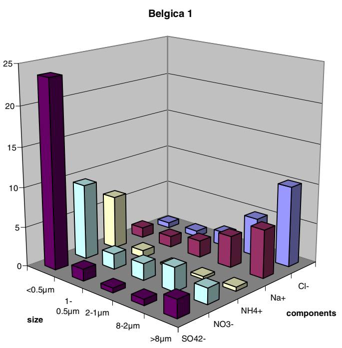


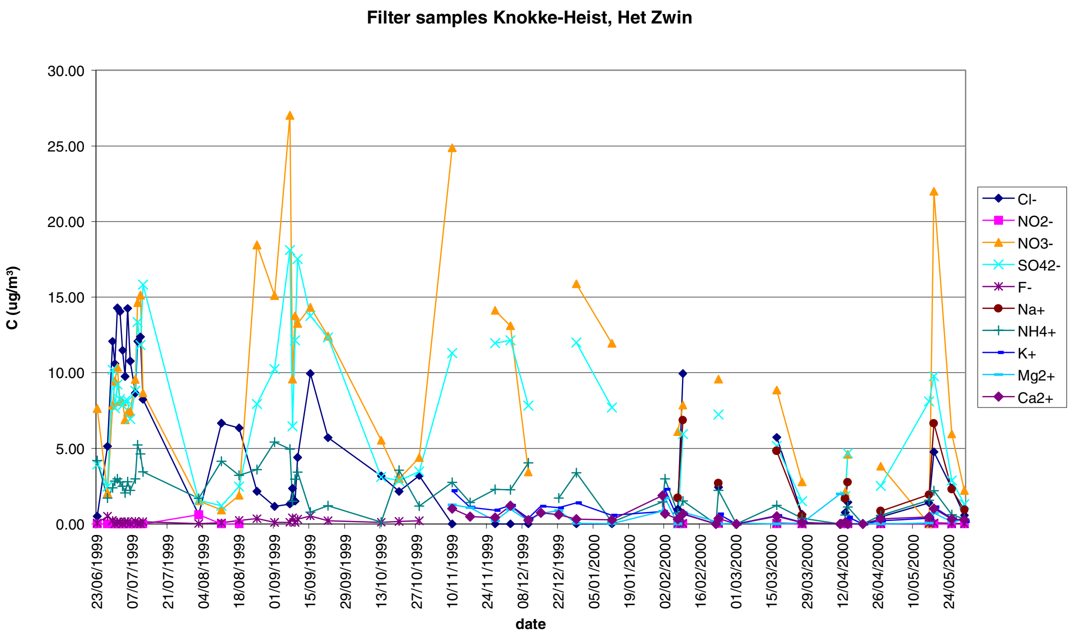
Fig 1: On the first Belgica sample (March 27, Belgian Continental Shelf) we can see a clear marine influence (coarse sea salt particles), combined with a continental history (small secondary sulphate, nitrate and ammonia aerosols).

4.2 Sampling at the Belgian Coast

4.2.1 Particulate matter

A) Nutrients

The sampling started on June 23, 1999 and was terminated at the end of May 2000. Over one week, around one thousand cubic meters of air were sampled, using a high-volume pump, and the particles were collected on a glass fibre filter. Afterwards, the particles were dissolved in Milli-Q water and analysed by Ion Chromatography. In *Figure 2*, the weekly averages of the different concentrations are depicted.

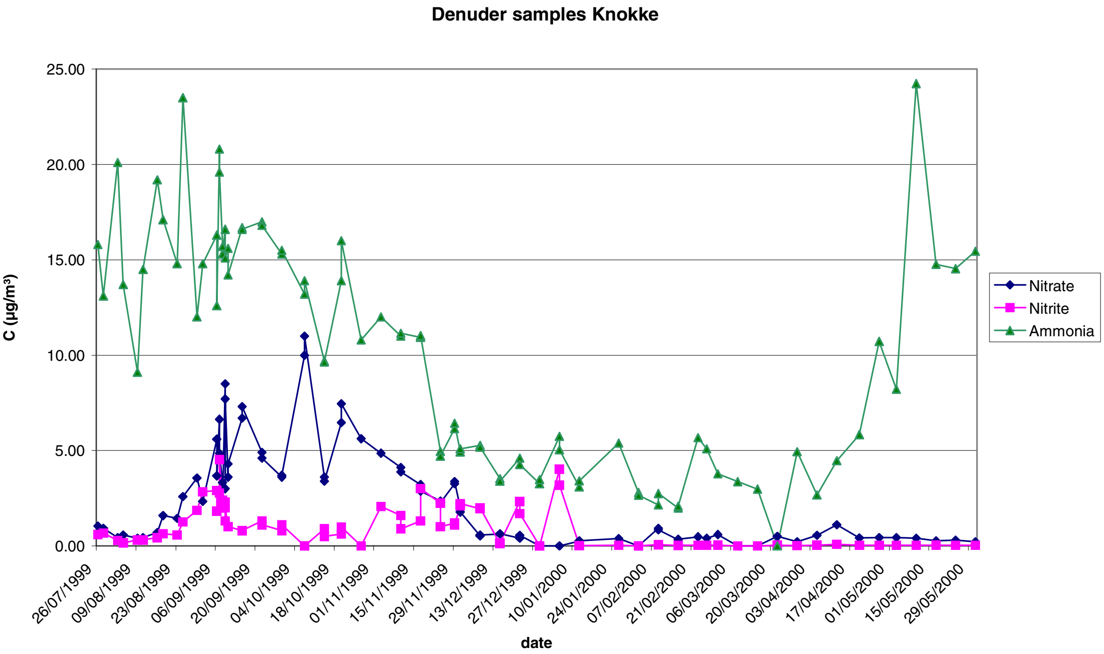


B) Trace metals

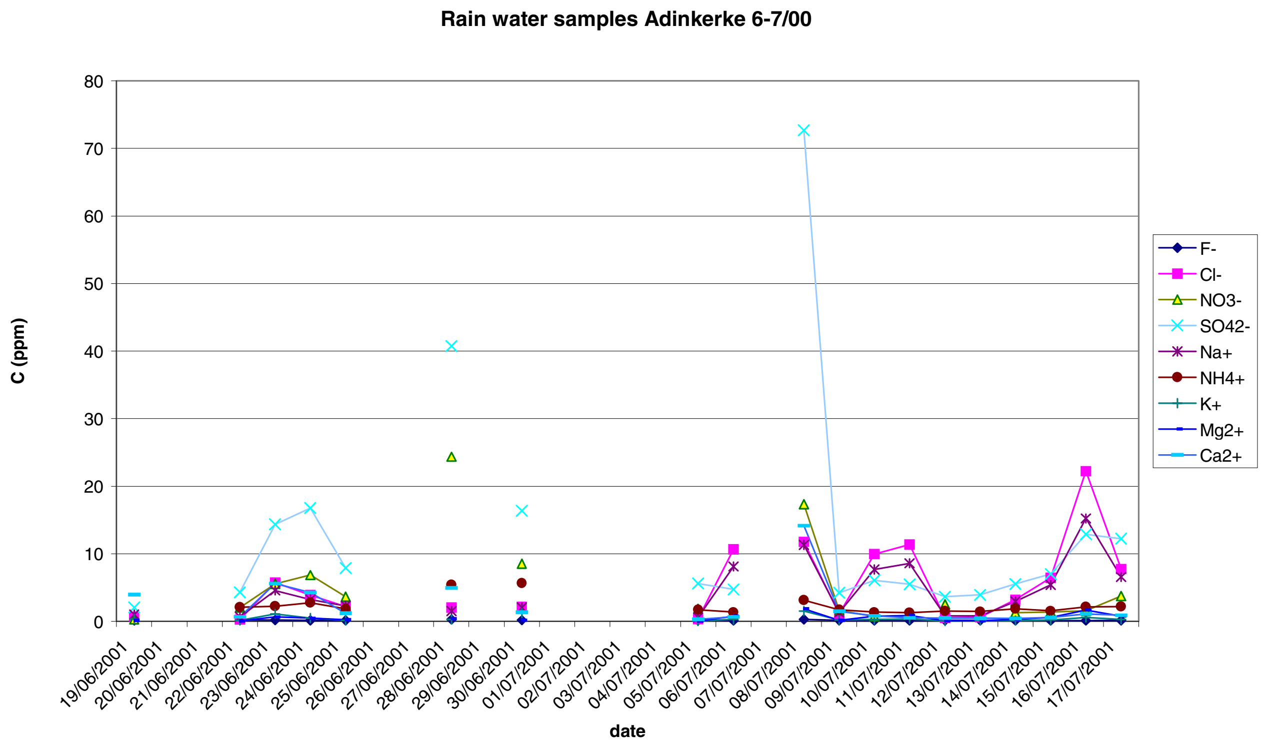
Trace metal samples were analysed but the results still have to be interpreted.

4.2.2 Gases

Gases were sampled with an annular denuder system, URG, 2000-01-K. Here, we can see a very high concentration of ammonia during August and September, decreasing over the winter period and increasing sharply in the beginning of May as a result of the fertilisation of the fields. The higher temperatures can also be responsible for the decomposition of ammoniumsalts, resulting in higher concentrations during spring and summer. The nitric acid concentration was rather low before September, and had normal to rather high concentration values in the period September-October.



4.2.3 Rain water



Rain water was collected over the months June and July, using an automatical sequential rain water sampler. The data have to be compared with the meteorological data and wet fluxes can be calculated.

Main conclusion

Almost all the sampling has been carried out and the results seem to be comparable with values found in literature. The sampling period lasted almost one year and major conclusions can be drawn. For the particulate fraction, there are clear fluctuating concentrations for all the different compounds, due to the changing wind direction and marine or continental influences. For the gases, and especially for ammonia, there is a clear seasonal variation in the concentrations. During spring and summertime, there is a higher concentration due to agricultural activities and the higher temperatures, with a sharp increase in these concentrations during the first fertilization activities in spring. Wet depostion fluxes can directly be calculated when the rain water results will be compared with meteorological data.

Aims for the coming year

Two sampling campaigns of one month, during February and June, are planned in Adinkerke. Most of the time will be used for the calculation of wet and dry atmospherical fluxes and the major results and conclusions will be drawn.

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ANNEX: Names and details of current principal investigators

Dr. Hugh Coe
UMIST
Dept. of Atmospheric Physics
P.O. Box 88, Manchester M60 1QD
UK
tel. +44 161 200 3935
fax. +44 161 200 3941
email. Hugh.Coe@umist.ac.uk

Dr. Serge Despiau
Faculté des Sciences et Techniques
Université de Toulon Var
F-83957 La Garde Cedex
France
tel. +33 94 14 23 63
fax. +33 94 14 25 72
email. despiau@isitv.univ-tln.fr

Dr. D. P. Eppel
Forschungszentrum Geesthacht
Institut für Physik
Max-Planck-Strasse, D-21502 Geesthacht
Germany
tel. +49 4152 87 1564
fax. +49 4152 87 1525

Dr. Gerald Geernaert
National Environmental Reseach Institute
Frederiksborgvej 399, DK-4000 Roskilde
Denmark
tel. +45 46 30 11 01
fax. +45 46 30 12 14
email. glg@dmu.dk

Prof. D. R. van Grieken
Centre for Micro- and Trace Analysis
Department of Chemistry,
University of Antwerp (UIA)
B-2610 Wilrijk
Belgium
tel. +32 8 820 2362
fax. +32 8 820 2376
email. vgrieken@schs.uia.ac.be

Dr. Ole Hertel
National Environmental Reseach Institute
Frederiksborgvej 399, DK-4000 Roskilde
Denmark
tel. +45 46 30 11 48
fax. +45 46 30 12 14
email. oh@dmu.dk

Prof. Dr. C. N. Hewitt
University of Lancaster
Institute of Environmental and Natural Sciences
Lancaster LA 1 4YQ
UK
tel. +44 1524 593931
fax. +44 1524 593985
email. n.hewitt@lancaster.ac.uk

Dr. Tim Jickells
University of East Anglia
School of Environmental Sciences
Norwich NR4 7TJ
UK
tel. +44 1603 456 161
fax. +44 1603 507 719
email. tjickells@uea.ac.uk

Dr. Søren Larsen
Risø National Laboratory
Dept. of Meteorology and Wind Energy
P.O. Box 49, DK-4000 Roskilde
Denmark
tel. +45 46 77 5012
fax. +45 46 77 5970
email: soeren.larsen@risoe.dk

Dr. Gerrit de Leeuw
TNO-FEL
Physics and Electronics Laboratory
P. O. Box 96864, NL-2509 JG The Hague
The Netherlands
tel. +31 70 3264221
fax. +31 70 3280961
email. deleeuw@fel.tno.nl

Dr. Kevin Noone
Stockholm University
Department of Meteorology
S-106 91 Stockholm
Sweden
tel. +46 8 162664
fax. +46 8 157185
email. zippy@misu.su.se

Prof. Doz. Dr. K. Heinke Schlünzen
Meteorologisches Institut
Zentr. für Meeres-und Klimaforschung
Universität Hamburg
Bundesstrasse 55, D-20146 Hamburg
Germany
tel. +49 40 42838 5082
fax. +49 40 42838 5452
email. schluenzen@dkrz.de

Dr. Michael Schulz
Chemical Institut
Universität Hamburg
Bundesstrasse 55, D-20146 Hamburg
Germany

Prof. Ann Sofi Smedman
Uppsala University
Department of Earth Sciences
Villavagen 16, S-75236 Uppsala
Sweden
fax. +46 18 55 11 24
email. annsofi@big.met.uu.se

Dr. Lise Lotte Sørensen
Risø National Laboratory
Dept. of Meteorology and Wind Energy
Frederiksborgvej 399, DK-4000 Roskilde
Denmark
tel. +45 46 77 5015
fax. +45 46 77 5045
email. lotte.geern@risoe.dk

Dr. Gunilla Svensson
Stockholm University
Department of Meteorology
S-106 91 Stockholm
Sweden
fax. +46 8 157185
email. gunilla@misu.su.se

Dr. Francisco Tampieri
Instituo per lo studio della Metodologie Geofische
Ambientali
CNR
Via Gobetti 101, I-40129 Bologna
Italy
tel. +39 51 639 8016
fax. +39 51 639 8132
email. franz@alice.fisbat.bo.cnr.it

Dr. Gianni Tinarelli
ENEL s.p.a. – CRAM
via Rubattino, 54, I-20134 Milano
Italy
tel. +39 2 7224 3936
fax. +39 2 7224 3915
email. tinarelli@cram.enel.it

Dr. Michael Tjernström
Stockholm University
Department of Meteorology
S-106 91 Stockholm
Sweden
fax. +46 8 157185
email. michaelt@misu.su.se

Dr. Elisabetta Vignati
Joint Research Centre
Environmental Institute
TP 460
I-21020 Ispra
Italy