

## **Air-Sea Exchange Studies at the North Sea.**

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### **1. Introduction**

The North Sea can be considered as a local 'inner' sea in which many processes are quite different from these over the open ocean. The surrounding land has a major influence, being the source for man-made aerosols and gases, whereas the North Sea acts as a sink for these. At the same time the North Sea is a source for marine aerosols and can be a source for biochemically produced gaseous species. When an air mass crosses the land-sea transition, the sudden change in surface properties results in a non-equilibrium condition between the water and the atmosphere, causing strong changes in the fluxes of momentum, heat and water vapour that drive the meteorological system.

Since almost two decades the TNO Physics and Electronics Laboratory has been involved in experimental studies on air-sea exchange processes at the North Sea. The emphasis was historically on the aerosols in the North Sea atmosphere, a complicated mixture of antropogenic and sea spray aerosol. Models have been developed describing the aerosol concentrations as function of meteorological parameters and air mass history (fetch). Studies on the lateral variation of the aerosol are conducted. In addition, techniques were developed to study the air-sea exchange of momentum, sensible and latent heat, water vapour and CO<sub>2</sub>. Both micrometeorological and geochemical measurements are made. To better understand the underlying processes, the influence of breaking waves is taken into account. Also size distributions of bubbles are measured in the water. Bubbles produce sea spray aerosol, and they are known to influence the gas transfer rate. However, the exact mechanisms and the magnitude of the influence of bubbles are as yet poorly known.

In addition to local measurements in the atmospheric and oceanic surface layers, remote sensing techniques are applied, mainly for boundary layer studies. Lidar (Light Detection And Ranging) is used to study boundary layer dynamics and surface characteristics, including optical properties. Satellite data are used to retrieve information on aerosols. From their variation in time and space, air mass properties are derived to describe their dispersal.

In all these studies, experiments and modeling are combined. The experimental data are used to develop and validate both empirical, numerical and physical models. Empirical models usually apply only in the area for which they were developed. However, they give some insight into the parameters that are important for the description of the observed phenomena, in particular when data are used from different locations with different characteristics. Physical models describe the underlying processes. If this is done correctly, they are applicable at different locations.

Below we summarise some of the studies made by the TNO Physics and Electronics Laboratory (TNO-FEL). A large bibliography is available on each of the topics discussed. In view of the limited space, references are not included in this contribution.

### **2. Aerosols**

Sea spray aerosol is produced at water surfaces by the interaction between wind and waves. Breaking waves produce bubbles that rise to the surface, break and produce film droplets and jet droplets. In high wind speeds, typically larger than about 9 m/s, spume droplets are produced by direct tearing from the wave crests. Although in principle these mechanisms are known, our current knowledge of the aerosol surface source function is still poor and estimates vary by several orders of magnitude.

Nevertheless, the aerosol concentrations in the atmosphere over the open ocean can be reasonably well predicted with the Navy Aerosol Model (NAM) [Gathman, 1989] using simple meteorological parameters like wind speed, relative humidity and the residence time of the air mass over the ocean. However, in coastal regions, such as the North Sea, the NAM predictions are usually poor. Comparisons with particle size distributions measured at Meetpost Noordwijk (MPN), a tower at 9 km from the Dutch coast, shows that large discrepancies occur due to the influence of aerosol advected from land.

To describe the aerosol concentrations at MPN, wind direction sectors had to be identified for different source regions. For each sector an empirical model was developed. All these models together form the TNO-FEL HEXOS aerosol model. It is based on 8 weeks of continuous measurements of aerosol particle size distributions and meteorological parameters during the HEXOS (Humidity Exchange Over the Sea) experiments in 1986.

The HEXOS model was validated with data collected during the MAPTIP (Marine Aerosol Properties and Thermal Imager Performance) experiments organised by TNO-FEL in 1993. During MAPTIP, aerosol data were collected by TNO-FEL on the MPN and also on a ship. The latter allowed us to also estimate the validity of the HEXOS model at some distance from the MPN. It appeared that the performance rapidly decreased with distance from MPN. This was not surprising since the HEXOS model is empirical and a large variety of sources is present in the densely populated and industrialised western part of The Netherlands, including the ports of Rotterdam and Amsterdam and some large cities, mixed with heavily used agricultural areas.

We also used the ship data to estimate the lateral variability of the aerosol in an air mass advected over the North Sea, with the MPN data as reference. Changes of both the aerosol concentrations and the meteorological parameters were clearly observed, and time lapses between the changes at MPN and the ship were close to the travel time of the air mass. Also the changes in chemical composition were as expected. However, the data set is too poor to identify the time constants for these changes in this complicated area. To this end, a dedicated experiment is required, combined with an appropriate modeling effort in which both the meteorological fluxes are described and aerosol physics and chemical reactions in both the gas phase and on the aerosol surface are included.

The HEXOS aerosol data were collected in the first place to identify the influence of sea spray aerosol on the exchange of water vapour between the water and the air. Several publications on this subject are available, either indicating that aerosols do have an influence, or, in contrast, that they don't have an influence. This discussion continues and new experiments in very high winds are required for a conclusive result.

Other applications of aerosol research over the North Sea than in the field of meteorology (water vapour and heat fluxes) are in the field of climate (aerosols scatter and absorb radiation and calculations show that their effects may be large enough to balance the global warming effect of greenhouse gases), air and water pollution and effects on the North Sea eco-system (atmospheric inputs contribute significantly to the total transport of pollutants) and in the field of electro-optic propagation (electro-optical detection systems are limited by the extinction due to aerosols).

### **3. Fluxes of CO<sub>2</sub>, momentum, heat and water vapour**

The air-sea exchange of momentum, heat and water vapour are important processes that determine the dynamics in the atmospheric boundary layer over the ocean. Knowledge on the air-sea exchange of gaseous species is important to understand the atmospheric effects on the oceanic eco-system and to understand the influence of the oceans on climate. TNO-FEL has assembled a micrometeorological flux package for the measurements of the basic air-sea fluxes, momentum, heat and water vapour, and CO<sub>2</sub>, one of the important greenhouse gases. CO<sub>2</sub> is a slowly dissolving gas for which the oceans act as a reservoir. The basic fluxes are needed to determine the CO<sub>2</sub> fluxes from micro-meteorological measurements. The North Sea may not be very important as a reservoir for CO<sub>2</sub> on a global scale. Our measurements at the MPN are mainly aimed at the developments of techniques, and to understand the mechanism of air-sea gas transfer (in the framework of international projects such as ASGASEX and ASGAMAGE).

Two methods are applied. The eddy correlation method is based on the determination of an ensemble average of the product of the turbulent components of the vertical air flow,  $w'$ , and the turbulent fluctuating component of the quantity of interest,  $x'$ :  $\langle w' \cdot x' \rangle$ . The inertial dissipation method is based on spectral analysis of the turbulent quantities, i.e. their power spectra and their co-spectra, to determine the inertial dissipation rate from which the fluxes can be determined. Both methods require measurements of the quantities of interest,  $x = X + x'$ , at a frequency high enough to resolve the inertial dissipation rate (usually up to 10-20 Hz), and with sufficient sensitivity. Here  $x$  is the measured component of the air flow (the horizontal components  $u$  and  $v$ , and the vertical component  $w$ ), temperature  $t$ , absolute humidity  $q$ , or the concentration of the gas of interest  $c$ .  $X$  is the mean value and  $x'$  is the turbulent fluctuating value. For measurements of  $\text{CO}_2$  with commercial instrumentation the sensitivity is often a problem over sea because the fluxes are low. Elsewhere new instrumentation is under development for research applications that is expected to have enough sensitivity. Awaiting the availability of these new instruments, another method is developed by TNO-FEL in cooperation with Risø National Laboratory (Roskilde, Denmark). This method is based on the use of two instruments to determine the  $\text{CO}_2$  scaling parameter  $c$  from the co-spectra obtained by cross-correlating the signals from the two instruments, thus reducing noise contributions.

Alternatively, fluxes of  $\text{CO}_2$  can be determined from measurements of the partial pressures of  $\text{CO}_2$  in the water and in the air. The measurement requires a value for the exchange coefficient. Recently we constructed an equilibrator system to apply this geochemical technique, both in laboratory measurements to study processes, and over the ocean for comparison with micro-meteorological measurements and to study the mechanism of air-sea gas transfer for this slowly dissolving gas.

#### 4. Breaking waves and bubbles

Breaking waves generate bubbles which in turn produce aerosols and enhance gas transfer rates. Aerosols are produced when the bubbles rise to the surface and the surface film opens. Recent laboratory experiments by Dr. Spiel (Naval Postgraduate School, Monterey, CA) show that the film cap rolls up and when enough momentum is gained large droplets separate off. Subsequently, the rising jet produces droplets. The enhancement of the gas transfer rate by breaking waves and bubbles can involve several mechanisms including turbulent transfer in the water, bubble mediated transport and the disruption of the surface layer.

Bubbles are measured with an optical technique using a diode laser to illuminate an open sample volume that allows for free flow. The sample volume is confined by conical tubes, such that the whole volume can be monitored with a small ccd camera. Bubbles in the sample volume cast a shadow on the ccd camera. Image processing techniques are applied to derive the diameter of each individual bubble, and to discriminate them from other objects (e.g., algae) in the sample volume.

This bubble measuring system, developed and constructed by TNO-FEL, is mounted on a float at a depth between 0.35 and 2 m below the instantaneous water surface. Two options are available for the deployment of this system. A large buoy is used with a battery pack and a television transmitter for data transfer to a receiver at a distance of maximum 10 km. A smaller buoy is available with cables for power supply and data. This smaller buoy is used for short term deployments and deployments in areas where tv transmission is not possible.

The BMS has been deployed at several sites in the North Sea, the North Atlantic and in the Danish coastal waters. The bubble spectra are similar to spectra published in the literature in similar ambient conditions, i.e. they are in the same range of concentrations. However, large variations have been reported in bubble concentrations, the shape of the bubble size distributions and the dependence of the bubble concentrations on ambient conditions (e.g., wind speed).

The aim of our bubble measurements is to model the bubble size distributions as function of ambient conditions (atmospheric and oceanic parameters), in relation to sea spray aerosol production and air-sea gas transfer. Therefore, the BMS is usually deployed to measure both bubble size distributions, aerosol concentrations, aerosol profiles and  $\text{CO}_2$  fluxes. The first results indicate that the shape of the bubble size distributions varies with wind speed, and the wind speed dependence of the concentrations is a function of bubble size. At the North Sea the situation is more complicated and no clear relations

have thus far been observed. One factor that likely plays a role here is the fetch, but also other factors may be important.

## **5. Remote sensing**

### **5.1 Lidar**

Optical remote sensing techniques involving lidar are used by TNO-FEL in studies on the boundary layer dynamics, i.e. turbulent and convective mixing processes, evolution of the boundary layer height and the depth of the entrainment layer, to measure wind speed and wind profiles, to detect and follow plumes, and to measure the boundary layer backscatter and extinction properties. A large number of lidar systems has been built by TNO-FEL in the last two decades. Presently, two lidar systems are in use, both working at 1.06  $\mu\text{m}$ . The TNO mini-lidar is a modified laser range finder with a low repetition rate. Hence this lidar can be used to study relatively slow processes such as the evolution of the boundary layer height and to measure the aerosol optical properties.

For faster measurements the TNO SMAL lidar with a repetition rate of about 10 Hz is available. This repetition rate allows for the resolution of structures that in turn allows for the determination of wind speed and mixing properties using advanced processing techniques. Effluent plumes can be detected and their size can be scanned.

The lidars have been applied in several studies in which the above concepts have been developed and the techniques have been proven to work. At present the lidar is mainly used as a tool to probe the boundary layer for application of the results in process studies involving atmospheric chemistry and aerosols.

### **5.2 Aerosol retrieval from satellite data**

Aerosol optical properties are retrieved from data obtained from optical instruments in satellites such as GOME and ATSR in ERS-2 and AVHRR in the NOAA-14 satellite. In these retrieval algorithms a radiative transfer model is applied. Application of aerosol retrieval algorithms requires a completely cloud-free pixel, a severe constraint. A further constraint is that the technique can thus far only be applied over terrain that provides a homogeneous dark background. The sea provides such a background and hence aerosols can be retrieved over water surfaces.

The aerosol optical depth can be visualised with imaging techniques. Hence an aerosol plume flowing out from land to sea can be identified. This technique is presently tested with AVHRR data for the Pacific Ocean off the Californian coast where the Los Angeles plume can be clearly observed. Ground truth data are measured with a ship following the plume to validate the aerosol retrieval and to study relevant processes and parameters that can be used to characterise the air mass and its evolution off the coast. The aim is to provide a parameterisation for application in other models.

## **6. Conclusion**

A brief review has been given of activities of TNO-FEL in atmospheric research, with emphasis on North Sea research. The techniques are applied for a variety of air-sea interaction studies, supported by modeling activities to understand the relevant processes. Both numerical and physical models are developed. Further, the facilities are used to develop instrumentation and techniques for process studies. Problem areas are the coastal influence on the processes in the North Sea atmosphere, and the influence of land-sea transitions. Atmospheric inputs into the North Sea are significant.

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