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On the use of satellites to obtain information on the occurrence of natural and anthropogenic aerosols over the boreal eurasian forest

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Abstract

The ALANIS-Aerosols project is a feasibility study on the use of existing satellite data for discriminating between natural aerosols emitted by boreal Eurasian forests and long-range transported anthropogenic aerosols. In this paper an overview is provided of different satellite products which are potentially useful to obtain this kind of information. Approaches that have been followed in the past are briefly summarized. Secondary production of aerosols from their precursor gases, in particular biogenic volatile organic compounds which play an important role in the formation of new particles through nucleation, is briefly discussed. These newly formed particles are initially too small to observe directly with optical instruments used for earth observation (EO) but through the use of proxies information can be obtained on global nucleation mode aerosol concentrations. Next we focus on aerosols with particle sizes in the optically active size range, roughly particles with a diameter larger than 50–100 nm, depending on wavelength. These particles can be observed with EO instruments as well as using several different types of in situ observations. The satellite data used are aerosol properties retrieved from the Advanced Along Track Scanning Radiometer (AATSR, flying on ENVISAT since 2002) using both the forward and nadir view with the AATSR dual view algorithm (ADV). Ground-based in situ data used here are aerosol properties measured in Hyytiälä, Finland. These observations are complemented with model calculations using the global atmospheric aerosol and chemistry model GLOMAP. Examples show the complementarity of different data sources to obtain information on the temporal and spatial information on the nature of aerosols over the Boreal forest.

1 Introduction

Aerosols in the atmosphere are composed of particles of both natural (soil dust, sea salt, sulphates and natural primary and secondary organic aerosol) and anthropogenic (e.g., particles produced from energy production, traffic, industrial and other human activities such as households and agriculture) origin. However, since the lifetime of

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aerosol particles is comparable to the time scale of intra- and inter-continental transport (i.e., 3 to 10 days), aerosol particles of anthropogenic origin are ubiquitous and the natural "background" aerosols are difficult to observe and quantify with confidence (Andreae, 2007).

Anthropogenic aerosols represent an "external" cause of climate change with respect to natural aerosols. Natural aerosols, in turn, may provoke variations in the climate system as part of a feedback mechanism in which climate change may induce changes in emissions of natural aerosol particles and their precursors. To further complicate matters, anthropogenic emissions interact with naturally occurring aerosols (or aerosol precursors) and it is difficult to discriminate natural from anthropogenic particles. Yet, the separation of anthropogenic from natural aerosols is essential for assessing human impacts. In particular, for properly assessing the effect of anthropogenic aerosols on atmospheric processes and climate, it is mandatory to effectively characterize the burden of natural aerosols as a reference level for inferring human-induced aerosol impact. This separation is also important for understanding the cloud-mediated effects of aerosols on climate, since cloud properties respond to aerosols in a nonlinear way and are most sensitive to the addition of particles when the background concentration is low (Andreae, 2007; Andreae and Rosenfeld, 2008).

In recent years there has been a growing interest in the influence of aerosols on climate. Aerosols influence climate directly through scattering and absorption of solar radiation and indirectly through their effect on cloud properties. Several extensive investigations and coordinated field campaigns have been carried out to assess the impact of anthropogenic aerosols from in situ measurements (Griffits et al., 1999; Raes et al., 2000; Ramanathan et al., 2001; Huebert et al., 2003; Kulmala et al., 2009). However, due to the intrinsic complexity of the problem, very few studies have been conducted using remote sensing data (Liu et al., 2005). Satellite remote sensing instruments do not directly measure the aerosol chemical composition needed to discriminate anthropogenic from natural aerosol components but can be used to infer information as described below.

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The general aim of the ALANIS Theme 3 – Aerosols (in short ALANIS – Aerosols) project is to investigate the feasibility of the use of existing satellite data, either alone or in combination with ground-based in situ or remote sensing data and/or with chemical transport models, for discriminating between natural aerosols and long-range transported anthropogenic aerosols over the boreal Eurasian forest. In this context, the interest in investigating anthropogenic in contrast to natural aerosol dynamics over boreal Eurasia stems from two main concepts. First, boreal forests directly modify the atmospheric aerosol content. In particular, ubiquitous features at boreal Eurasian forest sites are the regular occurrence of new particle formation events through the generation of secondary organic aerosols (SOA) and their subsequent growth by condensation of biogenic organic compounds that are emitted from the forests (Kulmala et al., 2001, 2004, 2008; Tunved et al., 2006, 2008; Spracklen et al., 2006). Second, due to particular wind circulation patterns the clean atmosphere over northern Eurasia is impacted periodically by long-range transported anthropogenic aerosols, such as those formed by energy production, traffic, industrial and other human activities such as households and agriculture (Arola et al., 2007; Stohl et al., 2007; Shindell et al., 2008).

Atmospheric reaction products of biogenic volatile organic compounds (BVOC) emitted from vegetation, especially boreal forest, are the main source of SOA (Kulmala et al., 2001; Hallquist et al., 2009). The geographic and seasonal distribution of BVOC is highly uncertain. The direct contribution of anthropogenic emissions to SOA is quite small but anthropogenic aerosol may serve as nuclei for biogenic SOA formation and growth (Kanakidou et al., 2000). Global total monoterpene or isoprene emissions are dominated by tropical ecosystems and boreal forests contribute only around 5–10 % by mass. However, emission maxima during summer, especially for monoterpenes, can be as large as in tropical regions. Monoterpenes dominate the production of aerosol mass over the boreal forest (Tunved et al., 2006); sesquiterpene-oxidation products are crucial for the initial stages of particle formation (Bonn and Moortgat, 2003). Isoprene is known to produce additional SOA mass (Claeys et al., 2004; Paulot et al.,

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2009), but at the same time it appears to suppress new-particle formation associated with monoterpene emissions (Kiendler-Scharr et al., 2009; Kanawade et al., 2011). Model analysis which combines BVOC emission algorithms with dynamic vegetation models is the state-of-the art to address these issues (Arneth et al., 2009). BVOC emission algorithms can be evaluated against available BVOC flux measurements (Arneth et al., 2007) and provide input for estimates of natural precursor emissions for the evaluation of the contribution of natural aerosol loading over boreal forests in aerosol-cloud-climate interactions. However, to-date process-understanding on sesquiterpene emissions is insufficient to be included in BVOC emission models, and most models concentrate on isoprene and monoterpenes as precursor sources. BVOC emission estimates based on top-down inversion modeling of satellite formaldehyde retrievals in combination with atmospheric chemistry models are promising; however, at present these estimates suffer from large uncertainties in the satellite information retrievals and associated with the chemical reaction pathways (e.g. Palmer et al., 2003).

Biogenic secondary organic aerosol (BSOA) influences climate mainly by affecting cloud properties, and to a lesser extend by scattering solar radiation back to space (e.g. Kerminen et al., 2005; Lihavainen et al., 2009). Possible offsets between stimulation of BVOC emissions by warming and inhibition by increasing CO_2 represent a chief source of uncertainty for the magnitude of proposed feedback loops (Arneth et al., 2010). SOA are efficient cloud condensation nuclei, with larger number concentrations possibly leading to less precipitative clouds. Faster chemistry due to elevated O_3 levels also increases the concentrations of SOA.

This paper presents an an overview of existing satellite products and methods that have been used to discriminate between natural and anthropogenic aerosols. The use of satellite products, in situ data, models and their complementarity to identify the occurrence of anthropogenic aerosols over the Boreal forest is illustrated. The work focuses on data over Finland as an example because of the availability of quality assured data from stations which are part of recognized networks such as EUSAAR (Philippin et al., 2009), AERONET (Holben et al., 1998) and GAW-PFR (Wehrli, 2008). In earlier

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publications the aerosol optical depth over Finland (Aaltonen et al., 2011) and the aerosol optical properties measured at the Finnish supersite Hyytiälä (Virkkula, 2011) have been analyzed. Global nucleation based on the use of satellite observations alone is discussed in Kulmala et al. (2011).

2 Overview of studies on the use of satellite data to discriminate between natural and anthropogenic aerosols

Few studies have been published on the discrimination between different aerosol types. OMI Absorbing Aerosol Index (AAI) data have been used to distinguish between absorbing (carbonaceous aerosol and dust) and non-absorbing aerosols (sulfate and sea salt) and MODIS Fine Mode Fraction (FMF) to further distinguish fine (carbonaceous and sulfate) and coarse mode (dust and sea salt) aerosols, resulting in one of these four aerosol types in each scene (Kim et al., 2007). The information derived from these two data products is clearly complementary.

The synergy between GOME and ATSR-2 has been used to derive both AOD and aerosol type (Holzer-Popp et al., 2002a, b). However the determination of the aerosol type in this algorithm is based on the a priori aerosol model selected by the algorithm to give the best match between satellite measured and modeled top-of-the-atmosphere (TOA) reflectance, so it is not as clearly utilizing complementary information as the approach by Kim et al. (2007).

The above mentioned approaches could be used to estimate the anthropogenic and natural aerosol contributions. However, further simplification is likely necessary, such as an assumption that all fine-mode AOD originates from anthropogenic sources and coarse mode AOD from natural sources. This assumption is not valid for secondary organic aerosol produced by boreal forests.

An approach to estimate the anthropogenic aerosol optical depth (AOD_{anth}) using fine mode fraction (FMF) and total aerosol optical depth (AOD) from MODIS over-ocean retrievals was developed by Kaufman et al. (2005). FMF for marine, anthropogenic, and

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dust aerosol were assumed to be constant, and derived from Terra MODIS measurements from regions dominated by these specific aerosol types.

This approach is more advanced than simply assuming that fine mode aerosols are exclusively anthropogenic and coarse mode aerosols are natural, since the contributions of natural dust and marine aerosol to fine-mode AOD are empirically accounted for. Kaufman et al. (2005) assumed that all biomass-burning aerosols are anthropogenic and all dust is of natural origin, while they estimated the FMF for both, not assuming that all anthropogenic is in the fine mode and natural in the coarse mode. Yu et al. (2009) further refined and developed the method introduced by Kaufman et al. (2005) by including seasonal and spatial variations in marine FMF. The crucial parameter in these approaches is the estimated fine mode fraction. This approach was applied over ocean only because the MODIS FMF over land is not considered reliable.

Another way to identify anthropogenic aerosols would be the quantification of biomass burning ignited by lightning (natural origin) and mineral dust due to human induced changes of land cover/land use and climate (anthropogenic origin). Ginoux et al. (2010) combined land use data and MODIS measurements to separate the natural and anthropogenic dust sources, assuming that any grid cell with land use of 10 % or more is anthropogenic, otherwise natural.

The methods summarized above were applied to situations with relatively high aerosol concentration as reflected by enhanced AOD. Over the EURASIAN boreal forests AOD is usually low. Although the number concentrations of natural aerosols emitted by boreal Eurasian forests are very high, their initial sizes are so small that they do not affect aerosol properties observed by optical instruments such as scattering, absorption or AOD. It takes several days before these new particles grow into a size range large enough to significantly contribute to the aerosol volume concentration (Tunved et al., 2006) and thus affect light scattering and AOD (e.g., Lihavainen et al., 2009). Therefore discriminating natural aerosol particles formed over the boreal forest from aerosols of anthropogenic origin is a challenge.

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In the ALANIS-Aerosol project the AATSR dual view algorithm ADV (Veefkind et al., 1998; Curier et al., 2009) is used to provide both regional and global information on aerosol properties. AATSR is the Advanced Along Track Scanning Radiometer flying on ENVISAT since 2002. It provides two views, one at nadir and 2 min later a 55° forward view. AATSR has seven wave bands in the visible and infrared regions, centered at 0.555, 0.659, 0.865, 1.61, 3.7, 11.0, and 12.0 µm. With a swath width of 512 km and nadir resolution of $1 \times 1\,km^2$, AATSR reaches global coverage in about five days. The dual view algorithm uses both views to eliminate the effects of land surface reflection on the radiance measured at the top of the atmosphere. Hence an external surface model is not needed in the retrieval. The IR channels together with the VIS/NIR channels are used for cloud detection (Robles Gonzalez, 2003; Curier et al., 2009). The cloud detection procedure consists of three standard tests which are applied to individual pixels. The tests are based on the evaluation of histograms of either brightness temperature or reflectance in both viewing angles. The VIS/NIR channels at 0.555, 0.659, 0.865 and 1.61 µm are used for aerosol retrieval.

The current version of ADV provides not only the AOD at several wavelengths but also the fraction of fine mode aerosol contributing to the AOD for two pre-determined aerosol types. This principle has been demonstrated with ATSR-2 data collected over the Indian Ocean showing that the relative contributions of sea-spray aerosol and anthropogenic pollution vary with distance from the coast (Roblez-Gonzalez et al., 2006). A study on the transport of biomass burning aerosol over Africa showed the changing mixing ratio of two types of aerosol with different absorption coefficients (Robles-Gonzalez and de Leeuw, 2008). Forest fires over the Iberian Peninsula were isolated from the background aerosol (Sundström et al., 2008) and very strong pollution, as characterized with AOD on the order of 3, was detected over China (Sundström et al., 2011). Over Eurasia the AOD is very low with a natural background value on the order of 0.05, which renders it difficult to obtain any additional information such as FMF or it's contribution to total AOD.

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3 Existing satellite data products that can be explored to investigate their potential in discriminating natural and anthropogenic aerosols

Table 1 summarizes several satellite data products that are potentially useful in discriminating between natural and anthropogenic aerosols. This table is not meant to be exhaustive, but rather listing the most suitable candidates for each type of satellite product available from currently flying satellites only. The potential use of the satellite products in Table 1 is discussed below. An initial attempt of using satellite products to obtain information on secondary aerosol formation through nucleation has been presented in Kulmala et al. (2011). This work was based on the use of proxies such as satellite-derived concentrations of SO_2, AOD and UV radiation. The correlation of AOD with short-lived trace gases such as formaldehyde, nitrogen dioxide and sulfur dioxide, all observed by satellites, was demonstrated by Veefkind et al. (2011). The potential extention to other satellite-observables and in particular BVOCs is discussed below.

3.1 Precursor gases and proxies

Aerosols formed within and above the natural boreal forest with diameters smaller than about 100 nm cannot be detected directly with optical instruments such as those used on satellites. An alternative approach to study these particles would be to use proxies for different parameters that influence the formation mechanism such as sulfuric acid concentration. Direct proxies for sulfuric acid concentration include the concentrations of SO_2, hydroxyl radical (OH) and condensational sink (CS) (e.g. Weber et al., 1997; Petäjä et al., 2009). Of these, only SO_2 concentration is directly available from satellites (e.g., SCIAMACHY, GOME-2, and OMI). However, CS can be estimated from AOD which correlates with aerosol surface area, and OH concentrations correlate with UV intensity (Rohrer et al., 2006); both AOD and UV intensity can be obtained from satellite observations.

Atmospheric volatile organic compounds (VOCs) are precursors of SOA. Formaldehyde (HCHO) and glyoxal (CHO.CHO) are two possible intermediates which can be

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produced during the atmospheric oxidation of VOCs. They can undergo further oxidation and thus contribute to the secondary organic aerosol formation. Therefore, information on formaldehyde and glyoxal levels and their variability could potentially be used to identify aerosols with biogenic origin. Column-integrated HCHO estimates are available from satellite measurements (e.g., Shim et al., 2005; Millet et al., 2006, 2008). Such measurements have been used to derive global constraints on surface emissions of BVOCs. HCHO data or glyoxal, CHOCHO, another oxidation product from biogenic BVOC could be potentially used to assess whether information on SOA production and concentrations can be obtained (Lerot et al., 2010). So far the studies have not been very quantitative in terms of SOA formation. Schurgers et al. (2009) hypothesized that BVOC emission in the southeast US contributes to the elevated AOD (seen both from MODIS and MISR). Veefkind et al. (2011) found that high HCHO levels in this region correspond to the pattern of higher AOD values, inferred from MODIS AOD data. Slowik et al. (2010) presented a substantial biogenic SOA event from the Canadian eastern forest with correlated increase in satellite AOD. The ratio of glyoxal to formaldehyde was used by Vrekoussis et al. (2010) to classify the sources of these oxygenated volatile organic compounds according to biogenic and/or anthropogenic emissions. The results indicate that biogenic emissions would favor glyoxal formation, observed to correspond to the highest Enhanced Vegetation Index, EVI.

3.2 Anthropogenic trace gases

As an alternative to the direct detection of aerosols the use of trace gas concentrations as an indication of anthropogenic pollution could be explored. Trace gas column concentrations are retrieved from satellites (SCIAMACHY, OMI) including anthropogenic pollutants such as NO_2, SO_2 or CO. The presence of these anthropogenically produced gases could be accompanied by the presence of aerosols. This is illustrated in a study by Veefkind et al. (2011) who used MODIS-retrieved AOD and OMI-retrieved columns of NO_2, SO_2 and HCHO on spatial correlations between AOD and these trace gases. The results from this study show low AOD to NO_2 ratios over the eastern United

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States and Europe and higher ratios over industrialized regions in eastern Europe and China. Comparison with SO_2 observations suggest that over the latter regions the aerosol content has a stronger contribution from sulphates than over regions where the SO_2 emissions are regulated. In their analyses, Veefkind et al. excluded regions where AOD is dominated by natural aerosols because no correlation with anthropogenic trace gases is expected.

4 Ground-based data

4.1 Ground-based in situ data

There are less than 20 stations providing continuous aerosol data from the boreal forest zone in Eurasia. The measurement sites currently in operation are shown on the map in Fig. 1. There are only few stations in the Eurasian boreal forest that provide observations on a regular basis and report to open data bases. For validation of aerosol information retrieved form satellites some more stations are available, such as AERONET and PFR sites mentioned in Sect. 4.2.

In the analyses presented in Sect. 6 we use data from the SMEAR II station at Hyytiälä (61°51′ N, 24°17′ E, 180 m a.s.l.). Hyytiälä has extensive facilities for measuring forest-atmosphere interaction and has been doing so since 1996 (Hari and Kulmala, 2005). Aerosol size distribution measurements were started with a twin-DMPS system at the end of January 1996, and have been ongoing since then. The DMPS measures submicrometer particles with sizes larger than 3 nm. The sampling is done at 2 m above the ground inside the forest canopy. The station is surrounded by Scots Pine forest. The nearest urban pollution sites are Tampere (ca. 50 km to the southwest) and Jyväskylä (ca. 100 km to the northeast). Of further interest for the current work are the optical aerosol measurments (scattering and absorption coefficients) described in Virkkula et al. (2011) and the CIMEL sun photometer measurements which are part of AERONET (Holben et al., 1998).

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4.2 Ground-based remote sensing data

Ground-based remote sensing over the boreal forest includes several sun photometers which are part of the AERONET network (http://aeronet.gsfc.nasa.gov/index.html) or the WMO-GAW PFR AOD network (http://www.pmodwrc.ch/worcc/index.html). Relevant sites located in the ALANIS-Aerosol Eurasian boreal forest study region are mainly located in Finland. The sun photometers in Helsinki, Hyytiälä and Kuopio are part of AERONET and data are available on the website within a few hours after measurement. The sun photometers in Joikionen and Sodankylä are part of WMO-GAW network and the data are available at the EBAS data base (http://ebas.nilu.no/). Other AERONET stations which could be used are located in Zvenigorod, Moscow, Yekaterinburg, Tomsk, Krasnoyarsk and Irkutsk. However, all of these stations are on the edge of the Boreal forest (57° N or further south) whereas the Finnish stations are all north of 60° N with the additional advantage of the availability of ground-based in situ data.

Ground-based remote sensing using sun photometers has similar limitations as satellite remote sensing as regards the occurrence of clouds and the availability of sun light. The CIMEL (AERONET) and PFR (Precision Filter Radiometer) (WMO-GAW network) data in Finland presented by Aaltonen et al. (2011) show large data gaps, but because these instruments measure from sunrise to sunset with a frequency of 15 min (CIMEL) and 1 min (PFR) more data are available then for AATSR with only one overpass every 2–3 days over Eurasia providing a snapshot around 10:00 a.m.

Aaltonen et al. (2011) classified the sun photometer observations in Finland according to AOD (larger or smaller than 0.2) and Ångström exponent (larger or smaller than 1.1) and associated the occurrences of clean continental background aerosol (natural aerosol in this paper) and anthropogenic aerosol with air mass origin using air mass trajectory cluster analysis. The observations are dominated by clean continental background aerosol whereas the occurrence of anthropogenic aerosol is associated with air masses transported from the east or from the south-east to south.

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5 Chemical transport model

Any transport model can in principle make a distinction between natural and anthropogenic contributions to the aerosol loading through the distribution and nature of sources and subsequent aerosol transport and transformation. The performance of the model depends on how well the sources, sinks and transformation (physical, chemical) processes and the meteorological fields are described. In the present study the global atmospheric aerosol and chemistry model GLOMAP (http://researchpages.net/glomap/) (Spracklen et al., 2005) is used. GLOMAP simulates the evolution of size and composition resolved aerosols, including their interaction with trace gases and clouds. The model includes sea spray and sulfate aerosol, elemental carbon, organic carbon, dust, and a simple scheme for secondary organic material based on monoterpene oxidation products. The host model for GLOMAP is the TOMCAT global Eulerian grid-point chemical transport model that has been developed over the last 10 years. It is used extensively for tropospheric chemical studies. TOMCAT is forced by meteorological analyses (ECMWF or UKMO) updated every 6 h. The model has a variable resolution of typically between $1^{\circ} \times 1^{\circ}$ and $7.5^{\circ} \times 7.5^{\circ}$ and 31 vertical levels between the surface and the tropopause. GLOMAP was the first global model that demonstrated the potential importance of atmospheric nucleation to the global aerosol number budget (Spracklen et al., 2006) and global cloud condensation nuclei production (Spracklen et al., 2008; Merikanto et al., 2009). GLOMAP has been used in several other applications (e.g., Mankletow et al., 2009; Spracklen et al., 2010). For the application of GLOMAP in the ALANIS-Aerosol project the SILAM (http://silam.fmi.fi), (Sofiev et al., 2006) fire emission scheme has been implemented to account for the presence of forest fires over part of Europe and Asia, and their influence on the atmospheric composition over Eurasia.

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6 Results

The AOD over Finland was retrieved using AATSR data and the ADV algorithm for solar zenith angles larger than about 60°. This limits the availability of satellite AOD data to the period from mid-April until the end of September with an increasing coverage of Finland as daylight hours increase. Further limitations are that AOD can only be retrieved in clear sky conditions, the AATSR swath and the presence of clouds. The AATSR swath of 500 km results in a return time of 2–3 days over Eurasia (5–6 days near the equator). The presence of clouds results in an often "patchy" pattern within an AATSR track. Swath and cloud occurence together hide the larger scale distribution on a single day and often there are insufficient data to show the spatial AOD pattern typical for the transport of anthropogenic pollution over a clean area. Because the focus of the ALANIS-Aerosol study is the use of satellite data, and for this particular part of the study the use of AATSR data, the later were used as stand-alone satellite data to make a first selection of cases showing clear or polluted air. This required that AOD over a large part of Finland and / or neighbouring countries would be retrieved and cover one or more of the in situ stations, in this case Hyytiälä. This left only few useful situations, e.g., for 2006 eleven useful retrievals were selected. Four of these were for clean situations and seven show areas with enhanced AOD indicating transport of aerosol from anthropogenic origin. These numbers may be biased by the occurrence of certain circulation patterns and their associated cloudiness (Kusmiercyk et al., 2007). In two of the cases with anthropogenic aerosol the plume clearly originated from Central Europe and in three other cases the source region would be Russia. For the other three anthropogenic cases the origin could initially not be decided based on satellite data alone.

Examples illustrating clean and polluted cases are shown in Figs. 2 and 3. Figure 2 is a composite of AATSR-retrieved AOD (at a wavelength of 550 nm) over the Boreal forest in Scandinavia, Finland and eastern Russia for three consecutive days on 1, 2 and 3 September, 2005. These days the AOD was very low with values of around

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0.05 over the whole area with somewhat higher AOD over a few areas in northern Finland, over the Norwegian mountains and over Estonia, Latvia and Lithuania. These higher AOD values are probably due to presence of clouds as indicated by the adjacent white areas. Undetected clouds would enhance the AOD and often the AOD is also higher near cloud edges. The somewhat enhanced AOD near the Norwegian coast may also be due to the presence of marine aerosol particles. The air mass back (96 h) trajectories (Draxler and Hess, 1998) shown in Fig. 2 were calculated for arrival at three locations in Finland (Helsinki, Hyytiälä and Värriö, see Fig. 1), on 2 September at 12:00. They show that the air masses originate from the clean North Atlantic which explains the low AOD values over the whole study area.

Figure 3 illustrates the effect of transport on the AOD over southern Finland during a short period from 2–6 May 2006. This period will be further discussed below together with results from ground-based in situ measurements corresponding to the cases selected from the satellite observations. These in situ observations were used to verify the conclusions based on satellite data alone. In this exercise only the Hyytiälä data were used because most of the polluted cases occurred over southern Finland which is closest to the pollution sources. Hence the clearest signal would be expected in Hyytiälä. Data used were aerosol scattering and absorption coefficients and particle size distributions. The latter were measured using a differential mobility analyzer (DMPS) in the dry diameter range 3–650 nm and an aerodynamic particle sizer (APS) in the diameter range from 650 nm–15 µm. Aerodynamic diameters were converted to mobility diameters assuming a particle density of 1.8 g cm^{-3} to be able to combine the DMPS and APS data to obtain the full size range; particles in the size range between 0.05–0.1 and a few µm are most effective as regards their effect on the scattering of light in the visible part of the solar spectrum which is observed by optical instruments such as the AATSR. The combined particle size distributions were averaged over 1 h and analyzed for the occurrence of pollutants based on total particle volume concentration (V). Virkkula et al. (2011) showed that in Hyytiälä the scattering coefficient correlates better with total particle volume concentration than with condensation sink

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(which is closer to total particle surface area concentration). The cases were classified as clean when $V < 10\,\mu m^3\,cm^3$, semi-clean when $10\,\mu m^3\,cm^{-3} < V < 20 \mu m^3\,cm^{-3}$ and polluted when $V > 20\,\mu m^3\,cm^{-3}$. Particle size distributions and time series of total number, area and volume concentrations for the cases shown in Fig. 3 are presented in Fig. 4.

Classification based on satellite data alone or on ground-based in situ data alone generally agree but in some cases they also disagree. This is ullustrated by comparison of the satellite and particle size distribution data in Figs. 3 and 4. Figure 3a shows the AOD for 2 May 2006: an area with enhanced AOD (on the order of 0.45) occurs over southern Finland and Estonia while further to the north the AOD decreases gradually to values of 0.1 and smaller. Both the most nothern and the most southern parts of Finland are covered by clouds (indicated by the white areas) and thus the retrieval of AOD was not possible. Hyytiälä is situated in southern Finland which was cloud covered. On either side of the cloud deck the AOD was about 0.45 but in view of the possible effect of clouds on the relative humidity, and thus swelling of the aerosol particles underneath the clouds resulting in enhanced extcinction, no assumption can be made on the spatial variation of the AOD and thus on the AOD in Hyytiälä. It is noted that when the cloud detection routine in ADV was switched off the retrieved AOD in the cloudy area has values between about 1.5 and 3, but there is no smooth transition from the area which was positively identified as aerosol (AOD 0.45). Hence we believe that the cloud detection algorithm correctly identified this area as cloudy rather than mis-assigning high pollution values as clouds. The total particle volume concentration V was around $20\,\mu m^3\,cm^{-3}$ in the morning and increased to $37\,\mu m^3\,cm^{-3}$ around the time of the AATSR overpass to drop to a steady concentration of around $30\,\mu m^3\,cm^{-3}$ during the rest of the day. This is clearly indicative of a polluted episode, following the criteria given above, and confirms the AATSR observation. It is noted that V was determined from particle size distributions measured in their dry state, i.e. at low RH.

The aerosol volume concentrations measured in Hyytiälä decreased substantially to reach a minimum value of about $10\,\mu m^3\,cm^{-3}$ in the afternoon of 3 May to increase

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again to 35 $\mu m^3\,cm^{-3}$ in the afternoon of 4 May and then steady decrease back to 10 $\mu m^3\,cm^{-3}$ in the morning of 5 May when we have the next AATSR overpass with reasonable area coverage over southern Finland. Figure 3b shows that the plume is optically much thinner than on 2 May with a maximum AOD of about 0.2 but the area of elevated AOD has a similar spatial extent as on 2 May. Around the time of the AATSR overpass *V* was about 13 $\mu m^3\,cm^{-3}$ and increased to 20 $\mu m^3\,cm^{-3}$ in the afternoon to drop to below 10 $\mu m^3\,cm^{-3}$ later in the day. The AATSR overpass on 6 May (Fig. 3c) partly overlapped with that on 5 May and the plume has moved more to the SE but still with a similar AOD maximum of 0.2. It is noted that the pollution had disappeared from Hyytiälä as clearly indicated by the low volume concentration of 5 $\mu m^3\,cm^{-3}$ at the time of the AATSR overpass. The spatial AOD pattern retrieved from the AATSR data shows the somewhat patchy behaviour with AOD varying between 0.05 and 0.1 near the Hyytiälä area and higher AOD near the Finnish southcoast.

The conclusions from the ground-based in situ and satellite data are confirmed by the 96-hour back trajectories arriving over Finland on 2 May 2006 (Fig. 3a). The air masses arriving in southern Finland (Helsinki and Hyytiälä) passed over forest fire areas, whereas the air mass arriving in Värriö in Northern Finland did not pass over any forest fires detected by AATSR. Figure 3a also shows the AOD measured on the same day (2 September 2006) over Russia indicating clear air in most of that part of the AATSR track and elevated AOD over the most southern part where the forest fires occurred on the same day and the day before. On 5 May (Fig. 3b) the air mass trajectories over southern Finland were also arriving from the SSE and turned around the sites just prior to arrival. The AATSR data indicate that the air mass arriving in Helsinki passed over the area in southern Finland which was still influenced by transported aerosol with enhanced AOD, whereas the airmass arriving in Hyytiälä had passed over areas with lower AOD (although not clean background). On 6 May (Fig. 3c) we see a different situation for the air masses arriving in Hyytiälä and Värriö versus the one arriving in Helsinki, which again comfirms the conclusions from the ground-based in situ and satellite observations while on the other hand they provide complementary

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information which contributes to explanation which is not available from the observations alone. First we see that all trajectories were from much closer regions than on the previous days (note that all trajectories were calcualted for 96 h), indicating stagnant air in the presence of a high pressure area. The air mass arriving in Helsinki appears to have resided over a relatively small area with enhanced AOD on 6 May as shown by the AATSR retrievals, and likely also on the previous day. In contrast, the air mass arriving in Hyytiälä had resided over the Baltic sea and passsed over clean areas in Finland prior to arrival. The airmass arriving in Värriö had been completely over land during the last 4 days.

GLOMAP model results are used as a third source of information. As an example, Figure 5 shows the global distribution of AOD for 2 May 2006 produced using GLOMAP. The large influence of forest fires on the AOD over Finland, Scandinavia and the area over the North Atlantic to the N and NW is clearly observed and supports the AATSR and ground-based observations. The spatial distribution over the areas for which AATSR-retrieved AOD was presented in Fig. 3 is similar to these satellite data, albeit with much coarser resolution which reveals less detail than the $10^{\circ} \times 10^{\circ}$ AATSR resolution.

7 Discussion and conclusion

The ALANIS-Aerosol project is a feasibility study on the use of existing satellite products for discriminating between natural aerosols emitted by boreal Eurasian forests and long-range transported anthropogenic aerosols. Discrimination between natural and anthropogenic aerosols using satellite observations only has been demonstrated before (see overview in Sect. 2) but usually requires high AOD (>~0.3) and certain assumptions (Holzer-Popp et al., 2002a, b). Over the boreal Eurasian forests the AOD is usually low (<0.2 over 2006–2010, with a natural background on the order of 0.05–0.1). The case presented for 2–6 May shows that useful information can also be obtained for these conditions. Particles formed by nucleation are too small to be optically active

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and it takes several days before they grow into sizes which allow them to be observed with optical instruments.

Particles larger than about 50–100 nm scatter light in the solar spectrum with an intensity that is large enough to be observed by optical instruments such as nephelometers and, provided that their concentrations are large enough, to scatter enough light to attenuate incoming solar radiation and thus influence the AOD.

In this paper we show how satellite and ground-based in situ data are complementary in their detection of transport of aerosol from elsewhere and affecting the AOD and concentration levels over Finland. When the satellite could not observe the aerosol plume due to clouds the local measurements of enhanced aerosol concentrations were indicative of the presence of pollution transported form elsewhere. When the local measurements indicated a clean case, the satellite showed that the plume had moved away from Hyytiälä and was situated more to the SE. Conclusions derived from the ground-based in situ and AATSR measurements are confirmed by air mass trajectories. Initial GLOMAP model results confirm the large influence of forest fires on the AOD over Finland, Scandinavia and the area over the North Atlantic to the N and NW. In future studies all information from satellite and ground-based observations will be used together with the GLOMAP model results to analyze this and other cases on the transport of pollution over Eurasia and the effects of anthropogenic aerosol on this specific environment.

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Table 1. Satellite data products which can potentially be used to discriminate between aerosols of natural and anthropogenic origin.

Data product	Satellite/Instrument	Satellite life-time	Website
SO_2	ESA ENVISAT/SCIAMACHY	2002 – present	http://www.gse-promote.org/services/so2/
	ESA ERS-1/GOME	1995 – present	http://disc.sci.gsfc.nasa.gov/Aura/
	ESA EUMETSAT MetOp-A/ GOME-2	2006 – present	data-holdings/OMI/omso2_v003.shtml
	NASA Aura / OMI	2004 – present	
UV at several wavelengths	NASA Aura / OMI	2004 – present	http://disc.sci.gsfc.nasa.gov/Aura/ data-holdings/OMI/omuvb_v003.shtml
AOD	ESA ENVISAT/MERIS	2002 – present	http://envisat.esa.int/instruments/meris/
	ESA ENVISAT/AATSR	2002 – present	http://envisat.esa.int/instruments/aatsr/
	NASA Terra/MODIS	1999 – present	http://modis.gsfc.nasa.gov/
	NASA Agua/MODIS	2002 – present	http://www-misr.jpl.nasa.gov/
	NASA Terra/MISR	1999 – present	
CHO.CHO	ESA ENVISAT/SCIAMACHY	2002 – present	http://envisat.esa.int/instruments/ sciamachy/
	ESA EUMETSAT MetOp-A/ GOME-2	2006 – present	http://www.doas-bremen.de/glyoxal_from_ scia.htm
	NASA Aura / OMI	2004 – present	http://disc.sci.gsfc.nasa.gov/Aura/ data-holdings/OMI/omso2_v003.shtml
HCHO	ESA ENVISAT/SCIAMACHY	2002 – present	http://www.iup.uni-bremen.de/doas/ hcho_from_scia.htm
	ESA EUMETSAT MetOp-A/ GOME-2	2006 – present	http://disc.sci.gsfc.nasa.gov/Aura/ dataholdings/
	NASA Aura / OMI	2004 – present	OMI/omhcho_v003.shtml
CO	EUMETSAT METOP /IASI	2006 – present	http://www.esa.int/esaLP/ SEMM36BUQPE_LPmetop_0.html
	ESA ENVISAT/MIPAS	2002 – present	http://envisat.esa.int/instruments/mipas/
	NASA Terra /MOPITT	1999 – present	http://terra.nasa.gov/About/MOPITT/ about_mopitt.html
	NASA Aura /TES	2004 - present	http://tes.jpl.nasa.gov/
NO_2	ESA ENVISAT/SCIAMACHY	2002 – present	http://www.temis.nl/airpollution/no2.html
	ESA ERS-1/GOME	1995 – present	http://disc.sci.gsfc.nasa.gov/Aura/
	ESA EUMETSAT MetOp-A/ GOME-2	2006 – present	data-holdings/OMI/omno2_v003.shtml
	NASA Aura / OMI	2004 – present	
LAI	ESA / Several instruments in GLOBCARBON	1999 – present	http://modis-land.gsfc.nasa.gov/lai.htm
	MODIS	2002 – present	
NDVI/EVI	ESA ENVISAT/MERIS	2002 – present	http://earth.esa.int/object/index.cfm? fobjectid=1618
	NASA Terra/MODIS	1999 – present	http://modis-land.gsfc.nasa.gov/vi.htm
	NASA Agua/MODIS	2002 – present	

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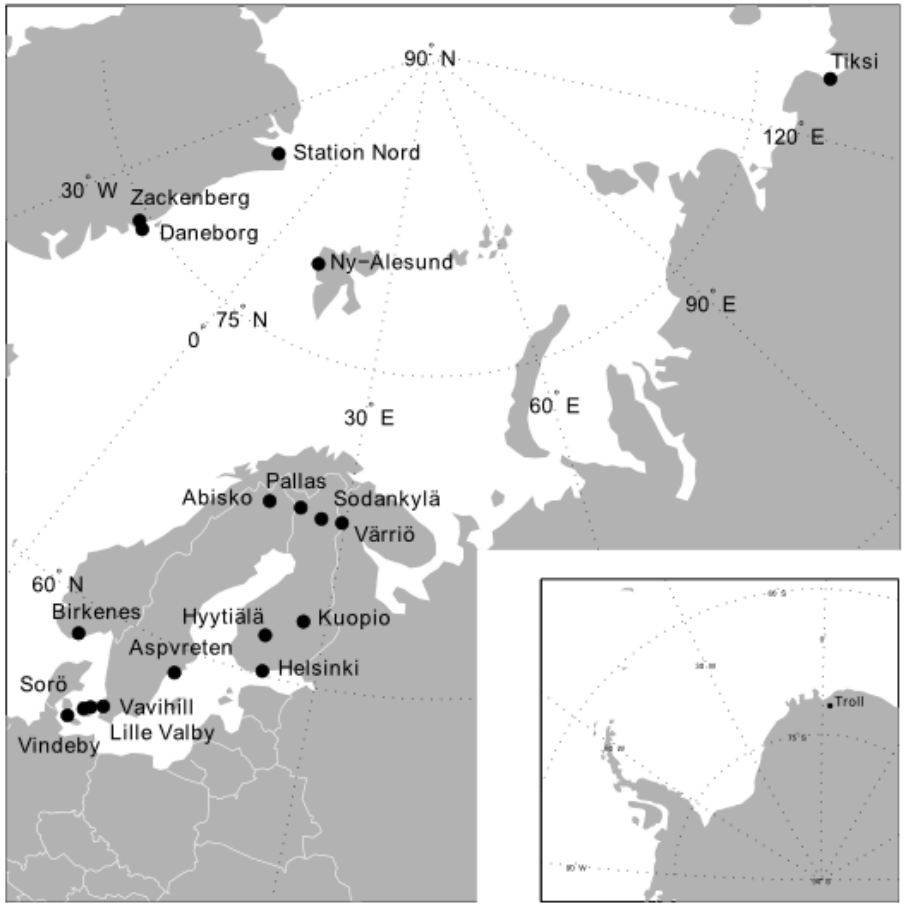

Fig. 1. Measurements sites in the boreal forest zone in Eurasia which currently are in operation.

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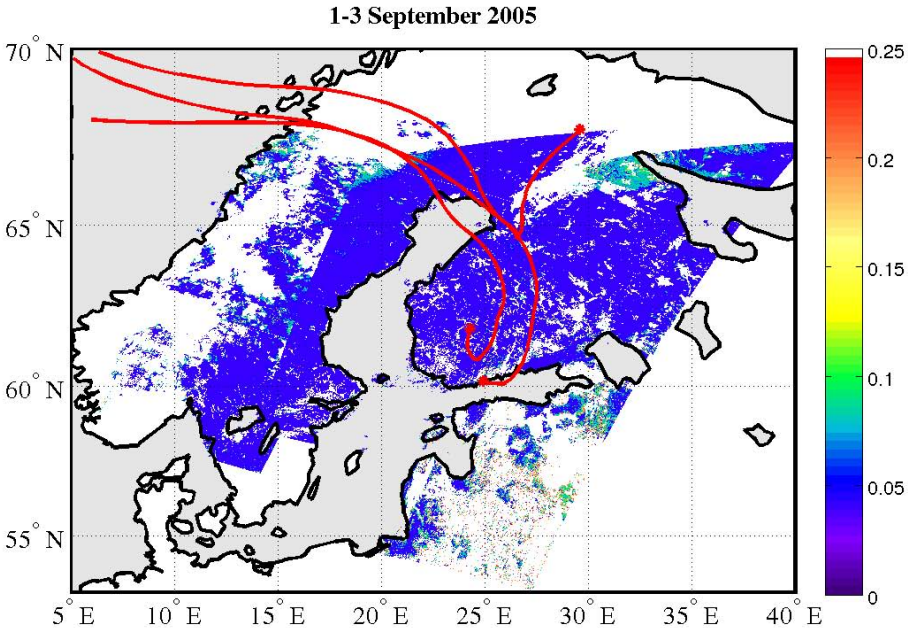

Fig. 2. AOD (550 nm) over Northern Europe retrieved from AATSR observations on 1, 2 and 3 September 2005. The figure is a composite of three tracks, with the most western track on 1 September, the middle one on 2 September and the most eastern one on 3 September. The AOD values are given on the colour scale on the rhs of the map. White areas indicate that no aerosol properties could be retrieved due to the presence of clouds or over the mountains near the Norwegian coast. North of 67° N the solar zenith angle was too small for aerosol retrieval. The red lines are the 96-hour back trajectories for air masses arriving in Helsinki, Hyytiälä and Värriö on 2 September at 12:00 GMT.

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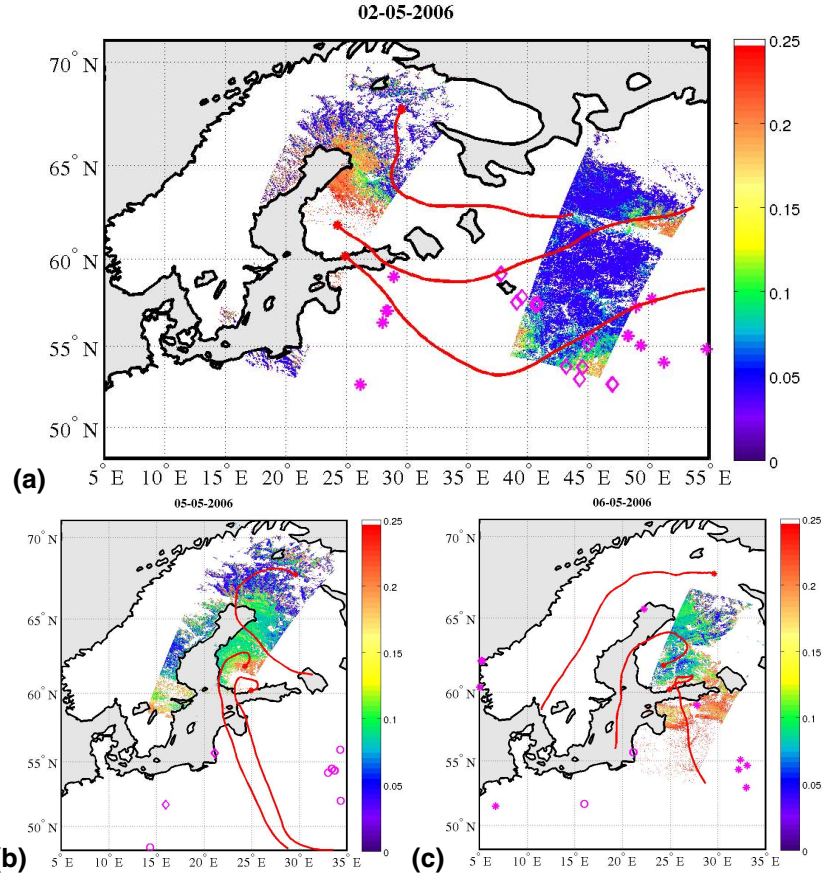

Fig. 3. AATSR-retrieved AOD at 555 nm over Finland and neighbouring countries for 2, 5 and 6 May 2006, see Fig. 2 for further explanation. The symbols in Fig. 3a indicate the presence of forest fires as detected by AATSR on the same day (*), one day before (◇) and two days before (o). The red lines are the 96-hour back trajectories for air masses arriving in Helsiki, Hyytiälä and Värriö at 12:00 GMT.

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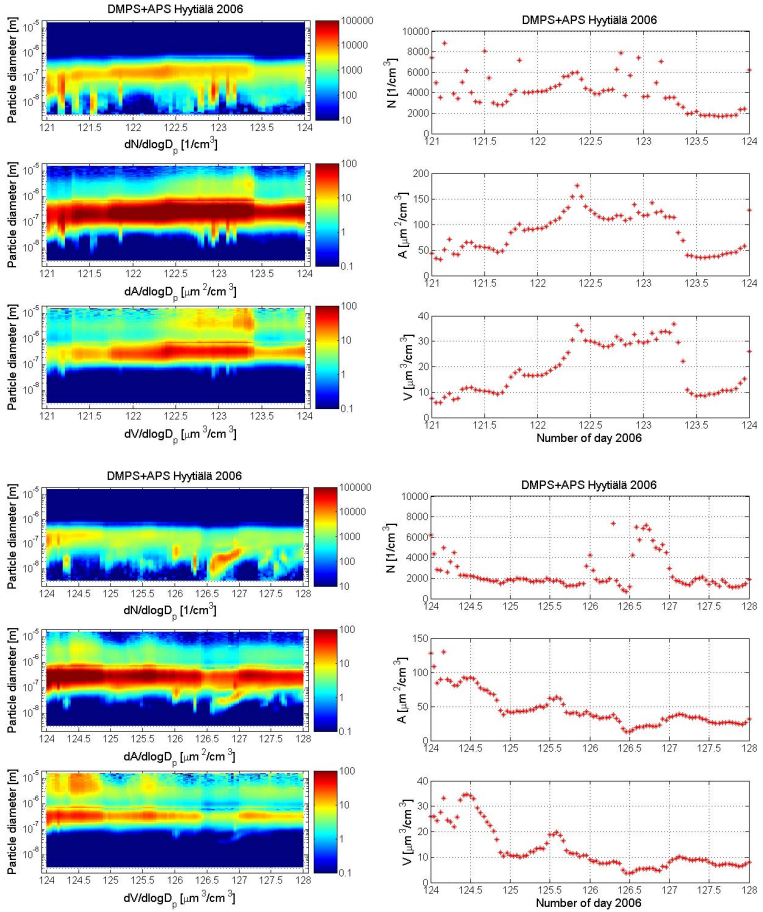

Fig. 4. Particle volume distributions (left) and total number, surface area and volume concentrations (right) measured in Hyytiälä for 2 May (top, doy 122) and for 5 (doy 125) and 6 May 2006 (doy 126) (bottom). The particle size distributions are plotted as time series of the particle diameter with the colour (scale on the right) indicating the particle volume concentrations $dV/d\log Dp$ in μm^3 cm^{-3}.

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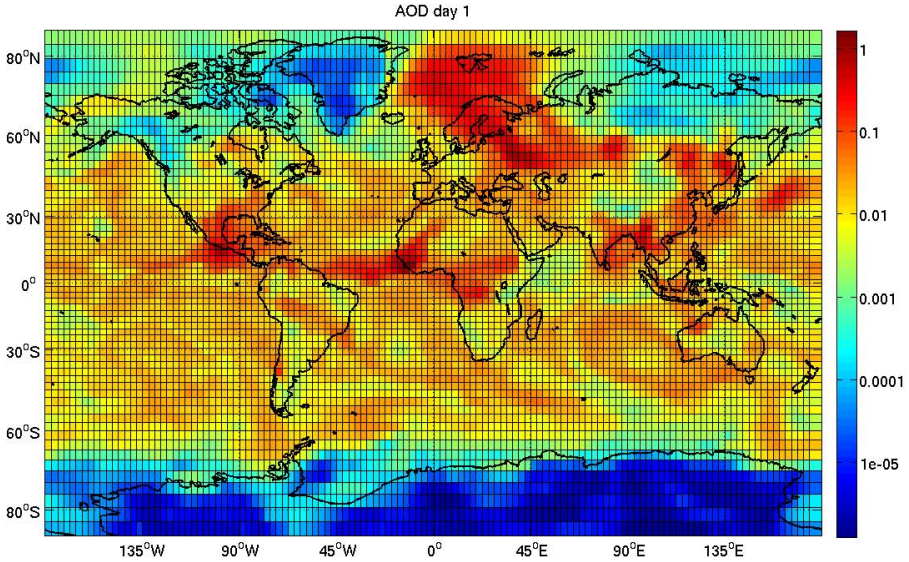

Fig. 5. GLOMAP-calculated AOD for 2 May 2006. The SILAM Fire Emission Scheme has been implemented and the figure shows the large influence of the Russian fires on the AOD over N and NW Europe.

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