Tracing the aerosol nucleation events with atmospheric transport model SILAM

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Introduction

This paper is intended to clarify the geographical extent of processes leading to a nucleation event and the role of atmospheric transport in it. The study is based on the inverse (adjoint) runs of atmospheric advection-diffusion model SILAM and general knowledge on basic mechanisms and time scales of nanometer-sized particle formation in the atmosphere. Results of two aerosol measurement campaigns were used as sensitivity source data for backward tracing:

- • in Värriö, Finland, Eastern Lapland, April – May 2003;
- • in Preila, Lithuania, Kura isthmus, May – August 2006.

Methods

The field experiments were carried out (Figure 1):

- from April 28 to May 11, 2003 in SMEAR I site (Station for Measuring Forest Ecosystem – Atmosphere Relation, 67°46'N, 29°35'E), located in Värriö nature park in eastern Lapland, ner the border of Russia, 100-200 km far from major pollution sources at Kola Peninsula.
- From May 25 to August 10, 2006 in Preila, Lithuania (55.55 N, 21.00 E), at the Environmental Pollution Research Station located on the eastern coast of the Baltic Sea. The measurement site was on a sandy beach at a 50 m distance from the seashore.

Campaigns included measurements of aerosol particle size distributions with EAS (electric aerosol spectrometer, Tammet et al., 2002. The EAS was used to measure the aerosol size distribution in the aerodynamic diameter range of 3 nm-10 μm. This study is focused on the nucleation (3-24 nm) and the Aitken mode (24-100 nm) particles.

The SILAM model applied in this study has been developed at the Finnish Meteorological Institute (Sofiev et al., 2006a). For aerosol, the SILAM considers advective and turbulent transport, and size-dependent dry (including sedimentation) and wet deposition.

The SILAM model has two modes of operation: forward and adjoint (Sofiev et al., 2006b).

- In forward mode, the input data contains the emissions from specified sources, meteorological fields produced by numerical weather prediction models, and land use. The output of the forward simulations consists of the 3D spatial concentration patterns developing in time and 2D dry and wet deposition fields.

- In adjoint mode, the model input in the specific case of this study contains measured aerosol concentrations (the so-called sensitivity source function or footprint of the observations), and the meteorological fields produced by the ECMWF numerical weather prediction model. The output (the so-called sensitivity distribution) is a 4-D probability field for the sources of observed concentrations. This output specifies the probability that the measured concentration is originated from a specific location or region.

Forward model runs were also performed in order to investigate the contribution of anthropogenic and natural emissions of sulphur, primary particulate matter, and sea salt to the aerosol detected. Anthropogenic emission data is based on the EMEP database.

Figure 1.

Locations of research stations:

Värriö, Eastern Lapland, Finland, 67°46'N, 29°35'E

Preila, Kura isthmus, Lithuania, 55.55 N, 21.00 E

Base map: from Google Earth

Results: Värriö 2003

The measured mass concentrations of the nucleation mode and Aitken nuclei are presented in Figure 2. The nucleation-mode peaks occurred on 30th of April (event 1), and on 5th and 9th of May (events 2 and 3, respectively). The highest peak of Aitken nuclei (also, coarser particles) was observed at April 30, just after event 1.

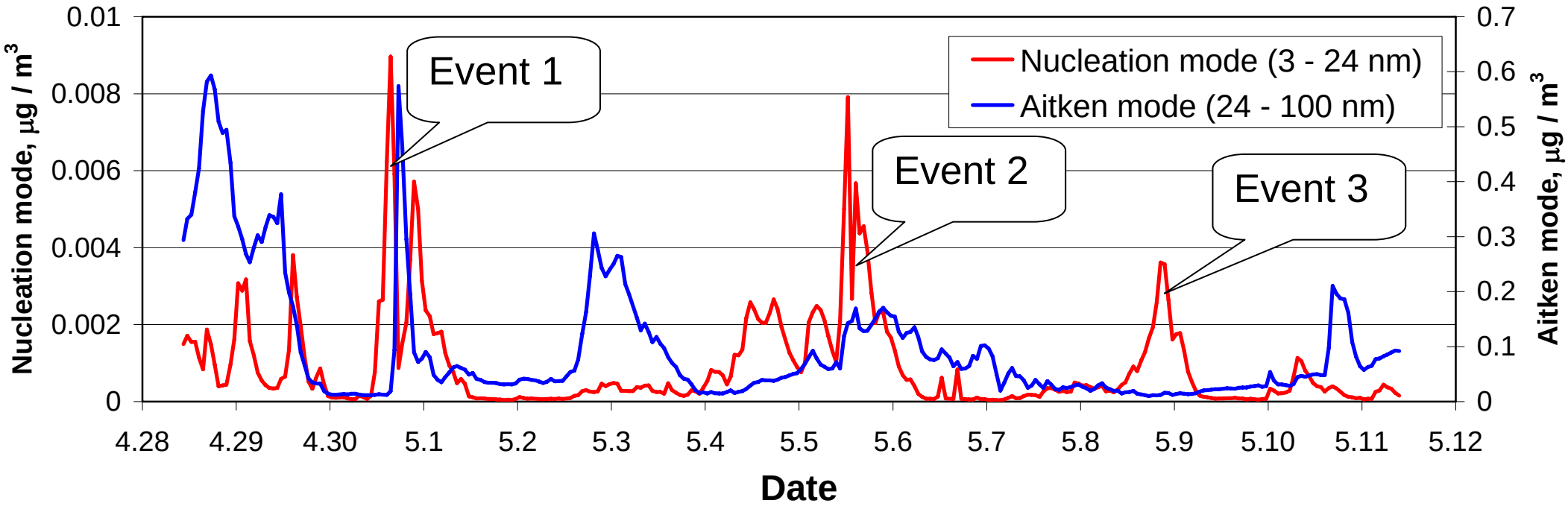


Figure 2. *Hourly average mass concentrations of aerosol in nucleation and Aitken modes measured by the EAS during the Värriö campaign in April-May 2003.*

Värriö, event 1

The first nucleation event is interesting due to the dramatic change of air masses at Värriö that interrupted its observation (Figure 3). It is rather complicated, on the basis of the performed SILAM run, to highlight the origin of its precursors than in the case of first event. We can expect either emissions from the Arctic Ocean or biogenic emissions from coniferous forests of Lapland during a few hours of transport over land areas before reaching the measurement site. Although the area was still covered with snow and the temperatures were slightly below zero, solar heating of tree crowns might induce some vegetation activity. As the air masses advected very lose to the Nikel metallurgy factory (Russia), we cannot exclude some triggering influence of gaseous emissions from there.

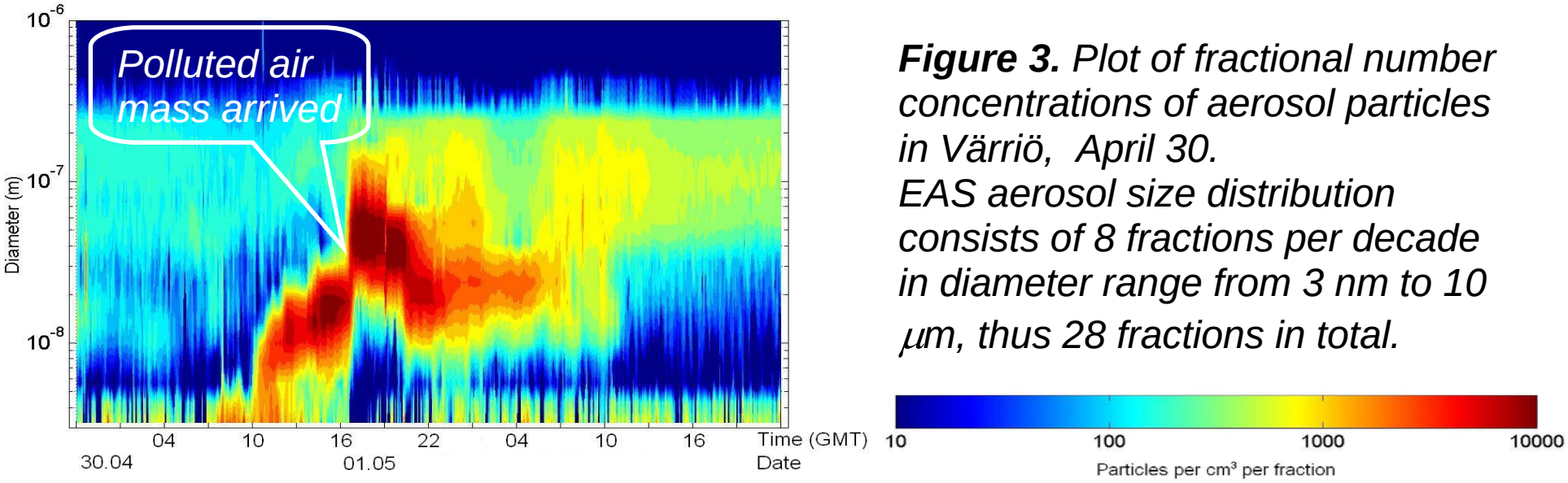


Figure 3. *Plot of fractional number concentrations of aerosol particles in Värriö, April 30. EAS aerosol size distribution consists of 8 fractions per decade in diameter range from 3 nm to 10 μm, thus 28 fractions in total.*

Värriö, event 2

The second nucleation event started on 5 of May at 11 a.m. GMT, when a large number of nanometer particles grew to Aitken sizes in late evening. Inverse computations show that the air masses spent two previous days over the continental areas of northern Sweden and central Finland (most of time over boreal forest) and were transported over the Botnian Bay (Figure 4). During and shortly after this nucleation event the forward model computations did not show any advection of substantial concentrations of sulphate over Värriö. Sunny and relatively warm weather during these days conditioned accumulation of biogenic aerosol precursors in the air mass. Event formed in the morning of May 5 over Southern Lapland.

Looking at fractional aerosol concentrations during the nucleation event 2, it is evident that uniform condensation growth of particles is disturbed. First, continuous growth of particles is seen in size range of 3-10 nm, but then a gap appears (Figure 5). Such a structure cannot be explained by condensation growth only. A likely explanation is an advective effect: the gap in time series may appear due to air mass that had no significant nucleation due to some reason.

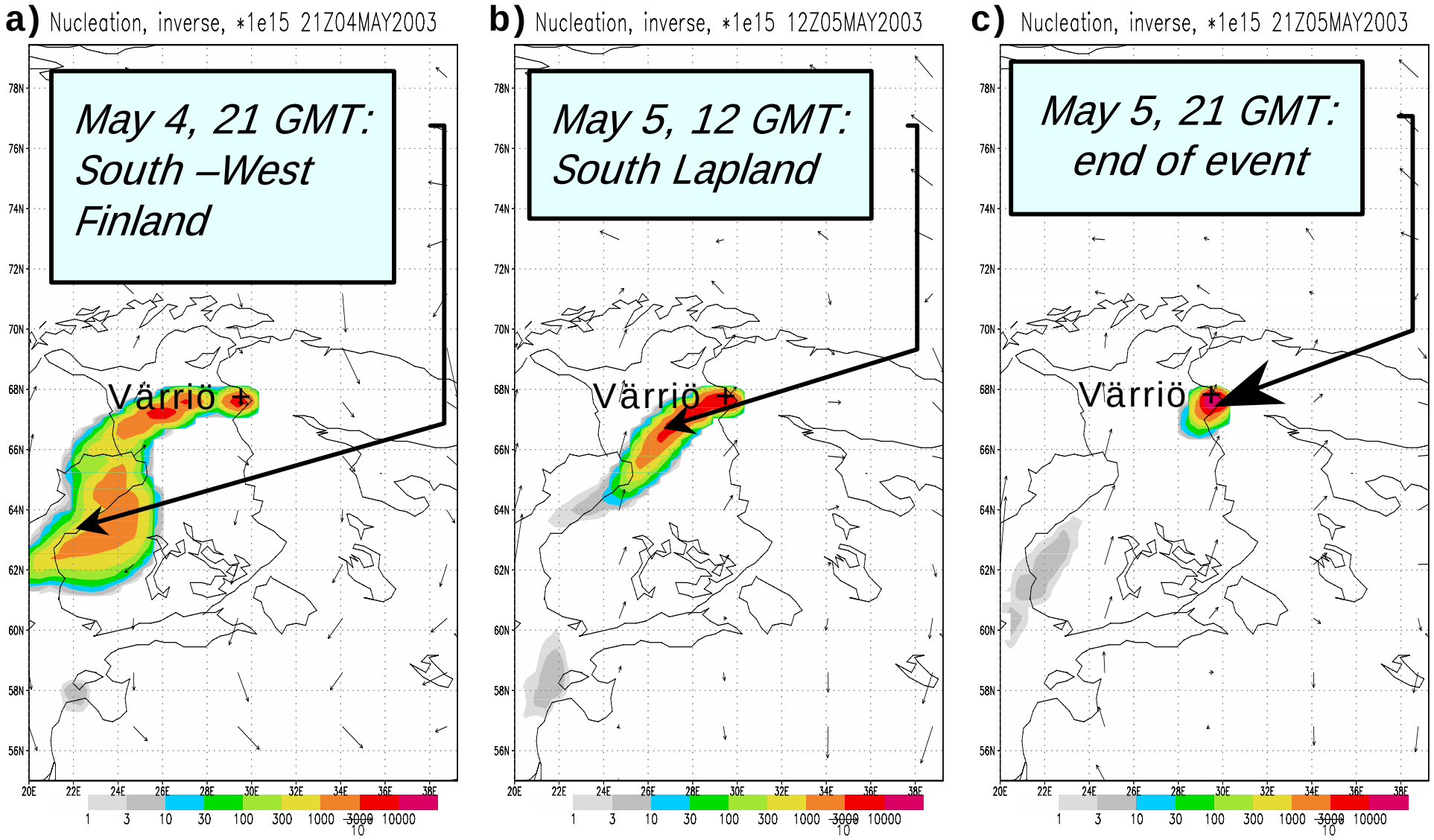
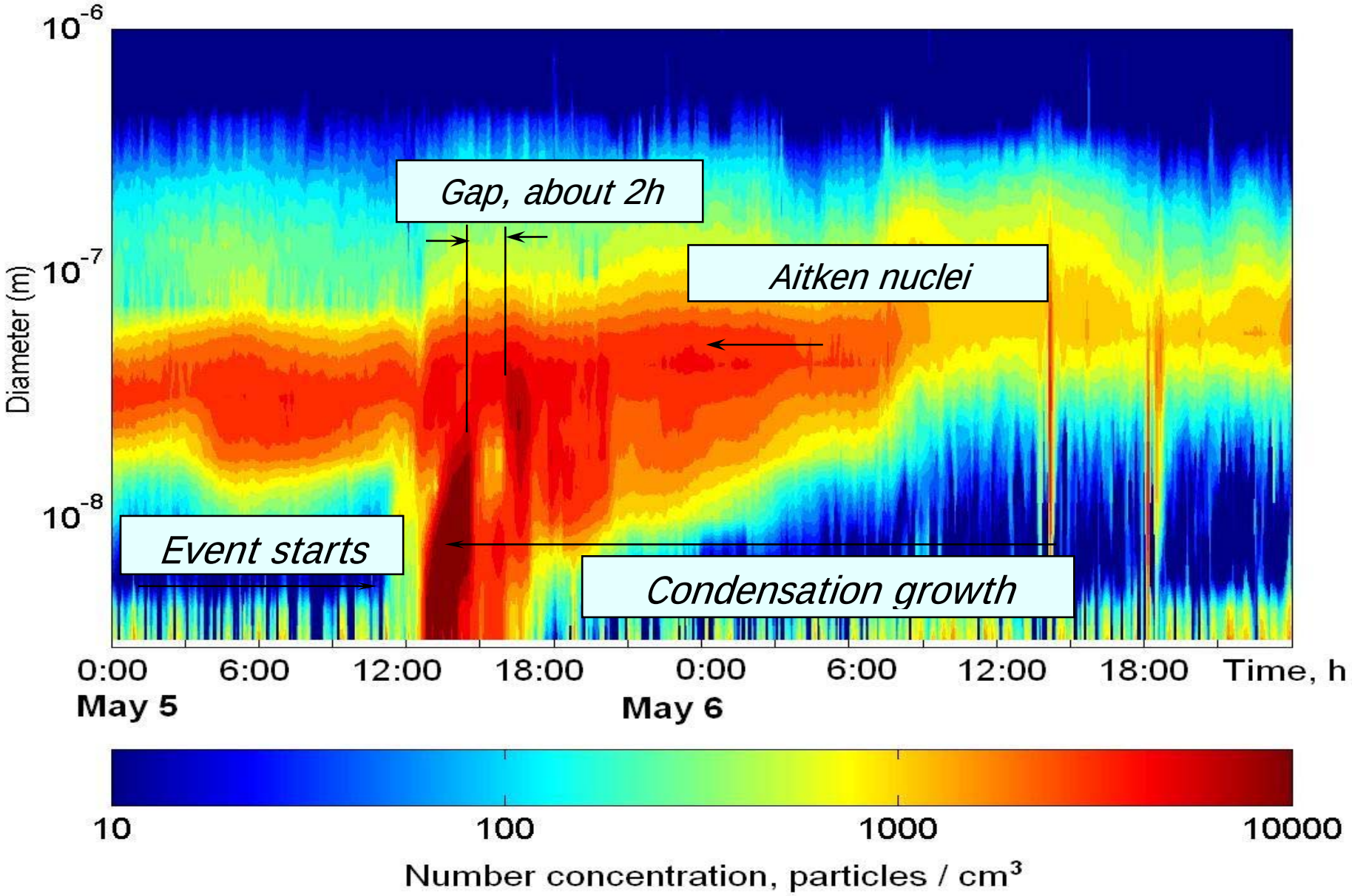


Figure 4. *Evolution of the footprint of nucleation mode particle mass concentration*, nuclea



tion event 2

Figure 5. *Evolution of particle size distribution during event 2*

<u>Värriö, event 3</u>In the case of nucleation event on 8 of May, the air masses were transported from the Norwegian Sea. A well-defined footprint was formed that extended over the Finnish and Norwegian Lapland from north-west to south-east (Figure 6). Thus, the leading role of marine emissions is expected.

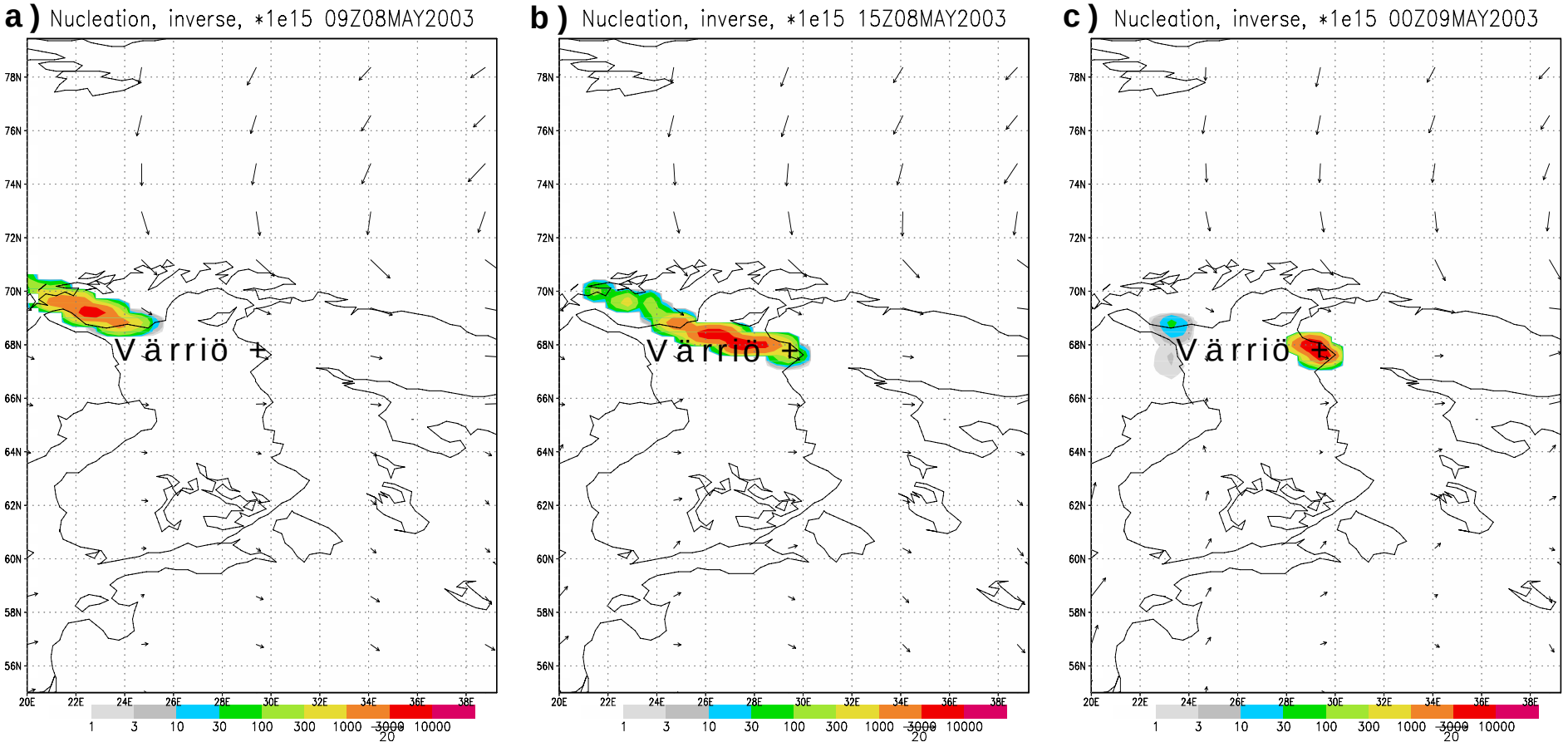


Figure 6. *Evolution of the footprint of event 3, 9:00 May 8 to 0:00 May 9 GMT.*

Results: Preila

Preila, triple event, June 15-17

During June 15-17 nucleation events were observed during three sequential days (Figure 7). Backward tracing with SILAM shows that the air masses had different origin. Event at June 15 formed in an continental air mass that spent entire 3 days in a steady high-pressure system located with the center near the measurement station. Air mass during following two events originated from north, as it is typical for days with a nucleation event (Figure 8).

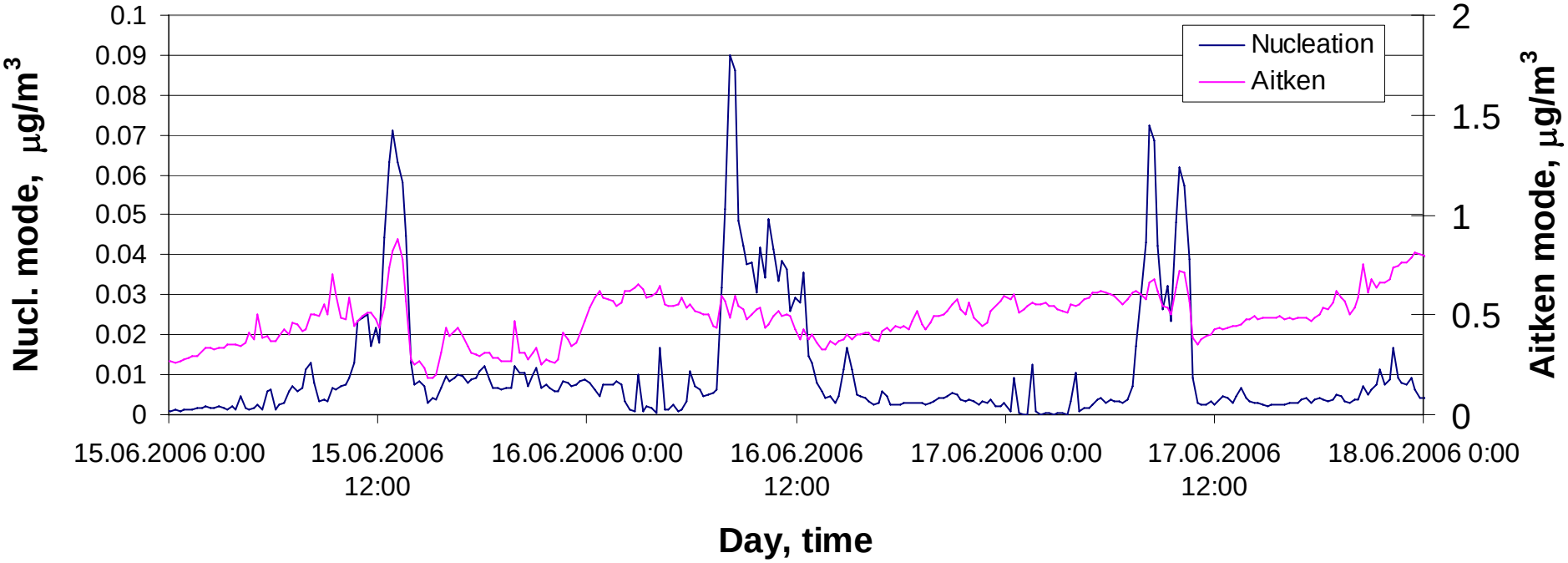


Figure 7. *Hourly average mass concentrations of aerosol in nucleation and Aitken modes measured by the EAS during June 15-17, 2006 in Preila.*

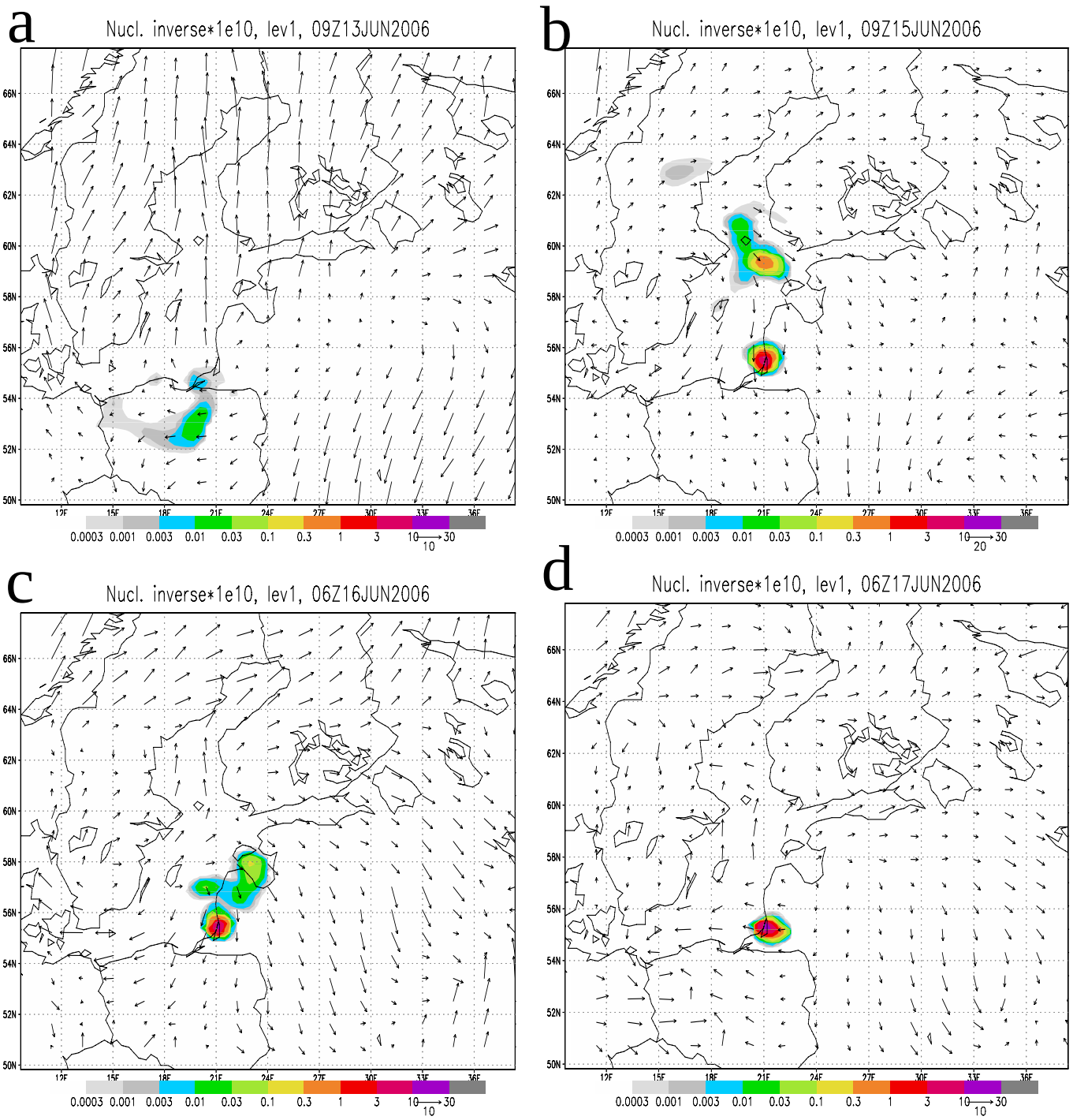


Figure 8. *Evolution of the footprint of the triple nucleation event:*

a) footprint of first event, 9:00 GMT at June 13 (2 days before event);

b) first event is occurring in Preila, 9:00 GMT at June 15, joint footprint of second and third events is seen in north;

c) second event is occurring in Preila, 6:00 GMT at June 16, footprint of third event over Gulf of Riga;

d) *third event is occurring in Preila, 6:00 GMT at June 17.*

Preila, double event at July 28-29

These two nucleation events waere rather typical: formed during an outbreak of clean air mass from the north. The results of adjoint SILAM run are given in Figure 9.

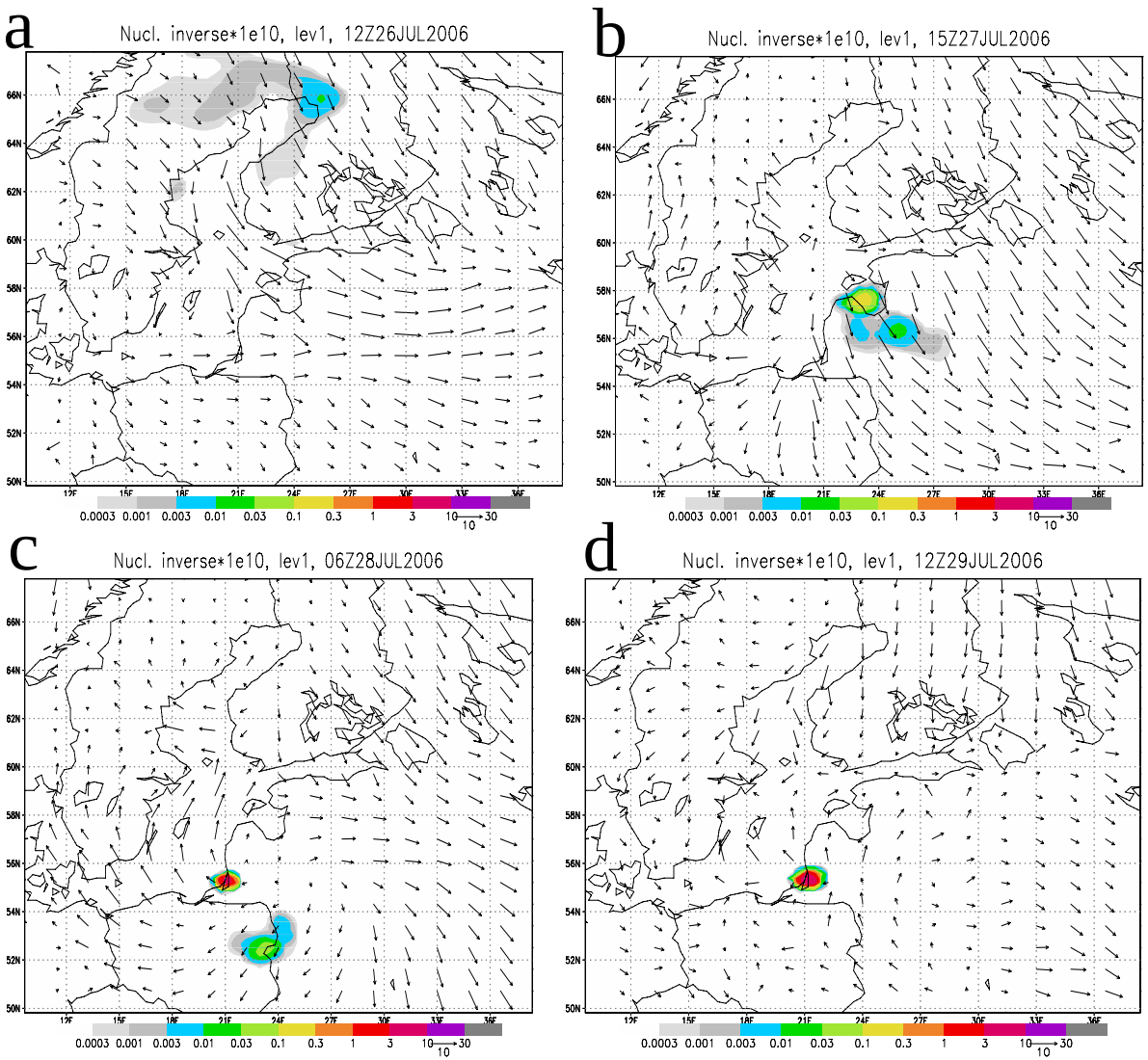


Figure 9. *Evolution of the footprint of the double nucleation event:*

a) footprint at 12:00 GMT at Jult 26);

b) footprint at 15:00 GMT at July 27);

c) first event is occurring in Preila, 6:00 GMT at July 28, footprint of second event over Eastern Poland;

d) *second event is occurring in Preila, 12:00 GMT at July 29.*

Conclusions

The footprint areas of seven observed nucleation events suggest that spatial scale of a nucleation event may reach about 1000 km and impact of atmospheric *transport to the aerosol processes recorded by an Eulerian (ground-based) observer may be significant. The nucleation events tend to be related with high pressure areas and outbreaks of air masses from the north. Formation of an intense event over extensive forested areas supports the theory on the role of biogenic VOC emissions. Need for coupling the models of atmospheric transport and aerosol dynamics is evident.*

References

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