



**Aerosol processing
and CCN formation of
an intense Saharan
dust plume during
EUCAARI 2008**

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Aerosol processing and CCN formation of an intense Saharan dust plume during the EUCAARI 2008 campaign

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Abstract

Atmospheric processing and CCN formation of Saharan dust is illustrated through the analysis of an intense Saharan dust event over northern Europe. *The analysis of this dust event was realized through the use of different sets of observations and through the use of numerical models. The altitude of the dust plume was assessed using the CALIPSO observations and our model results.* The major dust plume was transported over Europe between 2 and 5 km above sea level (a.s.l.). This altitude favored the interaction between the dust plume and the mountain ranges of Europe during its transport. This also led the dust becoming mixed with the European anthropogenic aerosol. The analyses of the simulation show that mineral dust particles accumulated soluble material through internal mixing over the Netherlands (51.97° N; 4.93° E). The value of the $CCN_{0.2}/CN$ ratio obtained over the Netherlands ($\sim 50\%$) is much greater than those observed over the Saharan region. In addition over the Netherlands, the CCN measurement reached 14 000 particles cm^{-3} at 0.63 % supersaturation level on 30 May. Our model results reveal that more than 70 % of the CCN concentration observed on 30 May can be explained by the presence of Saharan aged dust. The study reveals that heterogeneous reactions with inorganic salts converted this Saharan mineral dust into compounds that were sufficiently soluble to impact hygroscopic growth and cloud droplet activation over the Netherlands.

1 Introduction

Dust aerosol is considered as one of the most plentiful aerosol species in the atmosphere (Engelstaedter et al., 2006; Washington et al., 2006; Penner et al., 2001), and *it affects significantly the radiation budget* (Koehler et al., 2011; Forster et al., 2007; Haywood and Boucher, 2000). It is also known that dust may affect biogeochemical cycles, acting as a fertilizer for the ocean (Mahowald et al., 2005; Sarthou et al., 2003; Archer and Johnson, 2000). Dust is involved in heterogeneous, multiphase atmospheric chem-

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istry, affecting photo-oxidant concentrations and the composition of precipitation (Laurent et al., 2008; Bauer et al., 2004). Knowledge of the spatial and temporal distribution of dust aerosol and its properties is therefore crucial to the description of atmospheric processes and their climatic effects. The complexity of the interaction of dust with radiation and the biogeochemical cycle are among the most uncertain factors in climate studies and weather prediction. Although progress has been made in understanding the role of dust in the climate system, some uncertainties are still present (Foster et al., 2007). For instance, many uncertainties remain regarding the radiative properties and the atmospheric budget for dust (Ansmann et al., 2011; Bauer et al., 2010; Zender et al., 2004; Ginoux et al., 2004). Numerical models that simulate the processes of dust emission, its chemical and physical transformations and its deposition have improved in recent years to provide a more accurate description of the properties and processes of atmospheric aerosols (Todd et al., 2008). Furthermore, previous studies on the dust source locations and their variability have clearly demonstrated the usefulness of satellite observations in improving models and decreasing the current uncertainties (Tegen et al., 2013; Schepanski et al., 2007).

Dust is mainly produced by the wind erosion acting in arid and semi-arid regions. Half of the world's atmospheric dust originates from the North African deserts, with emissions ranging from 160 to 1600 Tg yr⁻¹ (Engelstaedter et al., 2006). The Saharan desert with the Sahel region is widely regarded as earth's largest source of dust (Engelstaedter et al., 2007; Tanaka and Chiba, 2006). Most of the North African dust emissions are known to come from the Bodélé depression (Koehler et al., 2010; Engelstaedter et al., 2007; Washington et al., 2006; Goudie and Middleton, 2001). A large part of the North African dust is known to come from the Bodélé depression, but other sources have been identified in northern Africa (Schepanski et al., 2007; Caquineau et al., 2002), which can be activated during cyclonic events (Bou Karam et al., 2009). Because Saharan dust can be transported over long distances in the atmosphere, it can affect natural and human environments far away from its sources. The bulk of Saharan dust is transported westward into the Atlantic Ocean, where it can impact the

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ecosystems of the American coast (Goudie, 2014; Prospero et al., 2002) and may alter the biogeochemical cycle in the Amazon Basin and Atlantic Ocean (Jickells et al., 2005; Swap et al., 1992). Europe and the Mediterranean basin can be affected by dust episodes originating in the Saharan region (Pappalardo et al., 2010; Papayannis et al., 2008; Mona et al., 2006; Collaud Coen et al., 2004). Furthermore, *in the intense Saharan dust event, the plume can reach the Scadinavian region* (Bègue et al., 2012; Ansmann et al., 2003).

The significant role of dust in providing ice nuclei (IN) has been rather well identified (Chou et al., 2011; Stith et al., 2009; DeMott et al., 2003; Sassen et al., 2003). A few studies have recently reported the ability of dust to act as cloud condensation nuclei (CCN) (Kumar et al., 2011; Koehler et al., 2010, 2009; Sullivan et al., 2009; Kelly et al., 2007; Perry et al., 2004). When dust is emitted, it is often composed of insoluble or only slightly soluble components. During their transport, the dust particles can accumulate soluble material through internal mixing, which drastically reduces the saturation required for activation (Dusek et al., 2006). The dust particles provide reaction sites for heterogeneous chemical reactions with atmospheric trace gases and pollutants that result in modified dust properties, such as enhanced hygroscopicity (Hatch et al., 2008; Levin et al., 1996). Roberts et al. (2002) have shown that a change of only 20% in the amount of soluble material can have a marked impact on the hygroscopic properties of aerosols that initially contain less soluble material, such as dust aerosols. Soluble coatings on dust are commonly observed in the atmosphere. However, not all dust particles undergo aging as the chemical processing depends on the chemical composition of the dust and thus on its source region (Sullivan et al., 2009). Several studies of Asian dust have shown that its atmospheric processing may have a considerable impact on the activation of Asian dust transported over a long range (Stone et al., 2011; Sullivan et al., 2007; Roberts et al., 2006; Perry et al., 2004; Chen et al., 1997). As far as African dust is concerned, significant sulfate coating on transported Saharan dust and an enhancement of the hygroscopic properties of this dust by the coating has been shown over the Mediterranean basin (Levin et al., 2001; Wurzler et al., 2000; Falkovich et al.,

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2001). Twohy et al. (2009) have shown that Saharan dust commonly acts as CCN over the eastern North Atlantic. To date, only a few studies on the ability of Saharan dust to act as CCN during its transport over northern Europe have been reported. Based on this observation, we present a study of the atmospheric processing of Saharan dust through the analysis of *the major Saharan dust event of the May 2008*.

An intense Saharan dust plume was transported over the Netherlands and the Scandinavian region between 25 and 31 May 2008. The synoptic analysis reported by Hamburger et al. (2011) revealed that the dust event took place in a meteorological situation characterized by strong convective activity associated with the advection of a frontal system over central Europe. *This dust event was firstly reported by Papalardo et al. (2010) in the framework of a comparative study between lidar measurements obtained from Cloud–Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) and the European Aerosol Research Lidar NETWORK (EARLINET).* Klein et al. (2010) have shown that the Saharan dust transported during this episode contributed significantly to the abundance and composition of Ice Nuclei in central Europe. More recently, Bangert et al. (2012) analyzed the impact of the Saharan dust transported on the radiation and cloud formation over western Europe during this dust event of May 2008. Through the use of the regional scale model COSMO-ART, they revealed that, on the one hand, the direct interaction of dust with radiation caused an additional reduction of 40 to 80 W m⁻² in the incoming short wave radiation, whereas the incoming long wave radiation at the surface increased significantly, by about +10 W m⁻². On the other hand, they showed that the number concentration of ice crystals was determined by Saharan dust, due to efficient heterogeneous freezing of the dust. The impacts of the interaction between the Saharan dust plume and convective activity on dust optical properties have also been reported recently by Bègue et al. (2012). Using the meso-scale model Meso-NH, they demonstrated *a dust size filtering induced by the convective activity*, which modified the dust optical characteristics in the measurements recorded over the Netherlands.

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The present paper extends the results of the first study of dust plume properties over the Netherlands, carried out by Bègue et al. (2012). *The measurements recorded over the Netherlands show a large increase in the number concentration of CCN, coinciding with the presence of Saharan dust over the Netherlands on 30 May 2008.* Thus, questions arise about a possible enhancement of the hygroscopicity of the Saharan dust plume by accumulation of soluble material during its transport over the Netherlands. This hypothesis is explored through a methodology combining observations and numerical tools. In this paper, first, the presence of a zone where mixing occurs between the dust plume and European anthropogenic aerosols is examined through an analysis of air mass transport from the emission of the dust aerosols to their arrival over the Netherlands. *Secondly, the impact of the aging of Saharan dust by coating on its hygroscopic properties is examined and discussed. We concentrate particularly on the influence of the chemical composition on the activation of Saharan dust transported over a long range.*

The description of the observations and the model used to investigate the evolution of the dust hygroscopicity is given in Sect. 2. The Sect. 3 presents the interaction between the plume and European anthropogenic aerosols. Then, a qualitative and quantitative evaluation of the enhancement of the hygroscopic properties by coating with soluble material are proposed in Sect. 4. Finally, Sect. 5 gives a summary and some conclusions.

2 Observations and model description

2.1 Observations

The observations were acquired during an intensive campaign named EUCAARI-IMPACT (where IMPACT stands for Intensive Observation Period at Cabauw Tower), which took place in May 2008. During this campaign, airborne measurements were made using the French ATR-42 aircraft. The calibration of the devices onboard the

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ATR-42 aircraft during the EUCAARI-IMPACT campaign and the evaluation of their performance are discussed in detail by Crumeyrolle et al. (2013) and are only briefly described here. The aerosol instrumentation sampled the particles via the ATR-42 community aerosol inlet (CAI) (Crumeyrolle et al., 2013). *This isokinetic and isoaxial inlet* has a 50 % sampling efficiency for particles with diameters around 5 μm (Crumeyrolle et al., 2008, 2010, 2013; McNaughton et al., 2007). The total ambient aerosol concentrations were measured by a Condensation Particle Counter (CPC, TSI model 3010) aboard the ATR42 aircraft every 1 s. The 50 % detection efficiency of the TSI 3010 CPC applies to particles of diameter larger than 10 nm and its relative uncertainty is about 5 % (Mertes et al., 1995). The Cloud Condensation Nuclei Counter (CCNC, DMT model no. CCN-100) onboard the ATR-42 aircraft was a continuous-flow streamwise thermal gradient CCN counter (Crumeyrolle et al., 2013). The design and operating principles of the instrument are based on the work of Roberts and Nenes (2005). The supersaturation was set at 0.2–0.4 % during all the research flights. The chemical composition and mass concentration of the aerosol were analyzed through the use of a Time-of-Flight Aerosol Mass Spectrometer (C-ToF-AMS, Middelbrook et al., 2012; Canagaratna et al., 2007) aboard the ATR-42 aircraft. The AMS provided information on the mass concentration of particulate organic matter (POM), nitrate, ammonium and sulfate. It should also be noted that the upper 50 % cut-off-diameter of the onboard AMS is about 500 nm (Crumeyrolle et al., 2013). As reported by Ansmann et al., 2011, airborne multiwavelength backscatter lidar has proved to be a powerful technique for detecting dust plumes and their properties. In order to analyze the optical properties of the dust plume, the LEANDRE New Generation (LNG) was used in its backscatter version. The LNG airborne backscatter lidar is currently used for aerosol characterization (De Villier et al., 2010; Pelon et al., 2002; Schepanski et al., 2013). During the EUCAARI-IMPACT campaign, the system was operated in backscatter mode with two elastic channels at 1064 and 532 nm. Energies of 10 and 50 mJ were emitted at these two wavelengths, respectively, at 20 Hz repetition rate with a full angle divergence of the laser of 4 mrd at 532 nm and 6.5 mrd at 1064 nm. The profiles of atmospheric extinction coefficient at

532 nm were retrieved using a standard lidar inversion technique (Cuesta et al., 2008; Klett et al., 1985; Fernald et al., 1972).

During the EUCAARI-IMPACT campaign, a complete set of instruments were deployed at the Cabauw Experimental Site for Atmospheric Research (CESAR, 51.97° N, 4.93° E), *which is selected as a supersite* (Kulmala et al., 2009). We used data recorded from the DMT-CCNC (model no CCN-100, Roberts and Nenes, 2005) which operated continuously during this intensive observational period with a supersaturation in the range of 0.1 to 0.7 %. We also used the observations from the C-ToF-AMS and the multi angle absorption photometer (MAAP 5012, Petzold and Schönlinner, 2004) in order to describe the chemical composition and mass concentration of the aerosol over Cabauw. In order to evaluate the temporal and spatial evolution of the pollution aerosol over Europe, the BC mass concentration recorded by other European ground sites with a MAAP 5012 were also used. The ground sites selected were the European Supersites for Atmospheric Aerosol Research (EUSAAR) sites at Monte Cimone (44.11° N, 10.42° E), Puy-de-Dôme (45.46° N, 2.57° E) and Hohenpeissenberg (47.80° N, 11.01° E).

The Cloud–Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) products were used to analyze the dust outbreak. The Cloud–Aerosol Lidar with Orthogonal Polarization (CALIOP) is a two-wavelength polarization-sensitive lidar on board the CALIPSO satellite mission. An overview of the CALIPSO mission is given by Winker et al. (2010). The CALIPSO products used in this work were selected at level 1 because the level 2 products were unavailable for data acquired prior to 14 September 2008. The main level 1 CALIPSO products are the total attenuated backscatter profiles with a vertical and horizontal resolution of 30 and 1 m respectively below 8.2 km (Winker et al., 2009). *A detailed technical description of these data sets is given in the CALIOP Algorithm Theoretical Basis Document* (<http://eosweb.larc.nasa.gov>).

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2.2 Back trajectory model: LACYTRAJ

LACYTRAJ is a kinematic back trajectory code using the ECMWF wind field developed at the Laboratoire de l'Atmosphere et des Cyclones (LACy, France) (Baray et al., 2012; Clain et al., 2010; Dufлот et al., 2010). This code was used to determine the sources of the air masses arriving above Cabauw. Each air parcel was advected using a bilinear interpolation for horizontal wind fields and time, and a log-linear interpolation for vertical wind field. This operation is performed with a time step defined by the user: 10 min in this work, over a six-day period. The sensitivity and comparative study carried out by Clain et al. (2010) with LACYTRAJ and other models such as FLEXPART highlights the capability of LACYTRAJ for back trajectory analysis. Details on this code can be found in Clain et al. (2010).

2.3 Meso-NH Model

The simulations in this study were performed with the meso-scale, non-hydrostatic atmospheric model Meso-NH. This model was developed jointly by the Centre National de la Recherche Meteorologique (CNRM, France) and the Laboratoire d'Aerologie (LA, CNRS) (Lafore et al., 1998). Meso-NH allows simulations from small scale (LES type) to synoptic scale (horizontal resolution ranging from a few meters to several tens of kilometers). Furthermore, the two-way interactive grid-nesting method allows the model to be run simultaneously on several domains with the same vertical levels but with different horizontal resolution. *Different sets of parameterization have been introduced for convection (Bechtold et al., 2001), microphysics (Cohard et al., 2000; Cohard and Pinty, 2000), lightning activity (Barthe et al., 2005), aerosol properties (Shure et al., 1998; Tulet et al., 2003, 2005, 2006) and dust aerosol processes (Grini et al., 2006; Tulet et al., 2010; Moktari et al., 2011). The turbulence scheme is one dimensional along the vertical axis (Bougeault and Lacarrere, 1989). The surface computations include four main surface schemes: Town Energy Balance (TEB, Masson, 2000) for managing the urban areas, ISBA for the natural and agricultural covers (Noilhan and*

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et al., 2003). The physical basis of the model is taken from Marticorena and Bergametti (1995), in which dust fluxes are calculated as a function of saltation and sandblasting processes. ORILAM drives both the dynamical processes and the thermodynamical equilibrium between gases and particles along the Model to Predict the Multiphase Partitioning for Organics (MPMPO) scheme for organic species (Griffin et al., 2005) and the Equilibrium Simplified Aerosol Module (EQSAM, Metzger et al., 2002) for inorganic species in order to solve the condensation and nucleation processes. The aerosol radiative properties (extinction coefficient, asymmetry factor and single scattering albedo) are computed within ORILAM. They are then processed as inputs by the ECMWF radiation scheme implemented and coupled online with Meso-NH and AROME (Aouizerats et al., 2010; Tulet et al., 2008).

Aerosol–Cloud interaction including impact-scavenging is parameterized following a kinetic approach proposed by Tost et al. (2006). The collection efficiency obtained from the scavenging schemes used in this study is calculated by taking the Brownian motion, interception and inertial impaction into account (Slinn, 1979). For the present study, we used the explicit and implicit scavenging schemes (Bègue et al., 2012; Tulet et al., 2010). For the implicit scheme the mass flux is calculated from the deep and shallow convection parameterized following Bechtold et al. (2001), which is based upon the Kain and Fritsch (1993) mass flux scheme. Deep and shallow convective drafts exchange mass horizontally with their environment through detrainment of cloudy air and entrainment of cloud-free air. Subgrid-scale entrainment and detrainment fluxes are diagnosed in terms of grid-scale thermodynamic and dynamic variables (Bechtold et al., 2000). The ICE3 microphysics scheme (Pinty and Jabouille, 1998) is used to resolve the wet deposition parameterization for the explicit scheme (Tulet et al., 2010).

2.3.2 CCN activation scheme

The supersaturation and the number of activated CCN were estimated from the aerosol chemical composition and size distribution computed by ORILAM. The CCN activation scheme used in this study is based on the parameterization proposed by Abdul-Razzak

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and Ghan (2000, 2004), which has been widely included in many models as reported by Ghan et al. (2011). The ability of this scheme to diagnose the number of activated CCN *has been well established in several studies* (Ghan et al., 2013; Bangert et al., 2011; Song and Zhang et al., 2011). The physical basis of the CCN activation scheme is derived from Köhler's theory (1936), in which water vapor condenses on inorganic salt particles to form water droplets. The Köhler theory is used to relate the aerosol size distribution and chemical composition to the number of activated CCN as a function of supersaturation (Abdul-Razzak et al., 2000). The maximum supersaturation is calculated from the aerosol properties and the updraft velocity. The scheme thus takes the effect of aerosol composition (Raoult term) and the surface tension effect (Kelvin term) on hygroscopicity into account. The Raoult term was parameterized by assuming additive behavior of the inorganic salt and the organic surfactant when these aerosol species were internally mixed (Abdul-Razzak and Ghan, 2000; Shulman et al., 1996). Following Abdul-Razzak and Ghan (2004), the Kelvin term was parameterized from Szyskowski's empirical equation (Szyskowski, 1908).

2.3.3 Simulation configuration

The numerical study was performed from the 25 May 2008 at 00:00 UTC to the 1 June 2008 at 00:00 UTC. The numerical domain is extended between latitudes 15.1 and 68.7° N and longitudes 14.9° W and 23.1° E, with a 25 km resolution. The vertical axis has a 60 levels non-linear resolution from the surface to 30 km. The forcing and initialization are driven by ECMWF operational analysis for the dynamics and the global chemical transport model MOCAGE (Dufour et al., 2004) for the chemical gaseous initialization. The dust modes is initialized by three log-normal distributions with median radii of 0.039, 0.32 and 2.5 μm and SD of 1.75, 1.76 and 1.70 respectively. The simulations performed included the implicit and explicit aerosol scavenging schemes. "Off-line" sensitivity analyses of CCN activation were performed for different supersaturations in order to assess the contribution of the aged dust to the CCN concentration measured over Cabauw.

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In order to obtain a correct representation of the gas-phase chemistry and the aerosol particle concentration, an emission inventory of gases and particles was used. The emission inventory was developed by the Netherlands Organization for Applied Scientific Research (TNO) from measurements made during the MEGAPOLI (Megacities: emission, urban, regional and Global Atmospheric POLLution and climate effect and Integrated tools for assessment and mitigation) campaign (Baklanov et al., 2008). This emission inventory includes an hourly-based description for the species: BC, OC, CO, SO₂, NH₃, NO_x and volatile organic compounds (VOC), emitted during the study period. The emissions are located in a domain extending between latitudes 34.2 and 54.9° N and longitudes 9.9° W and 19.9° E. The biogenic emissions were initialized from the MEGAN (Model of Emission of Gases and Aerosols from Nature) inventory with a resolution of 0.5°.

3 Interaction of an intense dust plume with pollution aerosol during its transport over northwestern Europe

3.1 Transport of intense plume over northwestern Europe

The back trajectory calculated with LACYTRAJ for the period of 25–30 May highlights the presence of an air mass over Cabauw on 30 May which comes from the central Sahara (Fig. 1). This is in agreement with Crumeyrolle et al. (2013), who used FLEXPART to show the presence of an air mass in the Boundary Layer (BL) and the lower free troposphere (LFT) at Cabauw, which originated in northern Africa. The back trajectory analysis also revealed that the Saharan air mass left Africa at the end of 26 May and reached Europe two days later. On 30 May, the Saharan air mass continued its spread to Cabauw via eastern France and Switzerland (Fig. 1). It should also be noted that the chronology obtained from the back trajectory analysis is consistent with that reported by Bègue et al. (2012) from satellite observations.

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An overview of the dust event at 01:00 UTC on 28 May and 02:00 UTC on 29 May 2008 is given in Fig. 1a and b through the dust burden and wind fields simulated by Meso-NH. *The wind flow at 700 hPa revealed an African vigorous flow moving northeast towards the Mediterranean basin (Fig. 1a and b). Over the Mediterranean basin, the Saharan air mass was embedded in a flow moving towards the northwest and moved toward the Scandinavian region (Fig. 1a and b).* According to Bègue et al. (2012), this flow was produced by a trough extending along the European and African coast and an area of high-pressure located over central Europe. *The advection of the Saharan plume toward the Scandinavian region was modulated by the strength and position of the high pressure associated with the frontal system.*

On 28 May 2008 at 01:00 UTC, the model results show *an area of high values of dust burden extending in a large area from central Algeria to Switzerland crossing through northwestern Italy (Fig. 1a). Inside this zone, the values are greater than 8 g m^{-2} , especially over northwestern Libya and Sardinia where values of 10 and 9.5 g m^{-2} , respectively, are simulated.* On 29 May, *the area of high values of dust burden has vanished (Fig. 1b). The simulated dust burden has decreased by more than half in the Italian and Mediterranean regions (values no greater than 4 g m^{-2}).* This considerable decrease is the consequence of the interaction between the dust plume and the convective activity. Bègue et al. (2012) have shown that precipitations scavenged the majority of dust over the Mediterranean and European regions, particularly the dust coarse mode. In contrast, the dust burden increased over Scandinavia, with the maximum value (around 7 g m^{-2}) simulated over Norway (Fig. 1b). Overall, there is fairly good agreement between the simulations presented here and those reported in the literature (Bangaert et al., 2012; Pappalardo et al., 2010).

From the CALIOP observations (Fig. 2a), the vertical structure of aerosols can be assessed over a long range, here from northern Africa to central Europe at 01:00 UTC on 28 May (see the CALIPSO track in Fig. 1a). As checked using the feature mask of CALIPSO products (not shown), observed total attenuated backscatter (ATB) signals ranging from 1×10^{-3} to $5 \times 10^{-3} \text{ km}^{-1} \text{ sr}^{-1}$ (corresponding to scattering ratio up

to 3) were due to the Saharan plume. Over the sources in the region (24.6° N; 7.7° E–36.5° N; 10.9° E), the dust plume stretched from the surface to 6 km a.s.l. (Fig. 2a). The major dust plume was transported towards the Mediterranean and European regions between 2 and 5 km a.s.l. It can hence be assumed that the dust plume interacted with the foothills of the African and European mountains during its transport. Thus, the elevation of the dust plume height when it came out of the African continent was very likely the result of the interaction between the plume and the foothills of northern African mountains (Fig. 2a). The Meso-NH simulation reproduced the features of the vertical structure and the observed ATB signals fairly well (Fig. 2b). However, the simulated plume over the Mediterranean and European regions was thinner than the observed plume. In consequence, the major dust plume over these regions was simulated between 2 and 4 km a.s.l. (Fig. 2b). As already discussed, Bègue et al. (2012) showed the interaction between the dust plume and the convective activity over Europe, particularly on 29 May, when the convective activity was the most intense. From CALIOP observations (Fig. 3a), the vertical structure of clouds and aerosols can be assessed over a long range, here from the Mediterranean to northern Europe at 02:00 UTC on 29 May (see the CALIPSO track in Fig. 1b). The observed ATB signals larger than $10^{-2} \text{ km}^{-1} \text{ sr}^{-1}$ were due to clouds. The high clouds were particularly observed over northern France (48.2° N; 4.0° E) whereas midlevel clouds were observed from Belgium to Scandinavia (Fig. 3a). The simultaneous presence of clouds and dust were particularly observed over the Netherlands and Scandinavia. The interactions between the dust and clouds took place between 2 and 4 km a.s.l. (Fig. 3a). The features of the vertical structure of the dust plume were acceptably reproduced by Meso-NH whereas the vertical structures of clouds were insufficiently reproduced (Fig. 3b). The main reason for this discrepancy can be attributed to the fact that a subgrid resolution of 25 km is not sufficient to reproduce the convective activity properly. It can be noticed that Meso-NH reproduced correctly the dust transport toward northwestern Europe.

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3.2 Temporal and spatial evolution of the anthropogenic aerosol

The BC mass concentration and the wind field at the surface simulated by Meso-NH on 28 and 29 May are depicted on Fig. 4a and b respectively. On 28 May, the high BC mass concentrations are mainly located in northern and central Europe (Figure. 4a). Model results show a large area of high BC mass concentration extending from the English Channel to Norway passing along the shore of the Netherlands and Denmark. Within this area, the values exceed $1.9 \mu\text{g m}^{-3}$, particularly over the English Channel and the coast of the Netherlands, where values of 2.5 and $2.2 \mu\text{g m}^{-3}$ are simulated respectively. A second belt of high BC mass concentration (1.9 to $2.2 \mu\text{g m}^{-3}$) simulated on 28 May extends in a large area from southeastern France to the Netherlands (Fig. 4a). On 29 May, the second belt of high BC mass concentration located over central Europe has disappeared and, in the northern region, the simulated BC mass concentration has decreased by more than half (value not exceeding $1.1 \mu\text{g m}^{-3}$) (Fig. 4b).

The simulated sulfate concentrations and the wind field at the surface on 28 May and 29 May are shown in Fig. 4c and d respectively. On 28 May, areas with high sulfate concentrations are located over northern Europe and Corsica. Over northern Europe, a wide area of high sulfate concentrations (maximum value of $6.2 \mu\text{g m}^{-3}$) extending from Norway to the Netherlands is simulated (Fig. 4c). On 29 May, the high sulfate concentration areas are mainly situated over northern and central Europe (Fig. 4d) with a local maximum over the English Channel where the maximum concentration of sulfate ($6.5 \mu\text{g m}^{-3}$) is simulated. Overall, the simulation results show that the concentrations of anthropogenic aerosol are particularly high over northern Europe. This analysis of the spatial distribution of the anthropogenic aerosol is in agreement with the work of Hamburger et al. (2011), *who have shown an accumulation of aerosol inside the Planetary Boundary Layer (PBL), and particularly over Cabauw*.

The observed BC mass concentration is compared with the concentrations simulated by Meso-NH in Fig. 5. The Mont Cimone station is located in the northwestern corner of Italy, which is the site with the lowest BC mass concentration among the sta-

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tions selected. The observed BC mass concentration ranged from 0.2 to 0.9 $\mu\text{g m}^{-3}$ with a maximum value occurring on 25 May. After 25 May, BC concentrations quickly decreased to under 0.5 $\mu\text{g m}^{-3}$ until 31 May (Fig. 5a). This temporal evolution appears to be correctly simulated. The BC mass concentration observed over Puy-de-Dôme ranged from 0.3 to 1.2 $\mu\text{g m}^{-3}$ (Fig. 5b). We note that the BC mass concentration evolution over Puy-de-Dôme follows a daily cycle with a maximum observed at midday and a minimum observed during the night. Over the Hohenpeissenberg station, the observed BC mass concentration ranged from 0.3 to 1.8 $\mu\text{g m}^{-3}$ with the maximum occurring on 25 May (Fig. 5c). The BC mass concentration increased to 1.8 $\mu\text{g m}^{-3}$ on 25 May and slowly decreased afterwards to 0.9 $\mu\text{g m}^{-3}$ until 31 May, which is in agreement with the simulation results. However, we note the presence of a time shift of 2 days between the maximum value observed (25 May) and simulated (27 May). It can be noted for the three stations mentioned above that the BC mass concentration tends to decrease during the study period. The Cabauw station, which is located in a rural area in the central part of the Netherlands, is the site with the highest BC mass concentration in comparison with the three others. In contrast to what happens at the other stations, the BC mass concentration observed here tends to increase during the study period *with values extending from 0.5 to 2.5 $\mu\text{g m}^{-3}$* (Fig. 5d). In spite of an underestimation (0.3 $\mu\text{g m}^{-3}$ on average), the temporal evolution of the BC mass concentration over Cabauw is fairly well reproduced by Meso-NH.

The discrepancies between Meso-NH and the observations may be attributable to several sources. First, a possible source of error can come from the fact that Meso-NH is an atmospheric forecasting model and drives the evolution of its meteorological fields itself. *Given that Meso-NH makes its own forecast, a drift can hence appear in the forecast and increase as the simulation advances away from the initial conditions. Another possible source of error* can come from the fact that the aerosol concentrations are calculated for grid cells with 25 km horizontal resolution. However, the overall temporal evolution of BC mass concentrations simulated by Meso-NH is in acceptable agreement with the observations. Moreover, both the observations and the simulations

show that the highest anthropogenic aerosol concentrations are mainly located over northern Europe.

3.3 Mixing of the dust plume with the European pollution aerosol

In order to highlight a possible interaction between the dust plume and European pollution, a vertical cross section of the dust and anthropogenic aerosol mass concentrations between the surface and 6 km a.s.l. along the trajectory obtained from LACYTRAJ between 28 and 30 May (Fig. 1) was drawn from the concentrations calculated by Meso-NH and is shown Fig. 6. During 28 May, a dust concentration of $500 \mu\text{g m}^{-3}$ spread along the western coast of Italy towards Switzerland at between 1 and 3.8 km a.s.l. Given that the plume continued its propagation along the eastern coast of France, the diminution of the vertical coverage of the dust plume (2–3 km a.s.l.) and the dust concentration ($400 \mu\text{g m}^{-3}$) simulated on 29 May can be explained by the interaction between the plume and the foothills of the Vosges mountains. On 30 May, the dust plume moved from Belgium to the Netherlands, where it separated into two layers. These two main dust layers were separated by a region of weak concentration (not greater than $250 \mu\text{g m}^{-3}$). *The first layer was located between 2 and 4.2 km a.s.l. with a dust concentration range of 370 to $400 \mu\text{g m}^{-3}$.* In contrast, the second layer was located near the surface and was thinner than the first (0.8–1.5 km a.s.l.) with dust concentration ranging from 320 to $370 \mu\text{g m}^{-3}$. Bègue et al. (2012) described this structure in two layers as resulting from the interaction of the dust plume with the convective activity. The present simulations show that this structure in two layers can be also due to the interaction of the plume with the foothills of the Vosges mountains during its transport toward the Netherlands.

An overview of the simulated carbonaceous (BC + OC) and inorganic salts mass concentration along the dust plume trajectory during the period of 28–30 May are given in Fig. 6. The simulated carbonaceous mass concentration reaches $7 \mu\text{g m}^{-3}$, with the maximum located at 1.5 km a.s.l. The vertical coverage of the carbonaceous component extends from the surface to 5 km a.s.l. Thus, we note the interaction of

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the carbonaceous component with the dust plume on 28 May during its spread towards Switzerland (Fig. 6). Moreover, the dust plume also interacted with the organic component on 30 May over Belgium and the Netherlands (Fig. 6). The simulated inorganic salts mass concentration reached $40 \mu\text{g m}^{-3}$ with the maximum located near the surface over Italy. The vertical coverage of the inorganic salts was lower than the carbonaceous component, in particular over central Europe where, on 28–29 May, the vertical coverage of the inorganic salts extended from the surface to 1 km a.s.l. On 30 May, the vertical extension of the inorganic salts increased and ranged up to 2 km a.s.l. over Belgium and the Netherlands (Fig. 6). The simulations thus show that the dust interacted with the organic salts mainly over Belgium and the Netherlands. It can also be noted that the mixing occurred essentially with the dust from the first layer. Although the mixing between the dust and the inorganic salts occurred mainly over Belgium and the Netherlands, it can be observed that the amount of inorganic salt mixed with the dust was greater than the carbonaceous component. The above results clearly show the mixing between the dust plume and the anthropogenic aerosol, particularly over the northern part of Europe. Now, we propose to quantify the impact of this mixing on the hygroscopic and CCN properties of the dust plume. In the following section, the consequence of this mixing over the Netherlands on the dust hygroscopic properties will be discussed in detail.

4 Enhancement of the hygroscopic capacity of the dust plume over the Netherlands

4.1 Chemical composition of the aerosol

The extinction coefficient obtained from the LNG lidar on board the ATR-42 aircraft on 30 May from 13:11 to 14:01 UTC over the Netherlands is shown in Fig. 7. *During this period the ATR-42 flew over a domain extended between latitudes 52.57 and 51.88° N and longitudes 6.34 and 4.99° E.* The vertical cross section obtained with LNG ob-

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servations reveals that the extinction coefficient ranges from 0.01 to 5 km⁻¹ between 0.8 to 3 km a.s.l. (Fig. 7a). As shown previously with the CALIPSO product (Sect. 3.1), *the meteorological context over northern Europe is characterized the presence of high and midlevel clouds during this period.* The observed extinction coefficients higher than 0.4 km⁻¹ were hence due to clouds. The LNG observations show a permanent cloud layer at 0.7 km a.s.l. (Fig. 7a). In these lidar observations, we also note the presence of clouds between 2.2 and 2.5 km a.s.l. embedded in a high scattering and depolarizing layer. These clouds layers are more heterogeneous than those observed at 800 m above the surface. The aerosol air mass indeed appears to be structured in two main layers separated by a region of weak extinction coefficient (from 0 to 0.06 km⁻¹).

It should be noted that this structure, obtained from LNG observations, is quite similar to the structure of the simulated dust plume over the Netherlands. The first aerosol layer is observed between 1.6 and 2.8 km a.s.l. with the extinction maximum (0.16 km⁻¹) located between 1.8 and 2.5 km high. *The second layer is separated in two parts by an area of low values (not greater than 0.01 km⁻¹) and is situated near the surface (0.6–0.7 km a.s.l.).* We note that this aerosol layer is continuously masked by the cloud layer (Fig. 7a). *The evolution of the extinction coefficient was reproduced by Meso-NH for comparison (Fig. 7b).* The simulated extinction coefficient compared relatively well with the observed one. It should be noted that this structure, obtained from LNG observations, is quite similar to the structure of the simulated dust plume over the Netherlands. The contribution of the dust to extinction was analyzed over this region from the Meso-NH simulation. The results reveal that 90 % of extinction is due to dust. Bègue et al. (2012) demonstrated that optical parameters (scattering, extinction, single scattering albedo) measured during this flight from 13:50 to 14:05 UTC was mainly due to the dust accumulation mode. This predominance of the dust accumulation mode is consistent with the findings of Crumeyrolle et al. (2013). Based on the SMPS measurements recorded onboard the ATR-42, they showed an enhancement of the accumulation mode particle number concentration in both BL and LFT. They also showed

an enhancement of the total mass concentration in the intermediate layer (1–3 km), suggesting efficient long-range transport of aerosol particles.

The mass concentration of inorganic salts (nitrate, sulfate, ammonium) and OC obtained with the AMS (type: c-TOF) onboard the ATR-42 during the same flight is shown in Fig. 8. Because of the presence of clouds, data were not recorded everywhere during the flight. The NH_4 mass concentration increased and reached its maximum value ($5.3 \mu\text{g m}^{-3}$) at 0.8 km a.s.l. from 13:11 to 13:25 UTC, and reduced afterwards to $0.2 \mu\text{g m}^{-3}$ between 1 and 2.8 km a.s.l., before oscillating between 0.1 to $5 \mu\text{g m}^{-3}$ until the end of the flight (Fig. 8a). The evolution of sulfate and OC looks similar to the evolution of the ammonium mass concentration. Thus, the sulfate and OC mass concentrations reached their maximum values (10 and $7 \mu\text{g m}^{-3}$ respectively) near the surface, and decreased together thereafter to $0.1 \mu\text{g m}^{-3}$ between 1 and 2.8 km a.s.l., before oscillating between 0.1 and $5 \mu\text{g m}^{-3}$ for the sulfate, and between 0.1 and $2 \mu\text{g m}^{-3}$ for the OC until the end of the flight (Fig. 8b and d). The nitrate mass concentration increased to $5 \mu\text{g m}^{-3}$ at 0.8 km a.s.l., and quickly decreased to $0.1 \mu\text{g m}^{-3}$ between 1 and 2.8 km a.s.l. Unlike other species, the amplitude of the nitrate concentration variation was very high (from 0.1 to $14.8 \mu\text{g m}^{-3}$) between 1.1 and 2.5 km a.s.l. from 13:30 to 13:55 UTC (Fig. 8c). It can also be observed that the nitrate concentrations decreased quickly to $0.1 \mu\text{g m}^{-3}$ at 0.5 km a.s.l. from 13:58 to 14:01 UTC. As reported by Crumeyrolle et al. (2013), peaks in nitrate may originate from natural (marine aerosol) or anthropogenic sources (industry exhaust). We note that Meso-NH reproduced the evolution and the magnitude of the concentration recorded onboard the ATR-42 over the Netherlands fairly correctly.

The observed mass concentration at Cabauw is compared to the simulated ones in Fig. 9. The evolution of the mass concentration recorded by the AMS (type: c-TOF) during the period from 25–30 May at Cabauw is marked by a significant increase in the concentration on 30 May for the four species. We note that the evolution of the three inorganic salts (ammonium, sulfate, and nitrate) is fairly similar. First, the concentration of the inorganic salts increased from 25 to 27 May. Then it decreased on 28 May,

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before finally increasing rapidly again from 29 to 30 May (Fig. 9). The observed ammonium mass concentrations ranged from 0.5 to $6 \mu\text{g m}^{-3}$ with the maximum appearing on 30 May (Fig. 9a). It can be also observed that the mass concentration of sulfate was less than that of the other species, with a maximum value of $4.8 \mu\text{g m}^{-3}$, also observed on 30 May (Fig. 9b). The evolution of the nitrate mass concentration was characterized by its wide range, extending from 0.1 to $17 \mu\text{g m}^{-3}$, observed on 28 and 30 May respectively (Fig. 9c). The evolution of the OC was characterized by a weak variability of the concentration compared to the inorganic salts. The OC mass concentration was confined between 1.6 and $5 \mu\text{g m}^{-3}$, except on 30 May where the concentration rose sharply to $7.8 \mu\text{g m}^{-3}$ (Fig. 9d). It is also worth noting that Meso-NH reproduced the evolution of the concentrations acceptably well for the different chemical species measured by the AMS at Cabauw.

Both the observations and the simulation show that the air mass over the Netherlands on 30 May included a mixture of dust with the anthropogenic aerosol. The fraction of these different species considered as internally mixed was analyzed from the Meso-NH simulation. The results reveal that, near the surface at Cabauw and at altitude (between 1 and 3 km a.s.l.) around this site, the composition of the mixture was fairly similar. More than 50 % of the mixture was made up of dust. The organic component represented an average of 15 % of the total mass, with more half of the contribution due to SOA (~ 10 % of the total mass). Despite of the mixing of the dust plume with BC and OC over Italy (Sect. 3.3), the contributions of BC and OC represented only 3.5 and 5 % on average respectively. The fraction of the inorganic salts was higher than the organic component, with 24.5 % on average. As expected on the basis of AMS observations, the fraction of nitrate was more significant than that of the other inorganic salts, with values around 13.5 %. Hence the major components were dust, followed by SOA and nitrate. Note that the simulated SOA concentrations were not compared to observations because of a lack of SOA observations over the Netherlands. Nonetheless, the chemical composition of the aerosol obtained as internally mixed was found to be consistent with previous studies of atmospheric processing of mineral dust particles

in urban areas (Almeida et al., 2014; Reddington et al., 2013; Kumar et al., 2011; Stone et al., 2011; Putaud et al., 2004).

It is reasonable to assume that the hygroscopic behavior of the mineral dust was caused by a coating of secondary sulfates, nitrates or organics. The chemical and physical processes and coating of inorganic salts (mainly sulfate and nitrate) change not only the mixing state and optical properties but also the hygroscopic properties of the aerosol (Leng et al., 2013; Rose et al., 2011; Wang et al., 2010; Jimenez et al., 2009; Reid et al., 1998). Gibson et al. (2007) have shown that the CCN activity of insoluble mineral dust components is enhanced dramatically when they are internally mixed with a small amount of an aqueous salt. Thus, the heterogeneous reactions with reactive gases, including nitric, hydrochloric and sulfuric acids, can convert insoluble mineral dust into slightly soluble compounds or compounds that are sufficiently soluble to play an important role in hygroscopic growth and cloud droplet activation (Ram et al., 2014; Sullivan et al., 2009). The dust hygroscopicity is controlled by its chemical mixing state, which is determined by its mineralogy and the chemical reaction pathways it experiences during transport (Sullivan et al., 2009). The results mentioned above confirm that the atmospheric processing of this Saharan dust led to an evolution of its hygroscopicity.

4.2 Hygroscopic properties of the dust plume

The concentration of CCN in a given population of aerosol is a crucial parameter for understanding the ability of a particle to act as a nucleating agent. This ability depends on its size as well as the coating of hygroscopic species (Ram et al., 2014; Gunthe et al., 2011; Dusek et al., 2010; Sullivan et al., 2009). The CCN activity was effectively predicted using Köhler theory (Köhler, 1936) based on physicochemical properties of the solute, such as its mass, molecular weight, density, size and activity coefficient. The CCN activity was calculated at the same supersaturation level that was chosen for making the measurements with the CCNC counter. The calculation was made by taking the simulated mass and number concentration of the different chemical species

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and their molecular weight into account, in addition to the simulated aerosol size. The evolution of the measured and simulated number concentration of CCN at 0.2 % supersaturation ($CCN_{0.2}$) during the flight of the ATR-42 on 30 May from 13:11 to 14:01 UTC is shown in Fig. 10a. The simulated $CCN_{0.2}$ concentration plotted in Fig. 11a was estimated from the aerosol accumulation (with median radius of $0.65 \mu\text{m}$) and fine modes (with median radius of $0.082 \mu\text{m}$). The observed $CCN_{0.2}$ concentration was relatively constant at around 80 particles per cm^3 at 0.8 km a.s.l. from 13:11 to 13:25 UTC, and *increased promptly thereafter* to 700 particles per cm^3 between 1 and 3 km a.s.l. Then, the observed concentration varied from 80 to 650 particles per cm^3 between 2.8 and 3 km a.s.l., before reaching its maximum value (900 particles per cm^{-3}) at 2 km a.s.l. from 13:50 to 13:55 UTC (Fig. 10a). It can thus be observed that, on average, the lowest $CCN_{0.2}$ concentration was mainly located near the surface. It is noteworthy that the evolution of the $CCN_{0.2}$ concentration is predicted fairly well by taking the aged Saharan dust particles into account.

To characterize the relationship between $CCN_{0.2}$ and total aerosol population in the atmospheric column, the $CCN_{0.2}/\text{CN}$ ratio was calculated as a measure of hygroscopicity of the aerosol population (Fig. 10b). When its ratio is 0 %, no activation of aerosol can occur to form cloud droplets whereas, when its ratio reaches 100 %, all aerosol particles can be activated to become droplets. The measurements of the CPC 3010 were used to provide the CN concentration required to calculate the $CCN_{0.2}/\text{CN}$ ratio. Unfortunately, CN concentrations were not recorded for the total length of the flight due to the presence of clouds. The value of the $CCN_{0.2}/\text{CN}$ ratio obtained from observations ranged between 10 and 50 %. The low values (less than 20 %) were mainly observed near the surface (0.7–1.5 km a.s.l.) whereas the maximum values of the ratio were correlated with the maximum values of the $CCN_{0.2}$ concentration. This is in agreement with the simulation results. In particular, it should be noted that the $CCN_{0.2}$ concentration peak from 13:50 to 13:55 UTC was associated with the maximum value of the $CCN_{0.2}/\text{CN}$ ratio (50 %). This was further corroborated by an enhancement in nitrate mass concentration (Fig. 8). The value of the $CCN_{0.2}/\text{CN}$ ratio obtained was much

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greater than those observed over the Saharan region. During the AMMA campaign, the CCN/CN ratio obtained was less than 15 % in the Saharan Air Layer (Crumeyrolle et al., 2008). This reinforces our conclusion that heterogeneous reactions with inorganic salts converted this insoluble Saharan mineral dust into compounds that were sufficiently soluble to impact hygroscopic growth and cloud droplet activation over the Netherlands.

Figure 11 depicts the evolution of the measured and simulated number concentration of CCN at 0.16, 0.30, 0.45 and 0.63 % at Cabauw. A large, rapid increase in the number concentration of the CCN can be observed for the four supersaturation levels between 29 and 31 May, with a maximum value of 14 000 particles cm^{-3} at 0.63 % supersaturation. The observed number concentration of CCN on 30 May was double the mean value observed during the EUCAARI-IMPACT campaign. Before 29 May, the CCN concentration at the four supersaturation levels was found to be fairly constant, except for the two weak peaks of CCN concentration observed on 27 and 28 May coinciding with higher BC mass concentration (Fig. 5). We note that these two weak peaks are not found in the simulation. A possible source of this discrepancy could be an underestimation of BC mass concentration by Meso-NH on 27 and 28 May (Fig. 5). Using the chemical transport model GEOS-CHEM, Riipinen et al. (2011) have shown that organic components have a significant influence on the growth of ultrafine particles and in CCN production. The simulated CCN concentration at Cabauw compared fairly well with the observations, and especially the high peak of CCN concentration, which was acceptably reproduced at the four supersaturation levels (Fig. 11). As previously observed with the ATR-42, this peak of CCN concentration coincided with a significant increase in the mass concentration of inorganic salts (Fig. 9), in particular for nitrate. On average, more than 70 % of the CCN concentration observed on 30 May can be explained by the presence of the aged Saharan dust (Fig. 11). The remaining 30 % could be attributed to other processes that can enhance the dust hygroscopicity, such as cloud processing, which is not taken into account in the simulations. Gibson et al. (2007) revealed that interaction between raindrops and the dust particles, both in and around

lands. In contrast, it was well identified from the simulation that the plume was mainly mixed with carbonaceous matter over Italy, likely leading to the adsorption of organic gases onto dust particles.

From the LNG observations onboard the ATR-42, the vertical structure of the aerosol layer over the Netherlands was assessed. The main aerosol layer was located between 1.8 and 2.5 km a.s.l., and the presence of aerosol was probed between 0.6 and 0.7 km a.s.l. in spite of the presence of clouds. *The Meso-NH simulations were compared well* with the LNG and AMS observations. It was shown that the extinction coefficient obtained from the LNG observations was mainly due to the Saharan dust. The presence of the dust plume over the Netherlands led to an enhancement of the accumulation mode particle number concentration in both BL and LFT, which was found to be in agreement with Crumeyrolle et al. (2013). The analyses of the simulation have shown that mineral dust particles accumulated soluble material through internal mixing over the Netherlands. It results that the major components of the mixture were dust, followed by SOA and nitrate. The value of the $CCN_{0.2}/CN$ ratio obtained over the Netherlands ($\sim 50\%$) was much greater than those observed over the Saharan region. In addition, the maximum of the $CCN_{0.2}/CN$ ratio was correlated with the maximum values of the $CCN_{0.2}$ concentration. This demonstrates that heterogeneous reactions with inorganic salts converted this Saharan mineral dust into compounds sufficiently soluble to impact the hygroscopic growth and cloud droplet activation over this region. The CCN measurement at Cabauw revealed a peak of the number concentration of CCN on 30 May, with a maximum of $14\,000\text{ particles cm}^{-3}$ at 0.63% supersaturation. As a result of the simulated CCN concentration, on average, more than 70% of the CCN concentration observed on 30 May can be explained by the presence of the Saharan aged dust. Thus, the atmospheric processing of Saharan dust is shown to be the main process by which this peak of CCN was produced. It is also known that, during the cloud processing, the mineral dust can enhance its hygroscopic properties through a series of additional processes including chemical reaction in the aqueous phase (Smoydzin et al., 2012; Sullivan et al., 2007; Matsuki et al., 2010). Further analysis of

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the dust microphysical properties, including chemical reaction in the aqueous phase, *could be considered in a next study*. In conclusion, our results confirm that changes in dust chemical composition due to atmospheric aging can play a significant role in determining the CCN activity.

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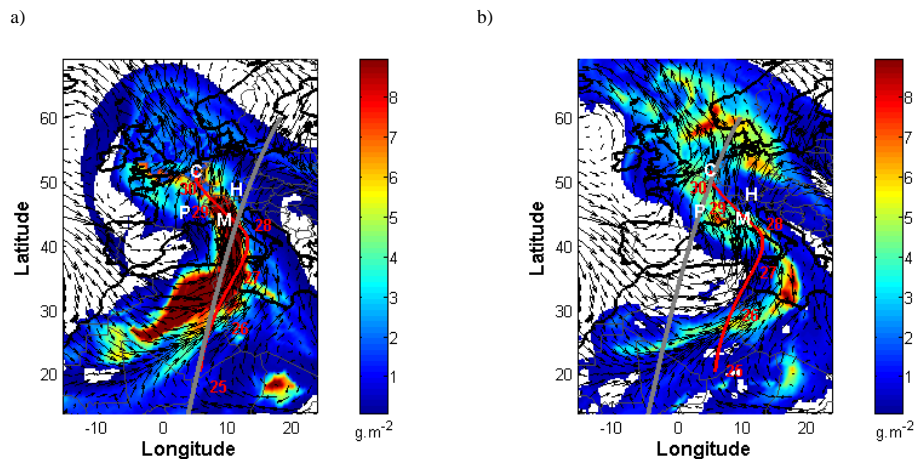


Figure 1. :Dust load (shading, $\text{g}\cdot\text{m}^{-2}$) and 700 hPa wind (vectors) simulated by Meso-NH at 12:00 UTC on (a) 28 May and (b) 29 May 2008. Nighttime CALIPSO overpasses are indicated by the gray line. Six-day backward trajectories calculated during the 25–30 May 2008 period are indicated by the red line.

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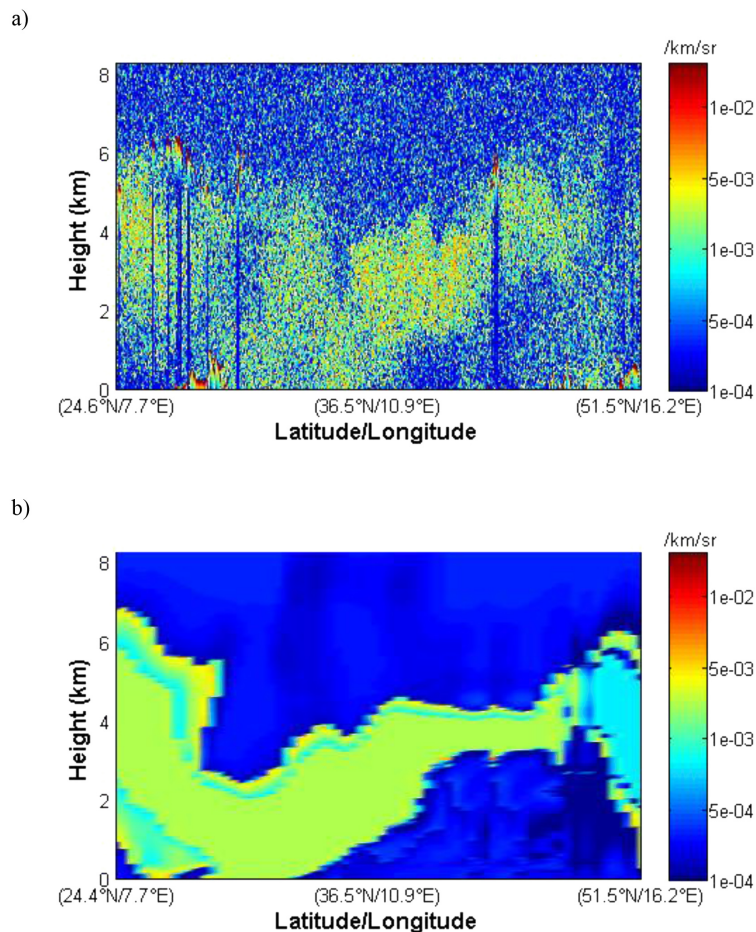
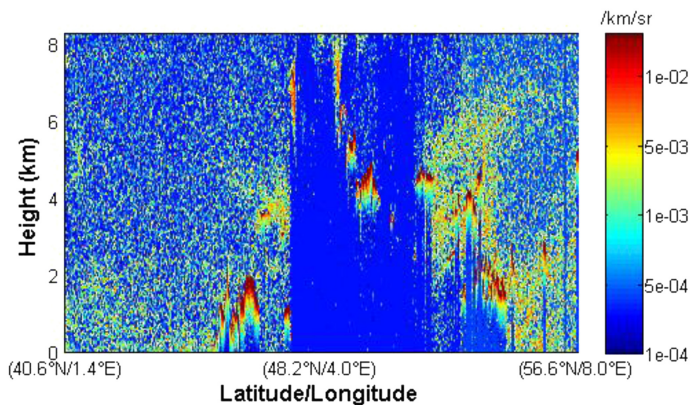


Figure 2. The total attenuated backscatter ($\text{km}^{-1} \text{sr}^{-1}$) at 532 nm from **(a)** the CALIPSO product and **(b)** the Meso-NH simulation for the overpass at 01:22–01:27 UTC on 28 May 2008. The CALIPSO overpass on 28 May 2008 is indicated by the gray line on Fig. 1a.

a)



b)

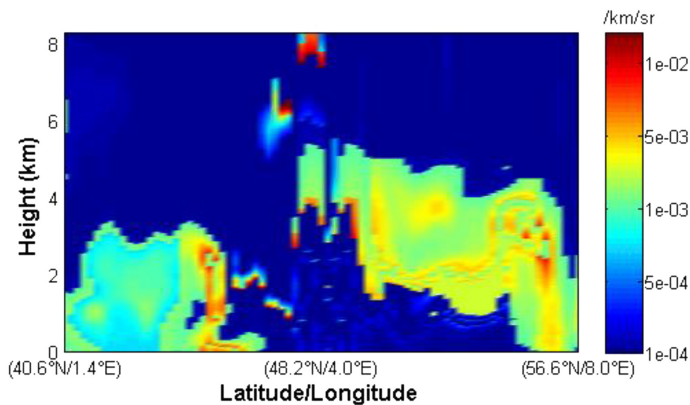


Figure 3. The total attenuated backscatter ($\text{km}^{-1} \text{sr}^{-1}$) at 532 nm from **(a)** the CALIPSO product and **(b)** the Meso-NH simulation for the overpass at 02:00–02:06 UTC on 29 May 2008. The CALIPSO overpass on 29 May 2008 is indicated by the gray line on Fig. 1b.

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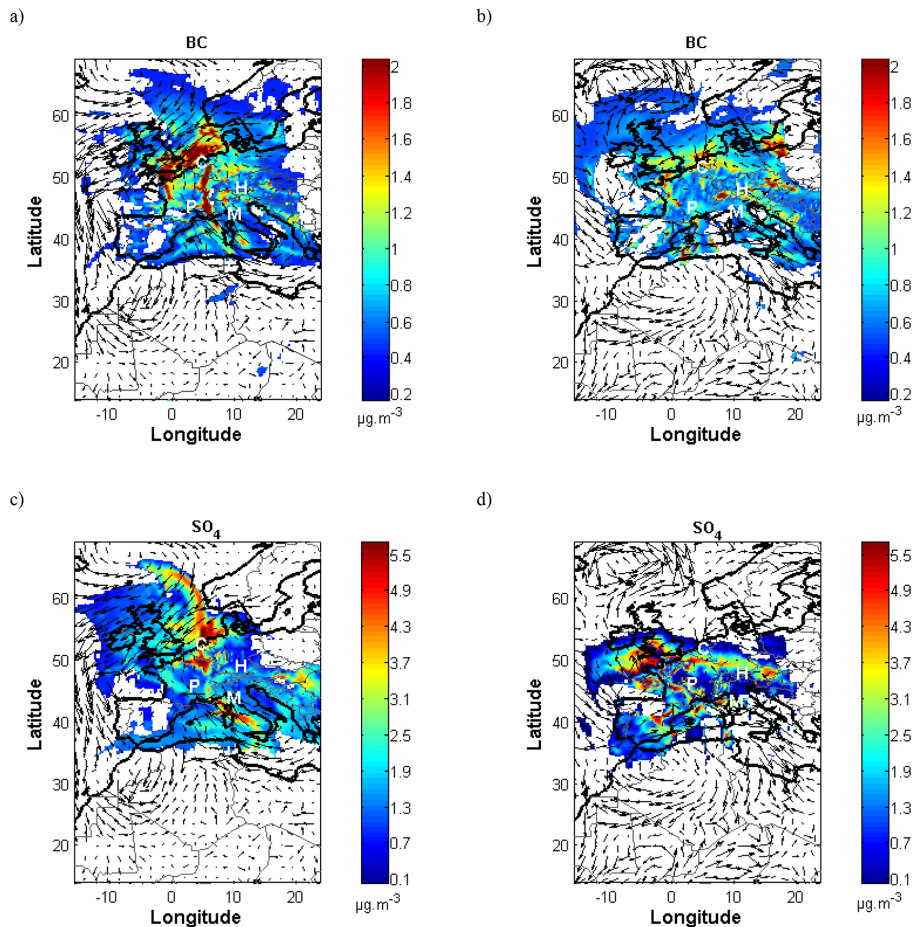
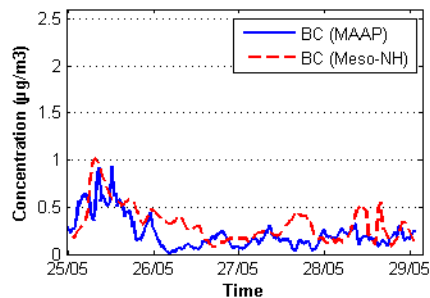


Figure 4. BC (a, b) and sulfate (c, d) mass concentration simulated by Meso-NH at 12:00 UTC on 28 May (a, c) and 29 May (b, d) 2008. The locations of the Cabauw, Puy de Dôme, Hohenpeissenberg and Mont-Cimone sites are indicated by C, P, H and M respectively.

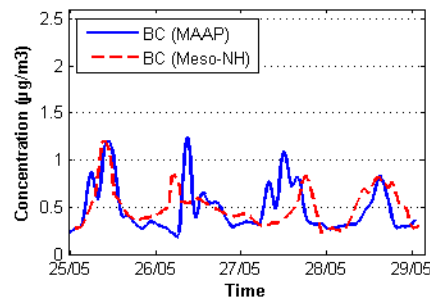
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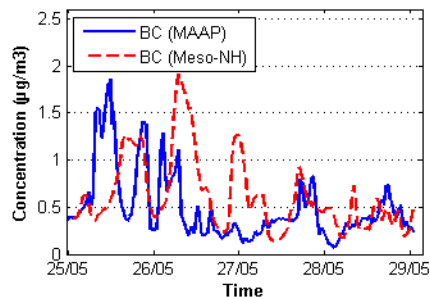
a) Mont Cimone



b) Puy-de-Dôme



c) Hohenpeissenberg



d) Cabauw

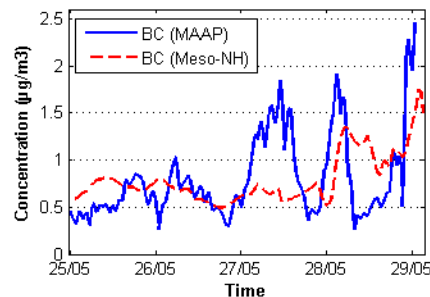


Figure 5. Evolution of the BC mass concentration ($\mu\text{g m}^{-3}$) simulated (red dashed line) and measured (blue solid line) between 25 and 31 May 2008.

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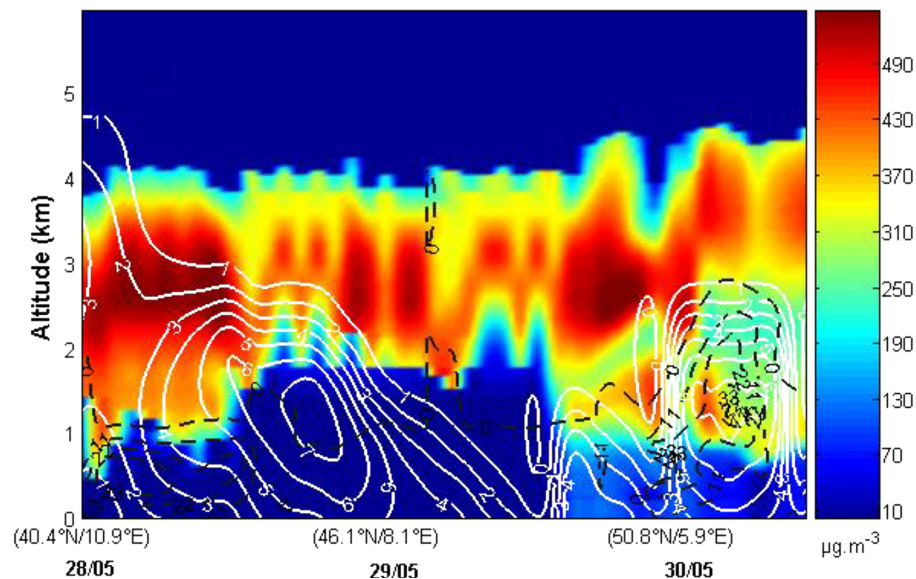


Figure 6. Vertical cross section of dust mass concentration (shading) with carbonaceous component mass concentration (white line, BC and OC) and inorganic salts mass concentration (black dashed line) following the dust plume trajectory over Europe between (40.4° N, 10.9° E) and (56.6° N, 8.0° E) obtained from the simulation.

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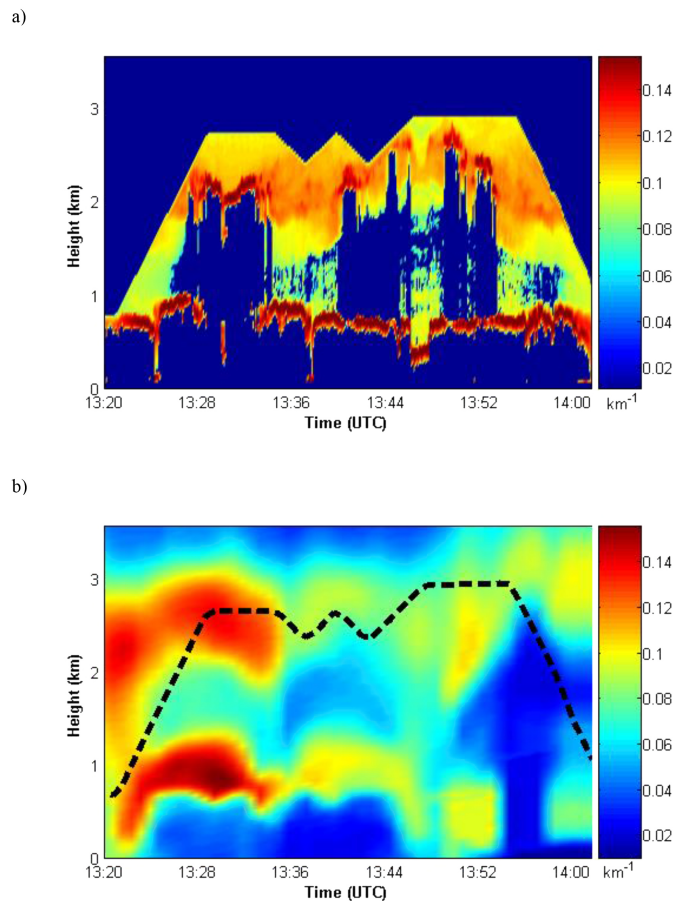


Figure 7. Vertical cross section of extinction coefficient (km^{-1}) **(a)** measured by LEANDRE and **(b)** simulated by Meso-NH around Cabauw between 13:28 and 14:01 UTC on 30 May 2008. The white line indicates the height of the aircraft.

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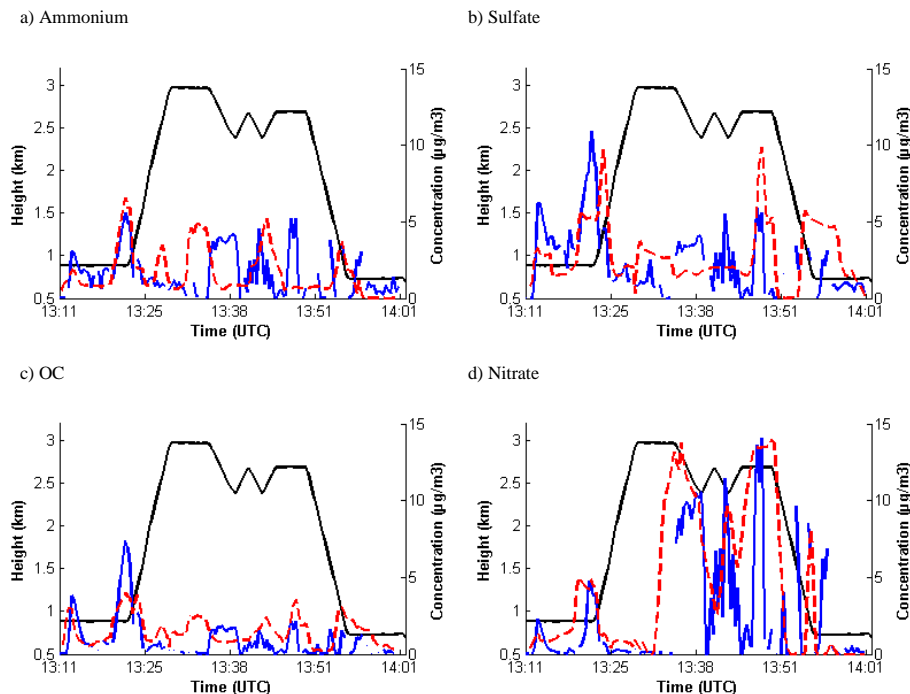


Figure 8. Evolution of (a) ammonium, (b) sulfate, (c) OC and (d) nitrate mass concentration ($\mu\text{g}\text{m}^{-3}$) obtained from AMS measurements (blue solid line) and simulated (red dashed line) around Cabauw between 13:11 and 14:01 UTC on 30 May 2008. The black line indicates the height of the aircraft.

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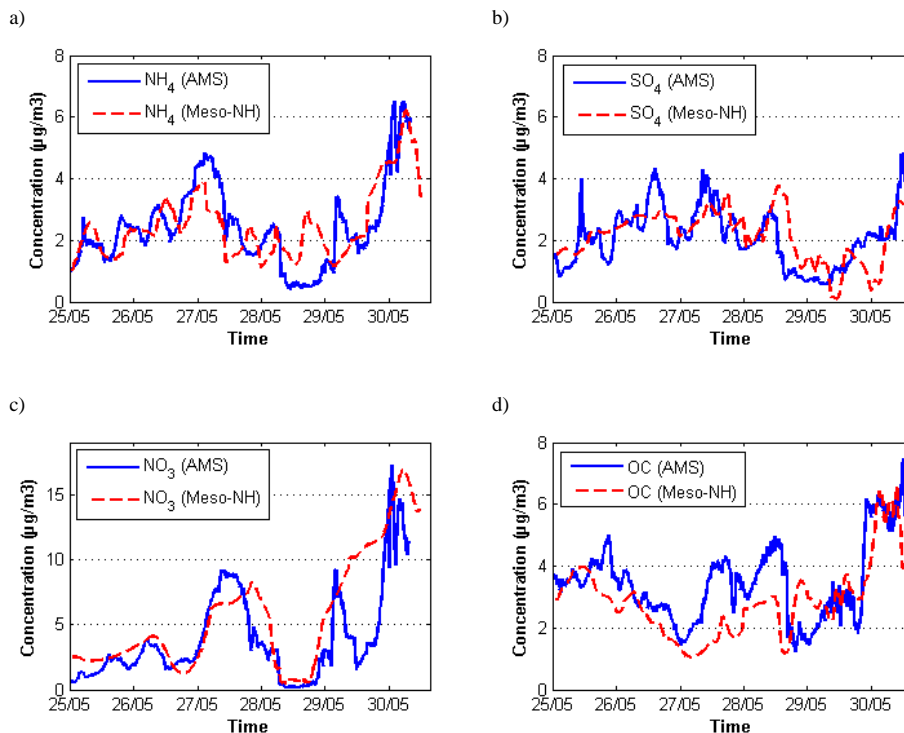


Figure 9. Evolution of (a) ammonium, (b) sulfate, (c) nitrate and (d) OC mass concentration ($\mu\text{g m}^{-3}$) obtained from AMS measurements (blue solid line) and simulated (red dashed line) over Cabauw between 25 and 30 May 2008.

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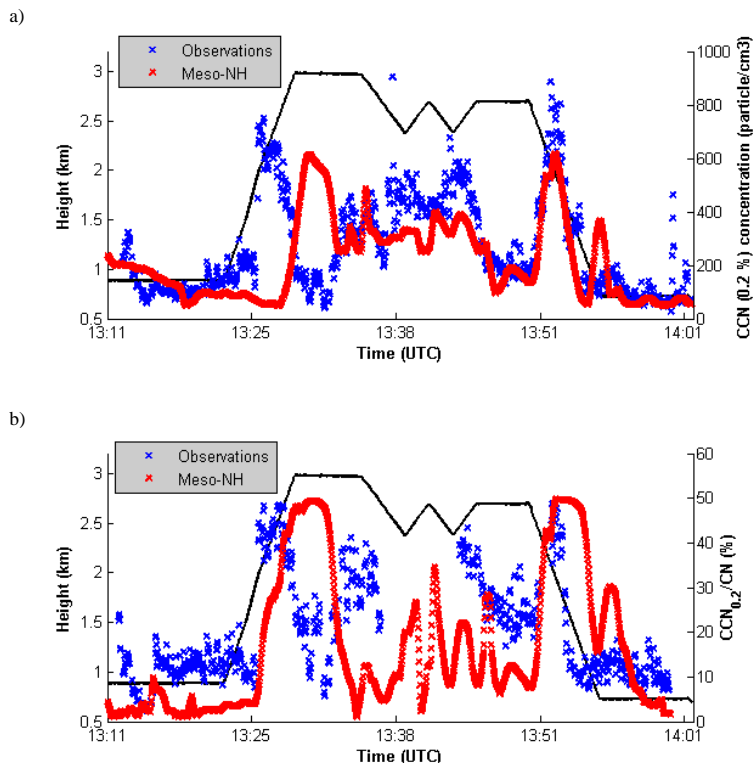


Figure 10. Evolution of (a) the CCN concentration and (b) the N_{CCN}/N_{CN} ratio at 0.2% supersaturation obtained from CCNC and CN concentration (blue line) and calculated from the simulation (red line) around Cabauw between 13:11 and 14:01 UTC on 30 May 2008. The black line indicates the height of the aircraft.

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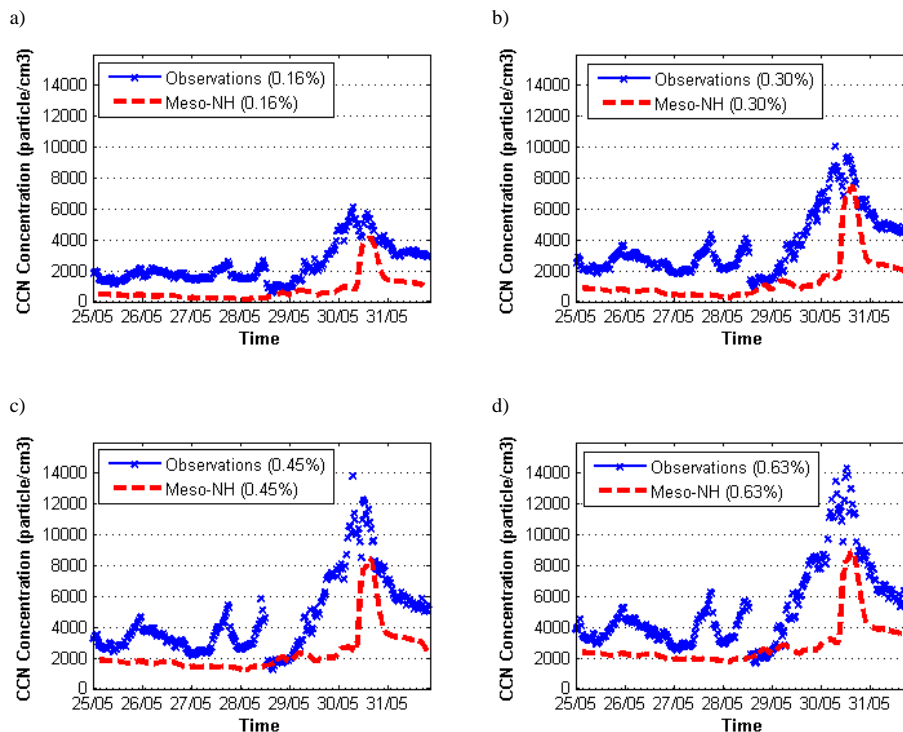


Figure 11. Evolution of the CCN concentration at water vapor supersaturation of **(a)** 0.16%, **(b)** 0.30%, **(c)** 0.45% and **(d)** 0.63% measured (blue line) and calculated from the simulation (red line) at Cabauw between 25 and 30 May 2008.