# MARINE AEROSOLS IMPACT ON ATMOSPHERIC CHARACTERISTICS OVER OCEAN SURFACE IN FRONTAL ZONES

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# ABSTRACT

Aerosols constitute one of an important component of the natural system at the global level. They have a direct radiative forcing due to scatterring and absorbing solar and infrared radiation in the atmosphere. Aerosols also alter the precipitation rate and formation of liquid water, ice and mixed-phase clouds, thereby causing an indirect radiative forcing associated with changes in cloud properties.

To evaluate the partidular degree of the marine aerosols impact on weather conditions, a numerical experiment with the Harmonie model was performed on a domain covering the North Atlantic. The experiment included couples of runs with an without marine aerosols. The comparison between the corresponding couples of results showed that marine aerosols have a significant impact on atmospheric variables such as temperature, specific humidity, precipitation, and vertical velocity. Most prominent differences were revealed to frontal zones with high gradients at all vertical levels in the atmosphere. These anomalies appeared as mesoscale cells of opposite signs following each other. The radiation absorption on marine aerosols resulted in temperature rising over large areas in the upper troposphere, while opposite effects were disclosed at lower layers. The aerosol involvement led to displacement in space and time of the humidity evolution and precipitation release. Thus, the simulation of weather conditions in frontal zones over the water surface requires accounting for the effect of marine aerosols.

Key words: marine aerosols, frontal zones, mesoscale cells, Harmonie model

# **INTRODUCTION**

Atmospheric aerosols are relatively small solid or liquid particles suspended in different layers of the atmosphere and heterogeneously distributed over the Earth in space and time. Aerosols can be of natural or anthropogenic origin. Natural aerosols include sea, volcanic and dust particles (typical sizes vary from 0,001 to 100  $\mu$ m), while human activity produces aerosols mainly due to burning fuels or biomass. The obvious direct effects of aerosols associate with changes in both radiation processes and precipitation formation. The most pronounced aerosol effect on the temperature regime of the atmosphere occurs during the daylight time, when particles absorb a part of solar radiation and contribute to local warming of the atmosphere. This tends to cause a temperature inversion and stable stratification, which has an influence on convection conditions.

The presence of various types of aerosols in atmosphere causes different effects on weather and climate namely self-feedback effect, photochemistry effect, smudge-pot effect, daytime stability effect, particle effect through surface albedo, particle effect through large-scale meteorology, indirect effect, semi-direct effect, black carbon-low-cloud-positive feedback loop (*Jacobson, 2002*).

Increased interest is directed specifically to aerosol-cloud-precipitation interactions. The presence of aerosols increases cloud drop concentration and reduces the effective drop size. A high density of nuclei population initializes coalescence growth, accelerates precipitation formation, increases cloud lifetime and lags precipitation. However, the second indirect aerosol effect is still not easily detected, even the cloud satellite, weather radar and aerosol ground-based data are used (*Sporre et al., 2012*).

#### 1. SEA SALT AEROSOLS (SSA)

The open ocean is one of the major sources of natural aerosols, producing annually  $10^{15}$ - $10^{16}$  g of sea-salt aerosols. This sort of aerosols forms by evaporation of sea-spray droplets produced by bubble bursting within foamy whitecaps and wind tearing off the wave crests.

Sea salt aerosols have a significant impact on the air-sea interaction, and play an important role in atmospheric chemistry, radiation, meteorology, cloud physics, climate, oceanography, and coastal ecology. Many investigations have focused on reactions occurring inside seawater drops and their effect on the atmospheric sulfur cycle. Under certain circumstances, uptake on SSA particles may act as a sink for condensable atmospheric gases, affecting the deposition rate of nitrogen to the ocean and possibly inhibiting nucleation of new particles. Sea salt aerosol affects the transmission of electromagnetic radiation in the atmosphere by scattering light, thus affecting visibility, the performance of electro-optical systems, and remote sensing (*Lewis et al, 2004*).

A major influence of SSA is on the clear-sky radiation balance over oceans and appears through changes to system albedo, thus impacting Earth's radiation budget. Recent studies are focused on the origin of sea salts in the atmosphere and sources of condensation nuclei. Wilson had shown that without such nuclei, water vapor would not condense into cloud droplets, rain, fog, or snow at ordinary air humidity. In 1881, Aitken suggested the ocean as the primary source of condensation nuclei. By analogy to laboratory observations of collapsing bubbles, Jacobs first assumed that in the ocean the breaking of waves and bursting of bubbles produce sea-salt aerosols. Boyce found experimentally that most of these particles are produced within foamy whitecaps formed when waves break. Fast-speed photography documented the exact mechanism of bubble bursting and drop formation (*Anguelova*, 2002).

Sea spray comprises three types of spray droplets, named after their production mechanisms: film, jet, and spume drops (Wu, 1992a; Andreas et al., 1995). As wind blows over the ocean, waves break and entrain air into the water forming clouds of bubbles beneath and foam patches on the surface. Once formed, the bubbles start rising to the surface due to their buoyancy. The bubbles reaching the open surface are responsible for the formation of sea spray. While they float on the surface, the film caps, separating the air inside the bubbles from the air outside. thin and the bubbles burst producing drops by two mechanisms (Resch and Afeti, 1991; Spiel, 1995). In the first mechanism, the shattering of the film caps generates 10s to 100s of small droplets called film drops. Film drops cover a size range from a physically possible minimum of 0.1 µm radius to a typical value of 50 µm with a peak about 1-2.5 µm. In the second mechanism, a water jet projects with high acceleration upward from the collapsing bubble cavity, becomes unstable, and breaks up into several droplets called jet drops. Depending on the bubble size, 1 to 10 jet drops are ejected into the air from the tip of the jet. Jet drop radii range from 1 to 300 µm, with a peak around 10 µm. These two ways of producing droplets are indirect, i.e., mediated by the bubbles within the whitecaps. A third, direct, mechanism for spray generation is via mechanical disruption of wave crests under strong winds. At high wind speeds, above 9 m/s, spume drops are torn from the wave crests and blown directly into the air. Spume drops are relatively large, with radii starting from 20 µm and reaching more than 500 µm (Andreas, 2002). Altogether, sea spray droplets cover a size range of 0.1-500 µm radii.

Driven out of the sea and entering the atmosphere, sea spray droplets get into an environment different from their watery milieu. Normally, the air is drier, with relative humidity (RH) less than 100%, and film, jet, and spume droplets start to evaporate. They eventually equilibrate with the ambient RH, and, becoming smaller, stable and long-lived, develop into sea-salt aerosols. Not all sea spray droplets transform into sea-salt aerosols. Many drops return to the sea almost immediately; the largest, > 100  $\mu$ m, stay less than 0.5 s in the air (*Andreas, 1992*). Under winds of 5 m/s, 20-100  $\mu$ m drops linger for at most 10 s, and settle down before reaching moisture equilibrium. Sea spray with radii < 20  $\mu$ m reside in the air for more than 10 s, and succeed in reaching moisture equilibrium. With that the process of formation of sea-salt aerosols ends.

While wind speed is certainly the most important factor in determining both the ocean surface production and the fraction of particles reaching the reference height, other aspects of the atmosphere and ocean may also be important, like water and air temperature, salinity, the presence of ice.

### 2. MODELING

As in the sphere of meteorology, numerical modelling is today one of the most important tools for research and applications in the field of atmospheric physical-chemistry.

In the future, the increasing computer power and understanding of physical processes pave the way for developing integrated models of the Earth system and gives a possibility to include interactions between atmosphere, environment, climate, ocean, cryosphere and ecosystems. Therefore, development of integrated Numerical Weather Prediction (NWP) and Atmospheric Chemical Transport (ACT) models is an important step in this strategic direction and it is a promising way for future atmospheric simulation systems leading to a new generation of models.

In the previous decades air pollution forecasting and numerical weather prediction (NWP) were developed separately, since the resolution of NWP models was too poor for meso-scale air pollution forecasting.

Due to modern NWP models approaching meso- and city-scale resolution and the use of land-use databases and remote sensing data with finer resolution, this situation is changing. As a result the conventional concepts of meso- and urban-scale air pollution forecasting need revision along the lines of integration of meso-scale meteorological models (MetMs) and atmospheric chemical transport models (ACTMs) (*Grell et al. 2005; Jacobson, 2006; Korsholm and Baklanov, 2007*). This combination is reasonable due to the facts that: (1) meteorology is an important source of uncertainty in air pollution and emergency preparedness modelling, (2) there are complex and combined effects of meteorological and pollution components on human health, (3) pollutants have effects, especially aerosols, on climate forcing and meteorological phenomena (radiation, clouds, precipitation, thunderstorms, etc.). So, this way of integrating modeling can be beneficial for model improvements in both communities: NWP and atmospheric environment forecasting. Within this line the HARMONIE model is one of those serving as a high resolution meteorological driver for ACT models.

#### **2.1. HARMONIE MODEL**

The non-hydrostatic convection-permitting HARMONIE model is developed in cooperation with Météo-France and ALADIN consortium. At the default horizontal resolutions is less than 2.5 km, the forecast model and analysis system are basically those of the AROME model from Météo-France (*Seity et al,* 2011, Brousseau et al, 2011). At coarser resolutions the ALARO or ECMWF physics parameterisations can be used, and/or the hydrostatic dynamics of ALADIN. The dynamical core is based on a two-time level semi-implicit Semi-Lagrangian discretisation of the fully elastic equations, using a hybrid coordinate in the vertical (*Seity et al, 2011*). Optionally, for larger domains and coarser resolutions the hydrostatic version of this semi-Lagrangian scheme can be used. An Eulerian dynamics core is available, but has been little used in recent years. In practice normally a rotated lat-lon grid projection is adopted by HARMONIE for the output grid.

A variety of sub-grid scale physical processes are taken into account by parametrization schemes. Basically, the parametrization adopted in HARMONIE are the same as those of the AROME model (*Seity et al., 2011*), developed by the meso-NH community.

The externalized surface scheme SURFEX is a set of models used for the description of the different types of surfaces: soil, sea and inland water bodies, and urban environments. It assumes a tile approach, distinguishing different surface types. Surface physiographies are prescribed using the 1 km resolution ECOCLIMAP database (ref.). SURFEX consists of a number of components, which are all described in the SURFEX scientific documentation.

The default upper air data assimilation scheme in HARMONIE is the 3DVAR scheme developed (*Brousseau et al, 2011*). Background error statistics have been calculated using the NMC method. An analytical balance condition is applied. The observation data types assimilated by default presently are conventional observations (TEMP, SYNOP, AIREP, PILOT, SATOB, SHIP, DRIBU) and AMSU-A / ATOVS radiances over sea. Additionally, it is possible to assimilate AMSU-A over land and sea ice, AMSU-B, geostationary and MODIS atmospheric motion vectors, SEVIRI cloud-cleared radiances, GPS zenith total delay, wind profilers, radar radial winds and profiles and Sea winds scatterometer data.

Variational bias correction is applied by default to all satellite data. Observation screening involves logical and representivity checks, background quality checks, black-or-white listing, multi-level and station level checks, redundancy checks and moving platform checks. Thinning is used to reduce the amount of observations. Analysis of surface variables is done within the spatial interpolation tool CANARI, applying optimum interpolation for the assimilation of screen level parameters T2m and RH2m, and sea surface temperature. This is to be extended with snow depth.

To reduce noise and spin-up, analyses can be initialized out by incremental digital filter initialization (DFI) (*Lynch et al. 1997*). In HARMONIE, initial and boundary conditions are normally taken from the larger scale HIRLAM or IFS (ECMWF) model. Lateral boundaries are overspecified, all variables being externally prescribed by the nesting model. Normally a relaxation zone of 10 grid points is adopted. Boundary relaxation is performed after the horizontal diffusion. At the upper boundary a condition of zero vertical velocity is imposed.

#### **3. EXPERIMENT SETUP**

To evaluate the sea salt aerosols impact on weather conditions, two types of numerical experiments with the Harmonie model are performed. The physics package includes six aerosol types: sea salt, land, soot/urban, desert dust, stratospheric sulphate background and volcano. The four earlier types must be initialized in one set if only aerosol simulations are performed, while the latter two aerosols are optional. At the first experiment, the aerosol module is switched off, i.e., aerosols are not taken into account (the reference experiment). At the second (modified) experiment, only the sea salt aerosol is included, while the rest are set up to zero values. The comparison of model output from two kinds of experiments allows to distinguish the contribution from the sea salt aerosol to atmospheric physics.

The model domain covers the North Atlantic region by 450 x 360 grid points with horizontal resolution of 25 km for keeping consistency with the AQMII project. The forecasts are integrated for 120 hours from 11 to 16 August 2010. Initial and boundary conditions are supplied by the ECMWF-IFS global model with time interval of 3 hours. For more detailed consideration of the effect of aerosols, two vertical cross-sections are located along the atmospheric frontal zone (fig.1).



Fig. 1. The weather chart of the 500 hPa geopotential height and surface pressure on 16 August 2010, 06 UTC with geographical location of the vertical cross-sections

#### 4. RESULTS

The role of marine aerosols in atmospheric processes was investigated by comparison of corresponded meteorological fields. The following three-dimensional analysis (in terms of vertical cross-sections and space distributions on standard pressure levels) is based on the differences between the reference and modified model runs.

In general, majority of prognostic meteorological variables considerably deviates from its reference analogues at the sea aerosol presence. These deviations develop in a form of small scale circulations with opposite signs geographically located each next to the other. Despite the fact that sea salt aerosol concentration is homogeneously distributed over the domain, the higher contrasts appear in differences for the intermittent mesoscale cells along frontal zones and within humid air masses. The focus of the study is on weather characteristics such as the air temperature, specific humidity, precipitation, vertical velocity and radiation fluxes.

The sea salt aerosol influence on the instantaneous short-wave (SW) radiation fluxes on the top of atmosphere as well as near the surface appears very similar in both numerical experiments. On particular, fields of the differences show large areas covered by small scale cells of opposite signs with average values of about  $\pm 20 \text{ W/m}^2$ . The instantaneous radiation fluxes, both SW and long-wave (LW), were calculated on the basis of its accumulated values during the forecast length. It should be noted, the increasing or decreasing of SW radiation within a particulate cell on the atmosphere top corresponds to the same changing near the surface. The maximum value of SW radiation provoked by aerosol particles reaches up to 114,5 W/m<sup>2</sup> at the top of the atmosphere and 143 W/m<sup>2</sup> near the surface (fig.2a). The relative growing of SW radiation fluxes in "clear" atmosphere in comparison to polluted conditions reaches up to 80,5 W/m<sup>2</sup> and 95,3 W/m<sup>2</sup> at the top of the atmosphere and near the surface correspondingly.

LW radiation shows weaker sensitivity to aerosol at a clear sky area, however this becomes ambiguous at front zone and cloudy area (fig. 2b). The domain averaged aerosol effect on both LW and SW radiation fluxes is negligible. The mean LW radiation differences on the top of the atmosphere and near the surface are as much as  $-0,078 \text{ W/m}^2$  and  $-0,083 \text{ W/m}^2$  correspondingly, whereas for the SW radiation they are of about  $0,12 \text{ W/m}^2$  and  $0,17 \text{ W/m}^2$ . The extreme values of differences in LW fluxes varies from -32,5 to  $31,3 \text{ W/m}^2$  near the surface and from -28,2 to  $29,2 \text{ W/m}^2$  on the top of the atmosphere.



Fig. 2. Differences (no aerosols – sea aerosols) of short-wave radiation fluxes near the surface (a) and long-wave radiation fluxes at the top of the atmosphere (b) on 16 August 2010, 06 UTC

The differences in the temperature fields between two experiments after the 5-day model integration are as large as  $\pm 3 \div 5$  K and reach extreme values up to  $\pm 10$  K at single cells, which are related to particular geographical regions, specific patterns of the atmospheric flow and mainly appear within the planetary boundary layer (PBL) (fig. 3a).

The results for the low/medium/high cloud cover and evolution of precipitation are in agreement with a well-known fact that aerosol particles act as additional condensation nuclei, changing the drop size distribution for the same water amount, extending a cloud lifetime with delaying a rainfall moment and affecting a precipitation rate. The differences in the specific humidity fields amount to  $\pm 10$  g/kg (figs. 3b, 4), in rain  $\pm 2.5$  g/m<sup>2</sup> (fig. 3c) and in vertical velocity up to  $\pm 1$  m/s (fig. 3d). The vertical cross-sections clearly show the slopped multi-centered deviations (fig. 5). They are associated with the first direct effect of aerosols; however non-linear dynamical processes developing on atmospheric fronts should be taken into account as well. The spatial distribution of differences in the humidity, precipitation and vertical velocity fields sufficiently depends on the geographical location and atmospheric flow regime. Worth to note that the evident aerosol impact realizes through meso-scale cells, while domain averaged values are weakly sensitive to aerosols on short time scales.

Largest differences in the temperature and specific humidity fields are observed within the layer between 500 and 1500 m with a maximum near the top of PBL (fig. 4). In particular, temperature at that layer is higher in the presence of aerosols due to absorbing the incoming solar radiation, while it causes the cooling of layers below. In the experiments with no aerosols, temperature is higher from the low to middle troposphere. This promotes the development of stronger and more active cells in the convective area behind the front (strong updrafts) along with earlier and heavier rainfall, correspondingly.

The complexity of non-linear interactions among aerosol particles and atmospheric physical characteristics can be explained by a chain of inter-related factors. For example, the presence of aerosols increases absorption of solar radiation during daytime. Variations in a SW radiative flux lead to changes in temperature, both in values and vertical profiles. This alters the moisture capacity and subsequent thermal inertia of the air. Higher amount of water vapor and aerosols in the atmosphere enhance the greenhouse effect at nighttime. On the other hand, variations in a temperature vertical profile make stronger or weaker updrafts and compensatory movements eventually modifying convection conditions and precipitation formation.









*Fig. 3. Differences (no aerosols – sea aerosols) in temperature (a), specific humidity (b), rain (c) and vertical velocity (d) at 925 hPa level on 16 August 2010, 06 UTC* 



*Fig. 4. Vertical cross-sections of temperature differences (no aerosols – sea aerosols) along I (a) and II (b) lines (see fig. 1) on 16 August 2010, 06 UTC* 



Fig. 5. Vertical cross-sections of specific humidity differences (no aerosols – sea aerosols) along I (a) and II (b) lines (see fig. 1) on 16 August 2010, 06 UTC

Thus, the simulation of weather conditions in frontal zones over the ocean surface is especially sensitive to the presence of sea salt aerosols and requires additional investigations for the better understanding of physical nature and model representation.

# CONCLUSIONS

Numerical experiments with the HARMONIE model have shown that the sea salt aerosols have significant influence on atmospheric variables such as the temperature, humidity, precipitation and radiation, despite the fact that their concentration is smaller in comparison with other types of aerosols. The most prominent differences associate with frontal zones of high gradients, where interaction between cold and warm air masses occurs. The anomalies appear in a form of intermittent mesoscale deviations with opposite signs following each other, which maximum amplitudes revealed in the middle troposphere. The treatment of the integral effect of sea salt aerosols requires additional attention and awareness, since extreme values of calculated differences considerably exceed its mean values averaged over the entire domain.

The presence of sea aerosols leads to increasing temperature in the lower and middle troposphere, intensification of development of convective cells along the front and provokes non-uniform changes of precipitation release.

# ACKNOWLEDGEMENT

The authors are very grateful to EuMetChem COST Action for financial support; ECMWF and DMI for access to High Performance Computing Facilities; Prof Alexander Baklanov for leading collaboration with OSENU group in Harmonie research modelling

#### REFERENCES

Andreas, E. L., 1992. Sea spray and the turbulent air-sea heat fluxes, J. Geophys. Res., 97, 11429–11441.

Andreas, E. L., J. B. Edson, E. C. Monahan, M. P. Rouault, and S. D. Smith, 1995. The spray contribution to the net evaporation from the sea: a review of recent progress, Boundary-Layer Meteor., 72, 3-52.

Anguelova, M. D, 2002. Whitecaps, sea-salt aerosols, and climate, Ph.D. Dissertation, University of Delaware.

Baklanov, A., Mahura, A., Sokhi, R. (Eds), 2010. Integrated Systems of Meso-Meteorological and Chemical Transport Models, Springer, ISBN:978-3-642-13979-6, p. 242.

Brousseau, P., Berre, L., Bouttier, F., Desroziers, G., 2011. Background-error covariances for a convective-scale data-assimilation system: AROME-France 3D-Var. *Quarterly Journal of the Royal Meteorological Society* **137**:10.1002/qj.v137.655, 409-422.

Grell, G., Peckham, S., Schmitz, R., McKeen. S., Frost, G., Skamarock, W., Eder, B., 2005. Fully coupled "online" chemistry within the WRF model, Atmos Environ 39(37):6957–6975.

Jacobson, M., 2002. Atmospheric pollution: history, science and regulation. Cambridge University Press, New York.

Jacobson, M., 2006. Comment on "Fully coupled 'online' chemistry within the WRF model," by Grell et al., Atmos Environ 39:6957–697.

Korsholm, U., Baklanov, A., Gross, A., Sørensen, J., 2007. Influence of offline coupling interval on meso-scale representations, Atmos Environ 43:4805–4810.

Lewis, E. R. and Schwartz, S. E., 2004. Sea Salt Aerosol Production Mechanisms, Methods, Measurements, and Models – A Critical Review, Geophysical Monograph Series Vol. 152, (American Geophysical Union, Washington).

Lynch, P., Giard, D. and Ivanovici, V., 1997. Improving the efficiency of a digital filtering scheme, Mon. Wea. Rev., 125, 1976-1982.

Resch, F., and G. Afeti, 1991. Film drop distributions from bubbles bursting in seawater, J. Geophys. Res., 96, 10681-10688.

Seity, Y., Brousseau, P., Malardel, S., Hello, G., P. Bénard, F. Bouttier, C. Lac, and V. Masson, 2011. The AROME-France Convective-Scale Operational Model. *Mon. Wea. Rev.*, **139**, 976–991. doi: <u>http://dx.doi.org/10.1175/2010MWR3425.1</u>

Spiel, D. E., 1995. On the births of jet drops from bubbles bursting on water surfaces, J. Geophys. Res., 100(C3), 4995-5006.

Sporre, M. K., Swietlicki, E., Glantz, P., Kulmala, M., 2012. A study of how aerosols affect low-level clouds over the Nordic Countries using MODIS, ground-based, ECMWF and weather radar data. Geophysical Research Abstracts. 14, EGU2012-7983.