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Saharan dust and ice nuclei over Central Europe

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Abstract

Surface measurements of aerosol and ice nuclei (IN) at a Central European mountain site during an episode of dust transport from the Sahara are presented. Transport is simulated by the Eulerian regional dust model DREAM. Ice nuclei and mineral dust are significantly correlated. The highest correlation is found between IN concentration and aerosol surface area. The ice nucleating characteristics of the aerosol with respect to temperature and supersaturation are similar during the dust episode than during the course of the year. This suggests that dust is always a dominant constituent of ice nucleating aerosols in Central Europe.

1 Introduction

In this paper we present surface measurements of atmospheric ice nuclei (IN) in Central Europe together with dust transport modeling of an episode in May 2008, when Saharan dust was transported over a distance of 2500 km northward across the European continent.

Mineral dust aerosols that are suspended in the atmosphere during dust storms are spread over a wide geographic range of the Northern Hemisphere (Prospero, 1999; Kaufmann et al., 2002; Sassen, 2002; Sassen et al., 2003). Dust aerosols directly affect the radiative balance of the earth-atmosphere system through scattering and absorption, changing the atmospheric dynamics and temperature profiles during major dust storms (Nickovic, 2005; Pérez et al., 2006). Furthermore, their cloud-active fraction modifies the size distribution and the phase of cloud particles by acting as cloud condensation nuclei (CCN) and as ice nuclei. This may in turn affect indirectly the atmospheric radiative balance and thus weather and climate (IPCC, 2007; Zeng et al., 2009), as well as the development of precipitation (Levin and Cotton, 2009). Mineral dust particles are particularly known to act as good heterogeneous ice nuclei (Prupacher and Klett, 1997; Field et al., 2006; Knopf and Koop, 2006; Zimmermann et al.,

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2008). The effect of desert dust on the glaciation of clouds has been reported from lidar backscattering and depolarization measurements. For instance, Sassen (2002) observed that the occurrence of dust from Asian sources over the western United States coincided with unusually warm cirrus layers (about 20 °C warmer than normal). These cirrus clouds were presumably produced by the dust-induced glaciation of supercooled liquid altocumulus. Similar observations were reported for Saharan dust events over southern Florida (Sassen et al., 2003) and over Central Europe (Ansmann et al., 2005). However the coexistence of dust and supercooled droplets does not necessarily result in immediate glaciation. From their measurements in Southern Morocco, Ansmann et al. (2008) conclude that ice formation is a strong function of cloud temperature and does not occur above -20 °C. Recently Wiacek et al. (2010) explored the availability of mineral dust particles as ice nuclei for interactions with clouds by analyzing 1.8 million trajectories from the major African and Asian dust source areas over a whole year. By far the largest fraction of cloud forming trajectories encountered conditions of mixed-phase clouds, where mineral dust can initiate primary ice formation.

Although ice nuclei are a necessary ingredient for the glaciation of tropospheric mixed-phase clouds and for the formation of precipitation via the Bergeron-Findeisen-process, their atmospheric concentration is rarely measured. From measurements in Japan Isono et al. (1959) concluded that the deserts of continental Asia are main sources of ice nuclei over Japan. Sassen et al. (2003) and DeMott et al. (2003a) report measurements of ice nuclei number concentration, using the Colorado State University's continuous flow diffusion chamber. In African dust plumes at 1–4 km altitude over Florida extraordinarily high IN number concentrations of up to 10³ L⁻¹ at -36.5 °C were observed. Recently Prenni et al. (2009) reported that ice nuclei sampled in the Central Amazon Basin were primarily composed of carbonaceous material of local biogenic origin and of dust from the Sahara.

Bertrand et al. (1973) found that the ice nuclei concentration (measured at -20 °C) at Abidjan, Ivory Coast, increased from 10 L⁻¹ to around 40–50 L⁻¹ during advection of Saharan dust particles in Harmattan air masses.

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Dust fall and red rains due to long range transport of Saharan dust into Central Europe have been reported in the literature since the 19th century (Ehrenberg, 1849). Ice core samples from non-temperate glaciers in the summits of the Alps show annual layers with reddish-yellow Saharan dust deposits ranging over many centuries into historical times (Tomadin et al., 1996). This transport phenomenon frequently takes place when subtropical air masses are advected from Northern Africa. Over the whole Sahara, a quasi-permanent reservoir of airborne dust can be observed. This reservoir is continuously supplied with dust generated by high surface winds in synoptic-scale extratropical surface depressions, meso-scale density currents related to convection over the mountain regions, small-scale dry convective mixing in the planetary boundary layer, and small-scale disturbances (Schepanski et al. 2007; Knippertz et al., 2009).

The long range transport of African dust into Europe is well documented. Measurements made by an Aerosol Lidar and a multi filter rotating shadowband radiometer in the period 1999–2008 at Lampedusa (Di Iorio et al., 2009) show the evolution of the aerosol vertical distribution in the Central Mediterranean Sea. Dust layers are present in the profile in 38% of the cases throughout the year, and in 57% in summer. The top altitude of dust layers is highest in late spring, up to 9 km altitude dust is then available for transport into northerly directions under suitable meteorological conditions (Barkan et al., 2004). From systematic observations made by the European Aerosol Research Lidar Network (EARLINET) during the period May 2000 to December 2002 Papayannis et al. (2008) found that the number of dust events was greatest in late spring, summer, and early autumn periods, mainly in Southern and Southeastern Europe and South-Central Europe. Multiple aerosol dust layers of variable thickness (300–7500 m) were observed. In less frequent cases dust reached Northwestern, Northern, or Northeastern Europe. The systematic occurrence of Saharan dust events is well documented over a period of many years at the high Alpine Jungfrauoch observatory by Collaud Coen et al. (2004), based on time series of scattering and absorption coefficient measurements. A climatology of dust event observations north of the Alps has been established at the WMO global observatory Hohenpeissenberg in Southern Germany

(Kaminski, 2006). It shows that on average a major dust event occurs each month throughout the year and even more minor events are recorded. Raman Lidar measurements (Mattis et al., 2008) at Leipzig, Germany, over the past 10 years in the framework of the German Lidar Network (1997–2000), reveal a frequency distribution of Sahara dust events peaking in late spring and during fall, which can be taken as representative for Central Europe and also for the Taunus Observatory site.

2 Methods

2.1 Measurements of ice nuclei and of aerosol physical properties

The number concentration of ice nuclei is measured since April 2008 on a regular daily basis at the Taunus Observatory of the Goethe-University on Mt. Kleiner Feldberg, a low mountain range (825 m a.s.l.; 50°13' N, 8°27' E) in Central Germany, approximately 25 km north of the city of Frankfurt am Main. This paper is focussed on ice nuclei observations during a major event of Saharan dust advection in May 2008. The entire ice nuclei record is presented and analyzed in a separate paper (Klein et al., 2010).

For the measurement of the ice nuclei number concentration, atmospheric aerosol was first collected on an inert substrate, which was later analyzed in the laboratory for ice nuclei. Samples were collected from a platform on the roof of the Atmospheric Physics Laboratory at hilltop. The air inlet was at 8 m above ground. A main flow of 60 L min⁻¹ of ambient air was pumped through a Sigma-2 inlet (VDI 1997) and a 25 mm ID×4 m length glass tube into the laboratory. The 50% cutoff diameter of this inlet system as calculated by FLUENT 3-D for 5 m s⁻¹ wind speed is 23 μm. Ice nuclei samples as well as samples for aerosol physical measurement in the 10 nm–20 μm size range were collected isokinetically from the main sample flow, using the pumps of the individual instruments together with appropriate inlet nozzles. The total aerosol number concentration was measured by a TSI 3785 Condensation Particle Counter (CPC). Aerosol size spectra were measured by a TSI 3936 Scanning Mobility Particle Sizer

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(SMPS) in the 10–414 nm diameter range and by a TSI 3321 Aerodynamic Particle Sizer (APS) from 523 nm–20 μm diameter. Spectra of particle number concentration were converted from aerodynamic diameter d_a measured by APS to volume equivalent diameter d_e by using the relation $d_a = d_e (\rho_p / \rho_0 \chi)^{1/2}$ (Hinds 1998), with $\rho_0 = 1 \text{ g cm}^{-3}$, $\chi =$ dynamic shape factor, $\rho_p =$ particle density. From these spectra the particle mass spectra and total mass concentration were derived. A dynamic shape factor of $\chi = 1.5$ and a particle density of $\rho_p = 2.6 \text{ g cm}^{-3}$ were assumed (Tegen and Fung, 1994, Kandler et al., 2007). For comparison to other measurements we consider below from our APS measurements only the mass of particles smaller than $d_{pe} = 10 \mu\text{m}$, for which we use the term PM_{10} . On average 98.5% of the particle mass is measured below $10 \mu\text{m dp}$.

For the sampling of ice nuclei, 10 L of air were pumped at 2 L min^{-1} through an electrostatic precipitator, which deposits aerosol particles on the surface of 47 mm \varnothing silicon wafers (Klein et al., 2010). These samples were then subsequently analyzed in the isothermal static diffusion chamber FRIDGE (Bundke et al., 2008; Klein et al., 2010) for their ice nuclei number. The samples were cooled to the activation temperature of -8°C to -18°C and exposed to relative humidities of 103–119% with respect to ice (RH_{ICE}). Ice crystals grown on the nuclei by deposition and condensation freezing were viewed by a CCD camera, and counted automatically. Samples were usually analyzed within 24 h. IN concentrations reported here will refer to -18°C and $\text{RH}_{\text{ICE}} = 119\%$, unless stated otherwise. After activation and counting of the ice nuclei samples by FRIDGE some of the samples were transferred to an Environmental Scanning Electron Microscope (ESEM) at University of Darmstadt. The elemental composition of individual particles found at those coordinates on the substrate where ice nucleation was observed in the images by FRIDGE, was measured. In addition a variety of meteorological parameters from the weather station of the German Weather Service DWD and of air pollution data are also available at the site.

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2.2 Modeling the dust transport

The DREAM dust model (Nickovic et al., 2001; Nickovic, 2002) was used to study the temporal and spatial evolution of the dust transport event. It is a regional model designed to simulate and/or predict the atmospheric flow of mineral dust aerosol. It solves the Euler-type equation for dust mass continuity. Eight size bins covering the particle diameters from 0.2 to 20 μm (Table 1) are used to describe the size distribution of dust (Pérez et al., 2006). Model data presented below are related to the cumulative mass of particle bins 1–7 called here $\text{PM}_{10}^{\text{dust}}$. DREAM is driven by the atmospheric NCEP/Eta atmospheric model. The mass concentration equation simulates all major components of the atmospheric dust cycle such as dust emission from sources, vertical and horizontal mixing and advection, and dry and wet deposition.

The simulation experiment in this study was performed by running the model in one-day sequences, in which new initial atmospheric conditions from a global model analysis were updated every day and boundary values of model variables were specified every six hours. Since there are not yet satisfactory three-dimensional dust concentration observations to be assimilated, the dust concentration field was initialized every day using 24-h dust forecasts from a previous day. Only in the “cold start” of the model on 18 May 2008 is the concentration set to zero permitting the model several days before the dust event started to spin-up the concentration. The model resolution is set to 50 km in the horizontal and to 24 layers extending up to approximately 16 km in the vertical and approximately 3 model layers in the planetary boundary layer. The exceptionally large south to north extent of the dust event dictated the use of a rather large model domain ranging from the Saharan sources to the Scandinavian Peninsula.

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3 Results

3.1 The Sahara dust episode of May 2008

During 22–30 May 2008 the 500 hPa pressure pattern over the Northeast Atlantic (Fig. 1) was dominated by a persistent low at about 1000 km south of Iceland, with a trough directed southeast towards the Iberian Peninsula and Morocco. Along the eastern side of the trough a strong southerly flow from the Western and Central Sahara across the Mediterranean Sea towards Central Europe prevailed. METEOSAT data indicate that the Western Sahara dust source regions were active during 30–40% of the time in May 2008 (I. Tegen, personal communication). Strong southerly winds through the entire troposphere (40 m s^{-1} at 600 hPa on 28 May) advected dust layers between 27 and 31 May, as indicated by the back-trajectories calculated by the German Weather Service DWD (Fig. 2). For 26–28 May the trajectories point to dust sources in the Central Northern Sahara (Northeastern Algeria, Tunisia, and Libya). The trajectory of 29 May extends to Northern Mauritania. This area northwest of Timbuktu is described as one of the most intense dust sources throughout the year (Prospero et al., 2002) The evolution of the spatial distribution of total column mineral dust during the event as modeled by DREAM (Fig. 3) shows a large scale meandering plume of dust, spreading from the Algerian/Libyan Sahara far northward across Europe to Scandinavia and the Norwegian Sea. In the vertical column over Germany, the modeled concentration peaked between 4.5 and 5.8 km altitude on 29 May (not shown).

Shallow low pressure systems over the western Mediterranean and Central Europe frequently caused clouds and precipitation along the transport path, but did not prevent the transport of significant amounts of dust across the Alps to Central and Northern Europe. Surface measurements of PM_{10} of the German Federal and Regional air pollution monitoring networks (Fig. 4a) exceeded the European legal limits for 24 h exposure ($50 \mu\text{g m}^{-3}$) over wide areas of Southern and Western Germany during the days of maximum dust advection on 28–30 May (Umweltbundesamt, 2008). The surface distribution of $\text{PM}_{10}^{\text{dust}}$ over Germany modeled by DREAM (Fig. 4b) closely resembles

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the measured structures and concentration levels (Fig. 4a), as can be seen for the 24 h means of 29 May. The plume axis is running from the southeast to the central west. In the model the maximum is somewhat shifted towards southwest as compared to the measurements. Both the peak levels of around $100 \mu\text{g m}^{-3}$ measured in the south and the $50\text{--}100 \mu\text{g m}^{-3}$ areas in the central west of the network are well reproduced by the model.

Next, we discuss the event as seen from the Taunus Observatory. Figure 5 compares the mean size distributions of particle number and mass that were measured by APS and SMPS before, during and after the passage of the dust plume. As expected the dust plume increased predominantly the number and mass of particles that are larger than $0.5 \mu\text{m}$ in diameter. Figure 6 shows the evolution of PM_{10} and $\text{PM}_{10}^{\text{dust}}$ at the Taunus Observatory as a function of time. In addition it shows the PM_{10} concentration measured at *Wasserkuppe* (HLUG, 2008a), an air pollution monitoring station at 950 m altitude on top of a mountain 110 km northeast of the Observatory. Some of the $15\text{--}25 \mu\text{g m}^{-3}$ positive offset of PM_{10} against $\text{PM}_{10}^{\text{dust}}$ during 24–26 May could be due to pollution (sulfate, nitrate, soot, organics, etc.) that is not covered in the dust model. The mean PM_{10} concentration in this area in May is $20 \mu\text{g m}^{-3}$ (HLUG 2008b). Both $\text{PM}_{10}^{\text{dust}}$ and PM_{10} start to rise simultaneously in the early morning hours of 27 May and undergo a series of five individual peaks until 31 May (labeled #1–5 in Fig. 6). While the measured and modeled peaks in all three data sets coincide well in time, the peaks #2 and #3 are overestimated by model, whereas peaks #4 and #5 agree well between measurement and model. We believe that these discrepancies mainly result from subscale phenomena that occur on these days and are not represented in the model, namely thunderstorm development, in particular orographically enforced convection over the Taunus hills. Radar echoes show the development of moderate convective precipitation over the Taunus in the afternoon of 28 (peak #2) and 29 May (peak #3) and the northward passage of two large cumulonimbus clusters over the Taunus ridge at 0–05 h and at 19–22 h local time on 30 May.

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3.2 Ice nuclei observations

The ice nuclei number concentration (at -18°C) at the Taunus Observatory during the dust event (Fig. 7) is significantly higher than our monthly mean value of 40 IN L^{-1} for May 2008 and 2009. Peaks of up to $200\text{--}312\text{ IN L}^{-1}$ were recorded on 29, 30 and 31 May. Several features point to mineral dust as the dominant contributor to the IN. First, the record of IN (Fig. 7) parallels the records of $\text{PM}_{10}^{\text{dust}}$ and of PM_{10} (Fig. 6) in surprisingly many details. $\text{PM}_{10}^{\text{dust}}$ shows five distinct maxima, the three largest of which are matched by IN peaks. IN are significantly correlated to the $\text{PM}_{10}^{\text{dust}}$ ($R=0.888$, $n=8$), but less to PM_{10} derived from the measurements (Table 2). Most likely the relation between IN and PM_{10} is obscured by the presence of PM_{10} constituents like soot, acids, sea salt and others that have a poor or even adverse effect on the nucleating ability of the aerosol. IN is also highly correlated to the particle mass in each of the size bins between $1.2\text{ }\mu\text{m}$ and $12\text{ }\mu\text{m}$ (bins 4–7) both for the APS measurements ($R=0.822$, $n=15$) and the model data ($R=0.838$, $n=7$). This size range covers the typical mass median diameters of $1\text{--}3\text{ }\mu\text{m}$, which are reported for mineral dust after long range transport (Duce 1995; Prospero 1999, and references therein).

Second, the IN number is significantly correlated to the aerosol surface, in particular in those size regimes that are dominated by mineral dust. Heterogeneous nucleation is viewed as a surface phenomenon, with the number of IN being related to the particle surface area and probability of occurrence of active sites per unit area of the nucleating material (Pruppacher and Klett, 1997; Phillips et al., 2008). High correlations of IN are found to the modelled total particle surface area ($R=0.919$, $n=7$; Fig. 7) and to surface in each of the size bins. The correlation of IN to the APS-derived particle surface is low for the total surface area of bins 1–7 ($R=0.370$, $n=15$) and the size bins below $1.2\text{ }\mu\text{m}$, but higher in the size range of $1.2\text{--}12.0\text{ }\mu\text{m}$, with a maximum ($R=0.846$, $n=15$) at $1.2\text{--}2\text{ }\mu\text{m}$. From our data we calculate a ratio ξ of activated IN to the total particle surface of $1.08 \times 10^9 \pm 3.1 \times 10^8\text{ IN m}^{-2}$ at 115% relative humidity with respect to ice (RH_{ICE}), and of $3.09 \times 10^8 \pm 9.1 \times 10^7\text{ IN m}^{-2}$ at 119% RH_{ICE} in FRIDGE. These data

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fit well into the observed dependence of ξ on RH_{ICE} (Fig. 8), which was obtained from field and laboratory measurements with the Colorado State University's Continuous Flow Diffusion Chamber (DeMott et al., 2003a,b; Archuleta et al., 2005) and recently compiled by Phillips et al. (2008). It is remarkable that this relation persists among data sets obtained with independent techniques and from very different atmospheric environments, such as Saharan dust over Florida (DeMott et al., 2003b) and Europe (this work), and background tropospheric air from Mt. Werner in the Western US (DeMott et al., 2003a). This and the proportionality of IN to aerosol surface add further evidence to support a key assumption of the parameterizations of heterogeneous ice nucleation which is based on the singularity hypothesis of nucleation (Phillips et al., 2008).

Third, during the days of the peak load of dust and IN (28–30 May) the fraction of activated dust particles, i.e. the ratio of IN relative to the total number of dust particles (based on the APS data), is 3–7%, and thus very similar to the laboratory data of Field et al. (2006) on Saharan dust. From expansion experiments with Saharan dust in the large cloud chamber facility AIDA they reported activated fractions of 3–8% at around -20°C . However, comparing these data one has to bear in mind that activation in the latter experiment is supposed to be mainly due to immersion freezing, whereas FRIDGE observes condensation and deposition freezing. Outside of the plume the activated fraction is at around 0.5–1.5%. We note very little difference between the IN activation spectra (number of IN as a function of temperature and relative humidity) of the samples taken during the dust episode and the average spectra from our two year record (Klein et al., 2010) at the Taunus Observatory (Fig. 8). For intercomparison these data are normalized with respect to the maximum number concentration IN_{MAX} in each spectrum. IN_{MAX} is usually measured at the lowest temperature (-18°C) and highest supersaturation ($\text{RH}_{\text{ICE}}=119\%$) in FRIDGE. Taking IN_{MAX} in each of the samples as 100%, we find that in the dust samples 20–30% of IN_{MAX} is activated at 97% RH_w and 15% at 93% RH_w . In the annual mean 30% of IN_{MAX} is activated at 97% RH_w and 20% at 93% RH_w . The regression line in the scatter plot of both data sets (Fig. 9, right) deviates insignificantly from the 1:1 line. Although the individual IN concentra-

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tions in these data range over more than 2.5 orders of magnitude, the ice nucleating characteristics of the particles appear to be similar. We thus conclude that mineral dust is the dominant contributor to deposition and condensation freezing IN at our site in Central Europe all over the year.

5 Finally, the ESEM analysis of individual aerosol particles that was carried out on the substrates after processing by FRIDGE (see Sect. 2) reveals that approximately 90% of the particles that acted as IN are silicates or Ca-carbonates (Table 3). According to Kandler et al. (2007) higher abundances of carbonates indicate source regions of the dust in the northern Sahara. In addition to the predominant silicate and carbonate
10 particles, a few carbonaceous and metal oxide grains were observed as IN. The high abundance of mineral dust particles among IN observed in the present study is in good agreement with recent findings of Pratt et al. (2009). Based on in situ mass spectrometric measurements in an orographic wave cloud over Wyoming (at an altitude between 7.9 and 8.3 km a.s.l.) these authors observed an average relative number
15 abundance of 50% for mineral dust in the fraction of particles acting as IN. However, in contrast to Pratt et al. (2009), we did not find biological particles acting as IN, although this particle group was found to be a major component at our sampling location in the size range of 1–30 μm (Ebert et al., 2004).

4 Summary and conclusion

20 The concentration of ice nuclei over Europe can be significantly affected by plumes of dust advected from the Sahara across large parts of the continent. Although the dust is transported mainly at several kilometers aloft, the surface concentration of ice nuclei and aerosol are dominated during such situations by dust. The surface concentration of ice nuclei closely resembled the plume structure derived by the DREAM dust transport
25 model. The high activation of 3–7% of the large aerosol particles during the episode adds further evidence to previous findings from laboratory and field studies on the good ice nucleating ability of desert dust. The highest correlation of IN was to aerosol

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surface area. The fundamental assumption of nucleation schemes, that the number of active IN of a particular species of insoluble aerosol is approximately proportional to the total surface area of its aerosol particles (Phillips et al., 2008) is supported. The areal density of activated ice nuclei per aerosol surface as a function of the relative humidity over ice (in the analysis by FRIDGE) agrees well to the empirical relationship obtained by the Colorado State University's group. The ice nucleating characteristics of the aerosol sampled during the dust episode is similar to that of the aerosol during undisturbed conditions. We thus conclude that mineral dust contributes significantly to the abundance and composition of IN in Central Europe during the whole year. Thus robust empirical relationships of IN to dust might in the future be used together with dust modelling as a first approach towards the prediction of IN for a given situation.

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Table 1. Dust size bins introduced in the model. d_{\min} and d_{\max} are minimum and maximum diameter and d_{eff} is effective diameter for each size bin.

Bin number	d_{\min} – d_{\max} , μm	d_{eff} , μm
1	0.2–0.36	0.3
2	0.36–0.6	0.50
3	0.6–1.2	0.90
4	1.2–2	1.56
5	2–3.6	2.6
6	3.6–6	4.4
7	6–12	7.6
8	12–20	14.2

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Table 2. Linear correlation coefficients between ice nuclei number concentration ($\#/l$) and measured or modelled (spectral) aerosol properties. The PM_{10} model data for the correlation analysis were linearly interpolated for the time of aerosol sampling (usually 2/d) within the 3 h time step in the model output.

Parameter correlated to IN	Model	Measurement
PM_{10}	$R=0.888, n=8$	$R=0.609, n=15$
Total particle surface of bins 1–7	$R=0.919, n=7$	$R=0.370, n=15$
Mass in size bins 4–7	$R=0.838, n=7$	$R=0.822, n=15$
Surface, bin 2 of model	$R=0.932, n=7$	
Surface, bin 4 of measurement		$R=0.846, n=15$

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Table 3. Relative number abundance (%) of the different particle groups that acted as IN ($N=112$).

particle group	mean value	95% confidence interval*
silicates	42.9	32.1–54.4
Ca-carbonates	46.4	35.4–57.8
C-rich	7.1	3.1–15.5
metal oxides	3.6	1.1–10.7

* Confidence intervals were calculated assuming a multinomial distribution.

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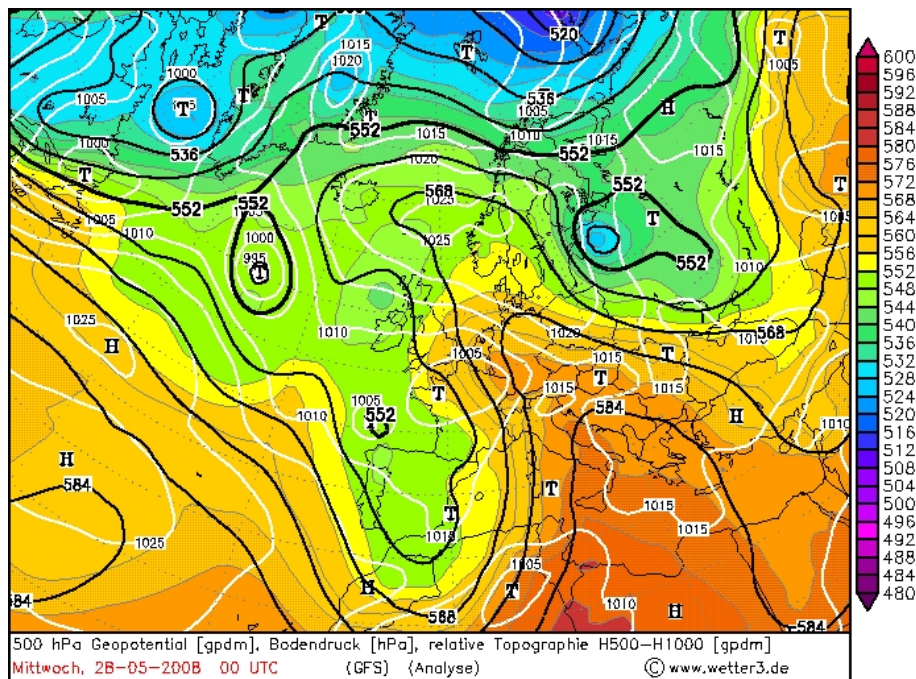


Fig. 1. 500/1000 hPa relative topography and surface pressure on 28 May 2008.

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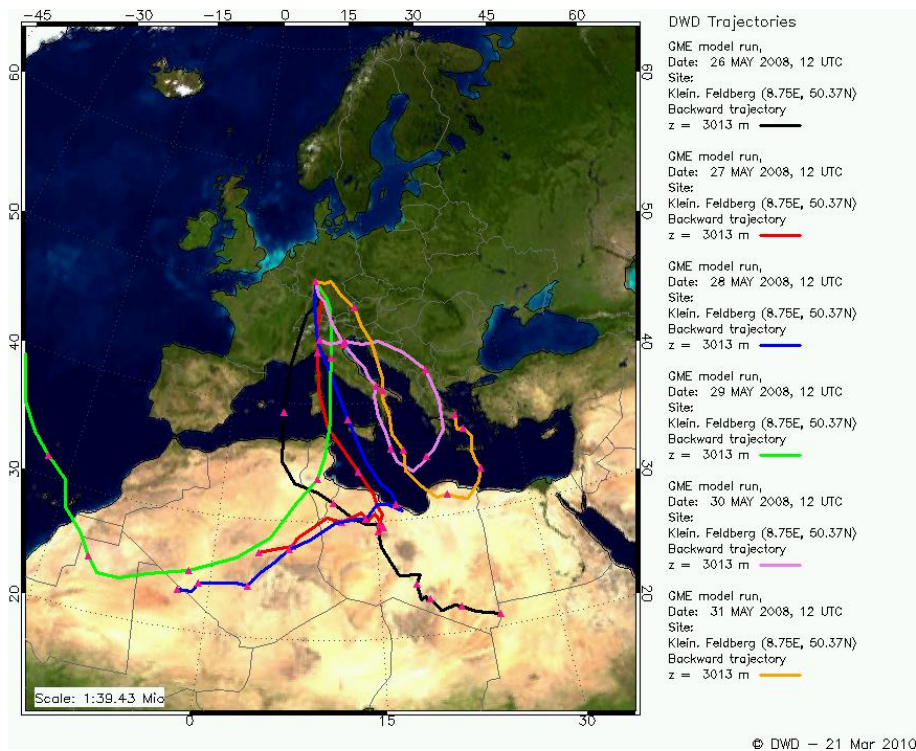


Fig. 2. Backward trajectories arriving at the Taurus Observatory during 26–31 May 2008, calculated by the German Weather Service (DWD).

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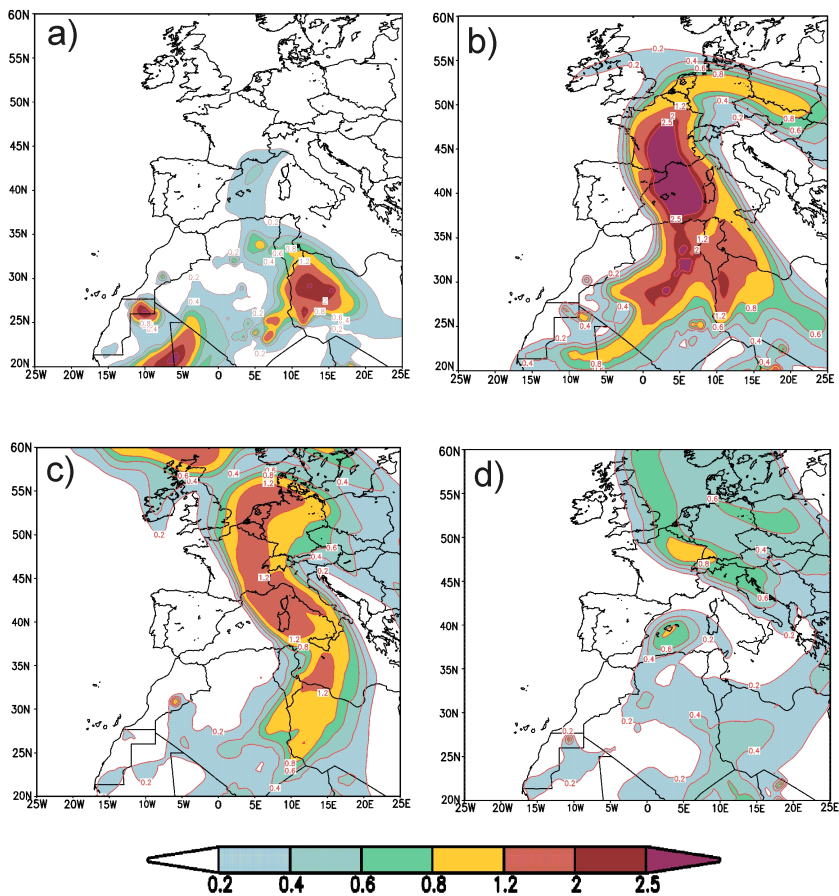


Fig. 3. Total vertical column dust load (vertically integrated concentration in $\mu\text{g}/\text{m}^2$) during the dust event as simulated by the DREAM model for 12:00 GMT on 25 (a), 27 (b), 29 (c) and 31 (d) May.

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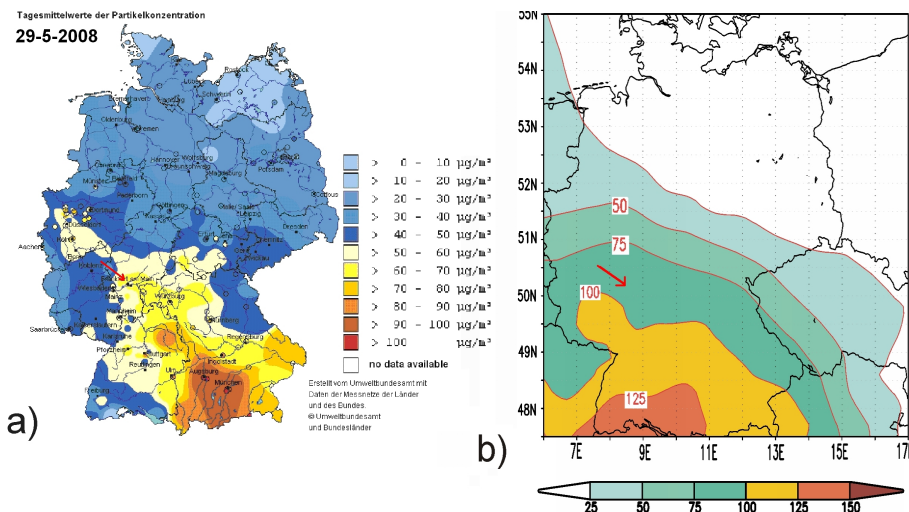


Fig. 4. Diurnal mean surface concentration by mass ($\mu\text{g}/\text{m}^3$) of aerosol particles with diameter $<10\ \mu\text{m}$ (a) *measured* in Federal and statewide networks on 29 May 2008 in Germany, (© data compiled by the German Federal Environment Agency, Umweltbundesamt); (b) *simulated* by DREAM model (size bins 1–7 in Table 1). (red arrows shows measurement-location Kleiner Feldberg).

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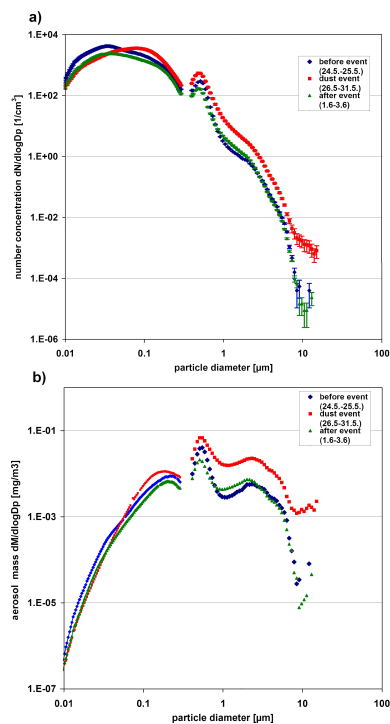


Fig. 5. Aerosol size spectra of **(a)** number concentration (upper panel) and **(b)** mass (lower panel) measured at Taunus Observatory by scanning mobility particle sizer SMPS and aerodynamic particle sizer APS before (blue), during (red) and after (green) the dust event.

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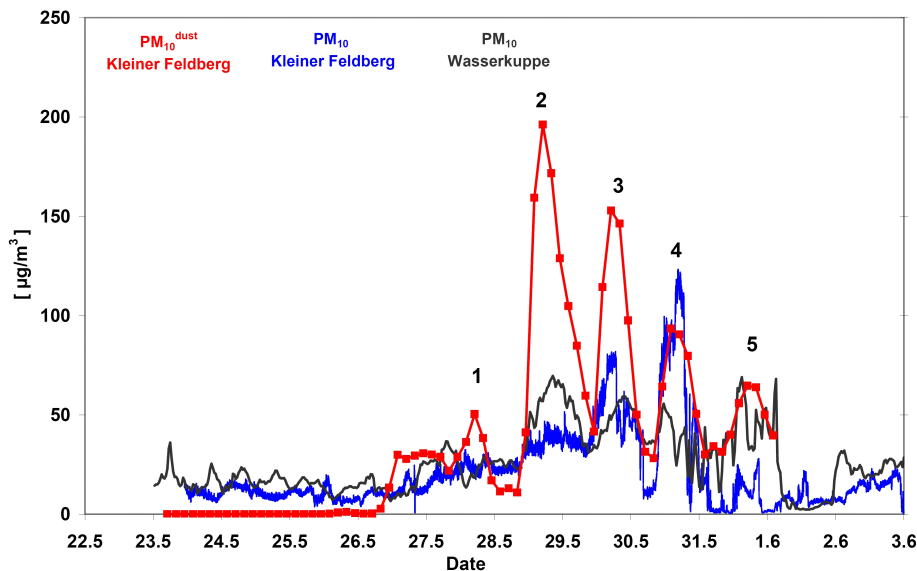


Fig. 6. Measured and modeled PM_{10} mass concentration during the dust event at the Taunus Observatory: *blue curve*: PM_{10} derived from APS size spectra of $d_p < 10\mu\text{m}$; *red curve*: PM_{10} calculated by DREAM model; *black curve*: PM_{10} measured at mountain weather station *Wasserkuppe* 110 km northeast of Observatory (HLUG 2008a).

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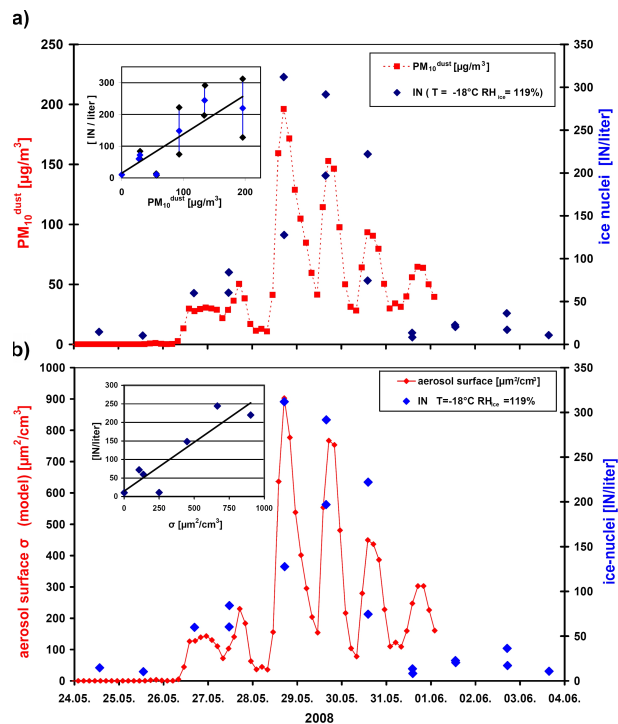


Fig. 7. Upper panel: PM_{10}^{dust} mass at the Taunus Observatory calculated by the DREAM model and number concentration of ice nuclei (#/Liter) measured at Taunus Observatory; Correlation Plot **(a)**: y [IN/liter] = $1.23 \times [PM_{10}^{\text{dust}}, \mu\text{g}/\text{m}^3] + 14.76$ (black diamonds – all samples, blue diamonds averages of subsequent samples taken within a period of one hour), $R^2 = 0.79$. Lower panel: Size-resolved surface dust concentration (σ) in model size bins 1–7 and IN number concentration measured at Taunus observatory; Correlation Plot **(b)**: y [IN/liter] = $0.26 \times \sigma [\mu\text{m}^2/\text{cm}^3] + 14.80$, $R^2 = 0.82$.

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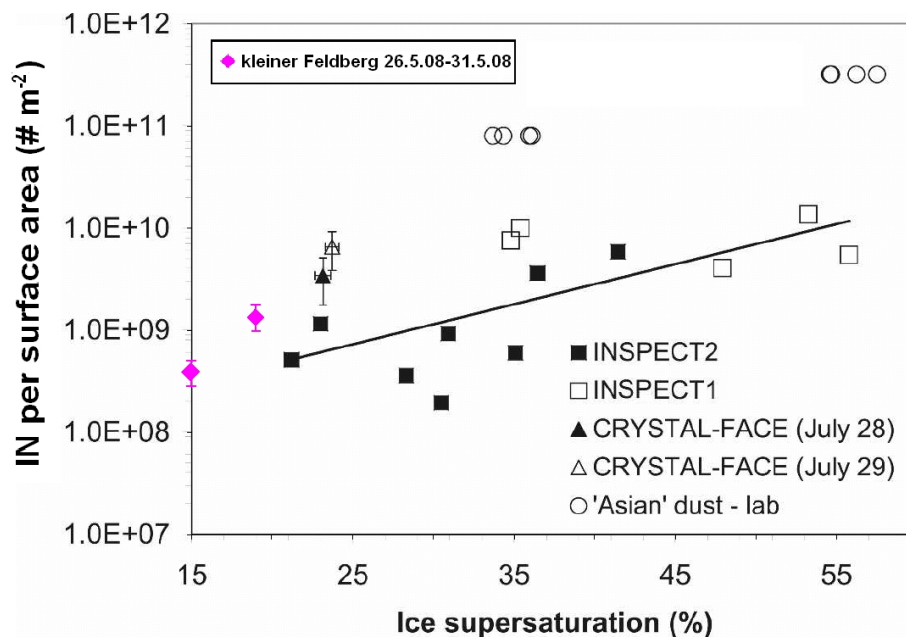


Fig. 8. Number of active ice nuclei per surface area of dust particles as function of supersaturation with respect to ice. Figure taken from Phillips et al. (2008), with data from this work (pink diamonds) added.

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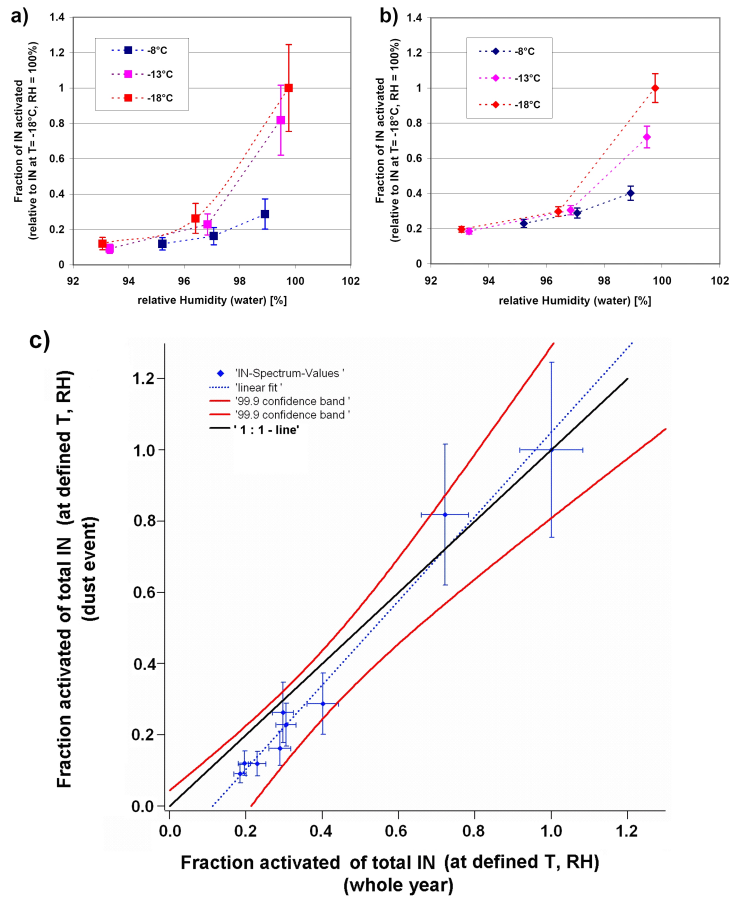


Fig. 9. Panel (a) fractional activation (% of maximum) of IN as function of water saturation during the dust episode. Panel (b) same as in (a) but from 352 samples at the Taurus Observatory during 2008/09. Panel (c) scatter plot of data from (a) vs. data from (b).