An overview from hygroscopic aerosols to cloud droplets: The HygrA-CD campaign in the Athens basin

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HIGHLIGHTS

• First coordinated measurements over Athens to study the impact of aerosols on PBL cloud formation (HygrA-CD campaign).
• Under continental and Etesian wind flow the PBL height was quite deep (~2–2.5 km), while under Saharan wind flow conditions, the PBL height remained quite shallow (~1–1.2 km).
• The origin of the atmospheric air masses, the prevailing meteorological conditions and aerosol hygroscopicity play a major role on the cloud formation at the vicinity of the PBL.
• Upward aerosol fluxes determine the number of the activated CCN at the cloud base.

GRAPHICAL ABSTRACT

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The international experimental campaign Hygroscopic Aerosols to Cloud Droplets (HygrA-CD), organized in the Greater Athens Area (GAA), Greece from 15 May to 22 June 2014, aimed to study the physico-chemical properties of aerosols and their impact on the formation of clouds in the convective Planetary Boundary Layer (PBL). We found that under continental (W-NW-N) and Etesian (NE) synoptic wind flow and with a deep moist PBL (~2–2.5 km height), mixed hygroscopic (anthropogenic, biomass burning and marine) particles arrive over the GAA, and contribute to the formation of convective non-prefecting PBL clouds (with droplets of ~16–20 µm mean diameter) with vertical extent up to 500 m. Under these conditions, high updraft velocities (~1–2 m s⁻¹) and cloud condensation nuclei (CCN) concentrations (~2000 cm⁻³ at 1% supersaturation), generated clouds with an estimated cloud droplet number of ~600 cm⁻³. Under Saharan wind flow conditions (S-SW) a shallow PBL (<1–1.2 km height) develops, leading to much higher CCN concentrations (~3500–5000 cm⁻³ at 1% supersaturation) near the ground; updraft velocities, however, were significantly lower, with an estimated maximum cloud droplet number of ~200 cm⁻³ and without observed significant PBL cloud formation. The largest contribution to cloud droplet number variance is attributed to the updraft velocity variability, followed by variances in aerosol number concentration.

1. Introduction

The interaction of aerosols with clouds, radiation and the hydrological cycle, referred to as “aerosol indirect climatic effects,” constitutes the largest source of uncertainty in assessments of climate sensitivity and anthropogenic climate change (Bony et al., 2013; Boucher et al., 2013; IPCC, 2013). This uncertainty stems from the highly coupled interactions between particles, clouds, dynamics and radiation across many scales. Limited understanding of these interactions further hinders their correct implementation in models, leaving open the possibility that the actual uncertainty is even larger than thought (Seinfeld et al., 2016).

Cloud condensation nuclei (CCN) are the seeds upon which cloud droplets form, and modulations in their concentration can have considerable influence on cloud formation and properties. Increasing concentrations of CCN in low-level warm clouds result in higher droplet concentrations and smaller droplet sizes relative to pristine clouds (Twomey, 1977). These microphysical changes increase shortwave albedo and long-wave emissivity (e.g., Boucher et al., 2013) and are less likely to form precipitation-size drops (Albrecht, 1989). Increased aerosol loads may increase the turbulence on the sides of clouds and reduce cloudiness by lateral mixing of cloudy with dry air (Xue and Feingold, 2006). These changes also delay the onset of freezing and decrease riming efficiency (Lohmann and Feichter, 2005; Lance et al., 2013). Enhanced aerosol concentration may also increase the lifetime and vertical extent of clouds with important feedbacks on precipitation efficiency and the hydrological cycle (e.g., Rosenfeld et al., 2008).

The ability of particles to act as CCN depends on their behavior when exposed to supersaturated water vapor, and is described by Köhler theory (Köhler, 1936) which considers the effects of particle curvature (Kelin effect) and soluble material (Raout effect) on the equilibrium water vapor pressure ($P_{eq}$) above the particle. According to Köhler theory, as long as the Raout effect ensures that $P_{eq}$ increases with water condensation, the aerosol remains in stable equilibrium with water vapor — even under supersaturated conditions (i.e. relative humidity above 100%). However, if the supersaturation exceeds a characteristic (or “critical”) value, $P_{eq}$ is dominated by the Kelvin effect, where it decreases with water condensation. Because of this, wet aerosol particle is incapable of reaching stable equilibrium with its environment (Köhler, 1936; Seinfeld and Pandis, 2006). Beyond this critical point, the particle experiences unconstrained growth and acts as a CCN that forms a cloud droplet.

The amount and nature of soluble material within particles strongly influences their ability to act as CCN; a larger amount of solute leads to an increase in CCN efficiency (i.e., lower critical supersaturation). In the absence of soluble material, adsorption of water vapour may also allow insoluble particles (biological material, mineral dust and volcanic ash) to become CCN active (Kumar et al., 2009, 2011a, 2011b; Lathem et al., 2011; Hatch et al., 2014; Laaksonen et al., 2016). The process may affect clouds in regions influenced by dust plumes (Karydis et al., 2011; Bangert et al., 2012; Bégue et al., 2015).

Hygroscopic Aerosols to Cloud Droplets (HygrA-CD) campaign aimed to study the aerosol-cloud droplet link for convective summertime planetary boundary layer (PBL) clouds in the Greater Athens Area (GAA). HygrA-CD was part of the Initial Training in Atmospheric Remote Sensing (ITARS) project (www.itars.net). The 6-week intensive observation period lasted between 15 May and 22 June 2014, which is late spring/early summer in the Eastern Mediterranean. During this season, PBL convection is generally favored. This condition, combined with sufficient humidity aloft, enables the development of clouds and local precipitation (Kotroni et al., 2001). June and July are also characterized by persistent northern winds, known as Etesians (Poupkou et al., 2011; Tyrlis and Lelieveld, 2013) that deliver high concentrations of aged aerosol particles from Eurasia throughout the region. This diversity of meteorological regimes and aerosol conditions provided the opportunity for a considerable range of relevant cloud formation conditions to be sampled.

2. HygrA-CD scientific objectives

HygrA-CD aimed to understand, i) the role of aerosols and meteorological conditions in cloud formation and their influence on cloud vertical extent, ii) the structure of the PBL in the GAA, and iii) the impact of aerosol-cloud interactions on the radiation balance.

The above goals were achieved by characterizing:

- aerosol properties (physical, optical, and chemical) both at ground and aloft in the lower troposphere, over the GAA,
- CCN number concentration at cloud-relevant water vapour supersaturation levels,
- tropospheric water vapour content and its variability over space and time,
- cloud dynamics, low-cloud variability related to atmospheric state, and
- properties of non-prefecting clouds,
- GAA basin flow and PBL structure through both observation (using lidars) and simulation (with a state-of-the-art weather forecast model).
A schematic of the main processes linking aerosol to cloud formation in the PBL is presented in Fig. 1. Aerosols activated as CCN are transported upwards in a convective PBL, from ground to near the top of the PBL, where they form clouds over the lifting condensation level (LCL). Fig. 2 shows a conceptual view of the mechanisms leading to cloud development as studied in the frame of HygrA-CD and main instrumentation/models involved (in blue).

3. Site location, instrumentation and modeling

3.1. Site location

The GAA is located in the Attica peninsula (around 37°58' N and 23°43' E) and hosts a large urban agglomeration of about 3.8 million inhabitants within about 415 km². GAA presents a very complex topography, with a major opening to the sea on the south-western part (Saronikos Gulf) and surrounded by four mountains: Egaleo (468 m) and Parnitha (1413 m) to the northwest, Penteli (1109 m) to the north and Hymettus (1026 m) to the east. The basin has a small opening inland to the northeast which allows a persistent northeast “Etesian” wind summertime flow pattern (May–August) to ventilate the basin. A synoptic description of the prevailing flow dynamics in the GAA is given by Melas et al. (1998).

During HygrA-CD, instruments were distributed over five sites within the GAA (shown in Fig. 3). Most of the instrumentation, however, was concentrated at the National Technical University of Athens (NTUA) (37.97° N, 23.79° E, 212 m a.s.l.) and the National Center for Scientific Research-Demokritos (DEM) (37.99° N, 23.82° E, 275 m a.s.l.) sites. Several other instruments were deployed at the Biomedical Research Foundation of the Academy of Athens (BRFAA) (37.93° N, 23.80° E, 130 m a.s.l.), the National Observatory of Athens (NOA) (38.06° N, 23.86° E, 495 m a.s.l.), the National and Kapodistrian University of Athens (NKUA) (37.98° N, 23.73° E, 280 m a.s.l.) and the Hellenic National Meteorological Service (HNMS) (37.88° N, 23.73° E, 10 m a.s.l.).

3.2. Instrumentation

A suite of in-situ and active/passive remote sensing (Fig. 2) instruments was deployed in the GAA during HygrA-CD. A complete list, together with detailed information on the physical properties measured, location, spatio-temporal resolution, operation period and relevant references for each instrument, is provided in Table 1.

Ground level aerosol instruments include:

- a carbon aerosol particulate device to measure the elemental/organic carbon (EC/OC),
- a scanning mobility particle size spectrometer (SMPS) and optical particle counters (OPC) to provide the aerosol size distribution and refractive index,
- particulate matter (PM₁₀, PM₂.₅, PM₁₀) samplers to provide the aerosol mass concentration,
- a nephelometer to provide the aerosol total scattering and backscattering coefficient,
- aethalometers to provide the equivalent black carbon (EBC) concentration,
- an ion concentration (IC) analyzer using filters to provide the water soluble ions in aerosol samples,
- a continuous stream-wise thermal-gradient CCN chamber (CFSTGC) to measure the CCN concentrations at different levels of supersaturation.

To retrieve the vertical profiles of the aerosol optical [extinction (aₐer) and backscatter (bₐer) coefficients, particle linear depolarization ratio (b₉₀°)] and micro-physical [number concentration (NC), effective radius (reff), refractive index (RI), single scattering albedo (SSA)] properties aloft (Müller et al., 2001), two synergetic lidar systems were used: the EOLE Raman lidar and the AIAS elastic backscatter lidar system. Moreover, a ceilometer provided the location and geometrical properties of clouds and the aerosol mixing height over the GAA, under specific conditions (Papayannis et al. in preparation-b) and taking into account various limitations, discussed by Wiegner et al. (2014).

The integrated aerosol parameters [aerosol optical depth (AOD), aerosol Ångström exponent (AE), aerosol optical properties (τₐeff, RI, SSA)] were retrieved from an AERONET sun photometer. A Doppler lidar provided the 3-dimensional wind field (direction and speed) permitting retrieval of the dynamic state of the atmosphere (e.g. turbulent dissipation rate) in the first 1–2 km above ground level.

A microwave radiometer (MWR) was used to provide vertical profiles of atmospheric temperature and humidity and related parameters. A Doppler polarimetric X-band radar was used to derive the vertical cross sections and profiles of the size distribution of large particles (including rain droplets) along with the radial wind velocity (Section 3.2.2). Radiosondes were also launched to obtain the vertical profiles of parameters that characterize the atmospheric thermodynamic state (pressure, temperature, humidity), static stability in the low troposphere (potential temperature, convective available potential energy, Richardson number, etc.), and the PBL height (PBLH).

3.2.1. In situ measurements

At the DEM station (Triantafyllou et al., 2016) (Fig. 3) the SMPS (SMPS 3936 model, TSI Inc.) was used to obtain the aerosol size distribution (10 to 550 nm electrical mobility diameter) with a time resolution of 5 min. The instrument has been calibrated against a reference instrument in Leipzig and participated in the European Center for Aerosol Calibration (ECAC) Instrument/Laboratory Intercomparison in June 2016 (Wiedensohler et al., 2012) showing a counting accuracy within 10% for the size range 30–550 nm under controlled laboratory conditions. An OPC (OPC 107 Dust Spectrometer model, Grimm GmbH) was used to acquire the particle size distribution in the range of 0.25–2.5 μm (optical diameter), with a time resolution of 5 min. According to Heim et al. (2008), for sizes larger than 0.8 μm (electrical mobility diameter), the OPC sizing accuracy decreases, while its counting accuracy is within 10% of the ideal 100% for sizes up to 1 μm. Furthermore, in situ aerosol sampling was deployed at the DEM station; a combination of SMPS and OPC size distribution data in the overlapping size provided the
diurnal variation of the total volume of aerosol size distribution, up to 1 μm (Vratolis et al., in preparation). A thermal-optical EC/OC analyzer (Lab OC-EC Aerosol Analyzer model, Sunset Laboratory Inc.) was used to measure the concentrations of organic carbon (OC) and elemental carbon (EC) in the size ranges up to 2.5 μm (aerodynamic diameter) with a time resolution of 3 h (Amiridis et al., 2012). The EC measurements repeatability and reproducibility relative standard deviations were 15 and 20%, respectively (Panteliadis et al., 2015).

A 3-wavelength (450–525–635 nm) nephelometer (Aurora model, Ecotech Pty Ltd.) was used to provide the aerosol scattering and backscatter coefficients, while two 7-wavelength (370–470–520–590–660–880–950 nm) aethalometers (AE-31 and AE-33 models, Magee Scientific Corp.) were used to provide the aerosol absorption coefficients, for PM_{10} and PM_{2.5} aerosols, respectively. The A31 and A33 aethalometers were also used to provide the EBC concentration. The single scattering albedo exponent was derived for 470 to 660 nm wavelengths from the nephelometer (PM_{10} inlet) and A33 aethalometer (PM_{2.5} inlet) data.

The CFSTGC (Continuous Flow Streamwise Thermal Gradient CCN Counter; Robert and Nenes, 2005) is constructed by Droplet Measurement Technologies (model CCN-100), was used to measure the CCN concentrations (available from 18 to 22 June) at 0.2–1% supersaturation. Measurements were taken for about 10 min at each supersaturation level, yielding a CCN spectrum consisting of 5 different supersaturations.
The sampling point was fully exposed to the top of the School of Mining and Metallurgical Engineering building. The following meteorological variables were measured 15 m above ground level at NTUA using a meteorological station (Vantage Pro 2 model, Vaisala Oyj) equipped with a GPS receiver, temperature, humidity, and pressure sensors, and also provided the water vapour mixing ratio profile (nearby sunphotometer), we retrieve only the $b_{aer}$ and the AE-related to extinction and backscatter coefficients. During nighttime the vertical profiles of $b_{aer}$, $a_{soa}$, $S$, and AE-related to extinction and backscatter coefficients are retrieved with 10–20%, 10–15%, 10% and 25% uncertainty, respectively (Kokkalis et al., 2012). During daytime, using as input a constant $S$ value (constrained by the mean AOD value obtained from a nearby sunphotometer), we retrieve only the $b_{aer}$ and the AE-related to backscatter coefficient values with an average uncertainty (due to both statistical and systematic errors) of 20–30 and 25%, respectively. EOEL also provided the water vapour mixing ratio profiles from 0.5 to 6 km height, during nighttime, with a statistical error $<5%$ at heights up to 2 km and $<10%$ from 2.5 to 6 km (Manouris et al., 2007). Although full overlap of EOEL occurs at 600–700 m above ground level, an experimental method has been applied (Wandinger and Ansmann, 2002) to derive the overlap correction vertical profile down to about 400 m.

The NTUA mobile aerosol backscatter depolarization lidar (AIAAS) (LR111-D200 model, Raymetrics SA) was operated at the DEM site (Fig. 3), only during daytime (due to personnel availability) 37 days out of 39. It is a truck-mounted system with two polarizations at 355 nm: parallel (p-parallel) and vertical (s-cross) (Papayannis et al., 2012). The full overlap of the system is obtained at about 350 m above ground level (Bougiaioti et al., 2009).
ground level. AIAS provides the vertical profile of $\delta_p$ in the lower free troposphere, using a calibration technique based on the $\pm 45^\circ (\pm 0.2^\circ$ uncertainty) methodology described by Freudenthaler et al. (2006) and Freudenthaler (2016). The relative uncertainty in the retrieved $\delta_p$ is about 15%.

A University of Leicester (UK) (emitting at 905 nm) laser ceilometer (CS135 model, Campbell Scientific Inc.) was deployed at the DEM site (Fig. 3). Every 30 s the instrument reported cloud base and aerosol mixing layer height, with aerosol layer extent determined by a gradient method with 30 min averaging (Vande Hey et al., 2014), accounting for other instrumental effects, but not for water vapour absorption (Vande Hey, 2015; Wiegner and Gasteiger, 2015). The range resolution of the instrument is 5 m. Full overlap of the system is of the order of 300 m above ground level (Vande Hey et al., 2014). This overlap is 50–100 m lower than the EOLE lidar, therefore combining both lidar profiles allows EOLE aerosol optical retrievals to be extended to lower altitudes (Wiegner et al., 2014).

A pulsed Doppler scanning lidar system (StreamLine Wind Pro model, HALO Photonics) at 1.5 μm was deployed at the DEM site (Fig. 3) by the Finnish Meteorological Institute (FMI). The system was operated in the vertical azimuth display (VAD) mode and the 3-beam Doppler beam swinging (DBS) mode. It mainly provided the vertical profiles of the radial wind (Henderson et al., 2005) and 2–3D wind fields (Wood et al., 2013), as well as the atmospheric turbulent properties (e.g. turbulent dissipation rate) (O’Connor et al., 2010). The wind velocity is provided with accuracy better than 0.1 and 0.5 m s$^{-1}$ for VAD and DBS mode, respectively. The range resolution of the measurements is 30 m, and the temporal resolution is 14 s and 15 min for DBS and VAD modes, respectively; the maximum range achieved is 2–3 km (or even 10 km height, under the presence of clouds) depending on the atmospheric aerosol load.

A microwave radiometer (RPG–HATPRO model, RPG Radiometer Physics), operated at NTUA by the INOE 2000 (Fig. 3), was used to detect the microwave radiation emitted by the atmosphere at several channels (22.2–31.4 GHz and 51.3–59 GHz) to provide temperature and absolute and relative humidity (RH) vertical profiles. The root-mean-square (rms) accuracy of temperature was $\pm 0.6$ K near the surface, increasing to $1.5–2.0$ K in the middle troposphere (Crewell et al., 2001; Lijieger et al., 2005). The rms of absolute humidity was 0.4 g m$^{-3}$. The integrated water vapour (IWV) and the liquid water path (LWP) retrievals had accuracies of 0.3–1.0 kg m$^{-1}$ and 20–30 g m$^{-2}$, respectively (Loehnert and Crewell, 2003).

The X-band (9.37 GHz) Doppler polarimetric radar (XPol) (Wurman et al., 1997) was operated by NOA, at an altitude of 510 m and a distance of 7 km from the DEM site (Fig. 3). It performed range height indicator scans from 0° to 60° elevation over DEM every 1 min (100 averaged signals with signal to noise ratio > 0 dB) to provide the wind radial velocity and spectral width (typical random standard error of 0.3 m s$^{-1}$), horizontal and differential reflectivity (calibration errors of 0.5 and 0.2 dB, respectively), differential phase shift (3° error) and co-polar correlation (1% error), with a range resolution of 120 m (at a maximum range of 24 km) and a vertical range resolution of 12–25 m (from 500 to 12,500 m in height). Reflectivity calibration is made typically in each experimental campaign using disdrometer data as a reference if available.

Retrievals also provided the rainfall rate (from polarimetric rainfall estimation algorithms), and particle size distribution parameters (median volume diameter and number density) for particle diameters $>0.3$ mm from horizontal differential reflectivity (Bringi and Chandrasekar, 2001; Kalogiros et al., 2012), as well as the turbulent kinetic energy dissipation rate within the radar volume ($1^\circ$ beam width) based on Doviak and Zrnic (1993).

An 8-wavelength (340, 380, 440, 500, 675, 870, 1020 and 1640 nm) sun photometer (CE-318-NEDPS9 model, Cimel SA), operated by NOA within NASA’s AERONET project (Aerosol Robotic Network) (Holben et al., 1998) was operated at the BRFAA site (Fig. 3). It was used to derive the integrated AOD (uncertainty $\pm 0.01–0.02$) (Smirnov et al., 2000), and the relevant integrated aerosol microphysical properties (Dubovik et al., 2006, 2011), such as: volume particle size distribution (with 15–35% uncertainty) in 22 size bins, fractional volume of non-spherical particles, complex RI (real part uncertainty 0.025–0.050 and imaginary part uncertainty of 0.03), SSA (uncertainty $\pm 0.03$ for AOD $>0.2$), and AE at the 440–870 nm wavelength pair (uncertainty of 0.03–0.04) (Schuster et al., 2006).

### 3.3 Modeling

The regional Weather Research Forecast (WRF) model (Skamarock et al., 2005) was used to forecast the meteorological parameters (temperature, relative humidity and wind speed) in the lower troposphere and also the PBLH (Banks et al., 2016). The model was configured with three domains of varying horizontal grid spacing: the parent European level ($12 \times 12$ km), and two one-way nested domains for Greece ($4 \times 4$ km) and GAA ($1 \times 1$ km). It is assumed that $1 \times 1$ km spatial resolution is sufficient to resolve most local features and cloud relevant parameters (e.g. surface sensible heat flux) in the complex terrain of Athens (Banks et al., 2016). The FLEXPART–WRF (Brioude et al., 2013) and HYSSPLIT (Draxler and Rolph; Stein et al., 2015) dispersion models are used to estimate the origin of the air masses arriving at the GAA during the campaign period.

FLEXPART data was provided on a daily basis during the experimental period in the form of back-trajectory maps and emission sensitivity maps to assist decision making of the measuring activities for the next two days. Driving the dispersion simulations with high resolution WRF meteorology allows a more detailed representation of the sources and transport of particles during the campaign. HYSSPLIT was mainly used for the clustering of the long-range transport back-trajectories.

### 4. Prevailing meteorological conditions

A cluster analysis of backward HYSSPLIT air mass trajectories, using the clustering technique provided by Fleming et al. (2012), allows for understanding the origin of air masses arriving in the GAA. These runs are performed for a 96-hour backwards period starting at 12:00 UTC of each campaign date. The arrival heights over Athens are 0.5, 1, 2, 3 and 4 km. Results of the cluster analysis are presented in Fig. 4 (upper left panel) and indicates the prevalence of three synoptic flow patterns during the campaign; namely continental (72.3%), Etesians (14.4%) and Saharan (13.3%). Analysis of the trajectories for each individual arrival height indicates similar characteristics for 0.5 and 1 km (Fig. 4, upper right panel) (78 trajectories), 2 and 3 km (Fig. 4, lower left panel) (78 trajectories) and 4 km (Fig. 4, lower right panel) (39 trajectories). Air masses arriving inside the PBL (0.5 and 1 km) are mainly continental and include a strong NE component that may imply biomass smoke transport at these heights (Diapoulis et al., 2014). The 2 and 3 km trajectories include a Saharan component mainly from deserts of Morocco and Algeria. This Saharan component is also evident at the 4 km clusters implying that desert dust is mainly transported above the Athens PBL.

During continental type flow Athens is influenced by W to NW winds. These days are characterized by a stagnant weather pattern with a weak atmospheric pressure gradient over Greece. This type of atmospheric situation can provoke the development of urban pollution episodes (Ziomas, 1998).

Etesian winds are mainly from N-NE directions and are caused by a gradient between strong high pressure NW of Greece and a low pressure area over Asia (Tyrlis and Lelieveld, 2013). Dynamic forcing by Etesian winds are mainly from N-NE directions and are caused by a gradient between strong high pressure NW of Greece and a low pressure area over Asia (Tyrlis and Lelieveld, 2013). Dynamic forcing by Etesians also leads to increased PBLH over Athens. The weakening of the Etesians synoptic flow (Poupkou et al., 2011) allows the development of local circulation systems (sea and land breezes) and a decreased PBLH (e.g. Melas et al., 1995, 1998) in accordance with Banks et al. (2016).
During Saharan-type flow, the winds flow predominantly from the SW. Typically this wind flow is associated with dust layers that are located above the PBL (Papayannis et al., 2009). The days corresponding to each typical synoptic meteorological condition (N-NE Etesian, W-NW Continental, and S-SW Saharan) are shown in Table 2 (only daytime measurements are considered). The Cimel-derived integrated AOD values (at 1640 nm) \(>0.1\) and the cases of cloud formation at the top of the PBL, along with the cloud vertical extent for these cases, are also indicated in Table 2. An interesting finding emerging from this table is that PBL clouds are formed in most of the cases under N-NE and N-NW synoptic flow, while under S-SW flow (mostly associated with medium to low Saharan dust transfer over Athens) PBL clouds were inhibited. The cloud vertical extent near the top of the PBL varied between 50 and 500 m.

In addition to the trajectory and AOD analysis, wind field profile measurements were made at the DEM site using the HALO wind lidar. Fig. 5 shows the diurnal evolution of the mean horizontal wind speed (upper plot) and direction (lower plot) within the PBLH for the period 11 May to 15 June. During most of the studied period, the mean wind speed ranged between 6 and 10 m s\(^{-1}\), with maximum values around 20–22 m s\(^{-1}\) on 14–15 and 29 May. These strong horizontal winds are associated with north-eastern (Etesian) synoptic flow, as shown by the wind direction measurements (cf. Fig. 5). The days that present diurnal variation of wind direction inside the PBL (e.g. 5 and 6 June) are typical days with sea breeze initiation during the afternoon hours. This is consistent with the results provided by WRF simulations (Banks et al., 2016).

### 5. HygrA-CD highlights — major findings

In this section we focus on major highlights and findings to date from HygrA-CD, which are detailed in a series of separate papers (see Section 6). To study the cloud microphysical and geometrical properties in relation to PBL top cloud formation, several data were combined. Thus, the lifting condensation level height derived from radiosonde data was considered as a key value for the cloud vertical extent at the top of the PBL. Moreover, the temporal gradient of the liquid water path measured by the microwave radiometer was used for identifying changes in the cloud water content when vertical transport of aerosols into shallow convective cumuli occurred. To characterize these upward aerosol fluxes, the co-spectra of the vertical wind component measured by the Doppler lidar and of the baer values retrieved from the AIAS data were calculated (Engelmann et al., 2008; Ansmann et al., 2010).

During HygrA-CD the mean Cimel integrated AOD values (Fig. 6) were of the order of 0.15 (at 500 nm), generally lower than the average AOD climatology (Gerasopoulos et al., 2011) for the area of Athens (e.g. 0.23 at 500 nm). This was due to the fact that no major dust cases were reported for the campaign period, aside from 20 and 24–29 May, where high AOD values (black and red lines) at 380 and 500 nm (0.35–0.40 and 0.25–0.30, respectively) and low integrated AE mean values (Fig.
6, green lines) at 440–870 nm (0.38–0.80) indicated the presence of moderate dust intrusion events over Athens. Finally, integrated SSA values, ranging from 0.85 to 1.00, showed normal to low aerosol absorption during the whole campaign period (Fig. 6, blue lines).

Regarding the PM$_{10}$ and PM$_{2.5}$ concentrations, the legislated maximum 24-hour average value for PM$_{2.5}$ (20 μg m$^{-3}$) was exceeded 3 times (on 23 May, 6 and 7 June) due to long-range transported fine particles from different sectors at different altitudes on 23 May and the north sector on 6 and 7 June. The PM$_{10}$ concentrations exceeded the legislated 24-hour average limit (40 μg m$^{-3}$) at 440 nm (0.38–0.80) indicated the presence of moderate dust intrusion events over Athens. Finally, integrated SSA values, ranging from 0.85 to 1.00, showed normal to low aerosol absorption during the whole campaign period (Fig. 6, blue lines).

Regarding the PM$_{10}$ and PM$_{2.5}$ concentrations, the legislated maximum 24-hour average value for PM$_{2.5}$ (20 μg m$^{-3}$) was exceeded 3 times (on 23 May, 6 and 7 June) due to long-range transported fine particles from different sectors at different altitudes on 23 May and the north sector on 6 and 7 June. The PM$_{10}$ concentrations exceeded the legislated 24-hour average limit (40 μg m$^{-3}$) twice (on 29 May and 6 June), with the first corresponding to a Saharan dust transport event and the second to long-range transported particulate matter from the north sector. From the concentrations of water soluble ionic species: Cl$^{-}$, NO$_3^-$, SO$_4^{2-}$, Na$^+$, K$^+$, NH$_4^+$, Ca$^{2+}$, Mg$^{2+}$, in PM$_{10}$ and PM$_{2.5}$ sampled at NTUA, on specific days, from 21 May to 7 June, it is clear that hygroscopic particles contained high concentrations of NO$_3^-$, NH$_4^+$ (1660–2430 ng m$^{-3}$ at PM$_{2.5}$), SO$_4^{2-}$ (3800–5280 ng m$^{-3}$ at PM$_{2.5}$), Na$^+$ (840–1140 ng m$^{-3}$ at PM$_{10}$), and Cl$^-$ (355–1290 ng m$^{-3}$ at PM$_{10}$) indicating predominantly anthropogenic (fine particles from N-NE directions) or marine (coarse particles from N-NE or S-SW directions) origin. These results are consistent with previous measurements in the area (Theodosi et al., 2011; Triantafyllou et al., 2016), and can therefore be considered representative of the region and season.

Moreover, Fig. 7 presents the mean relative contribution of the main inorganic ionic species in PM$_{10}$ (left panel) compared to that in PM$_{2.5}$ (right data), showing a considerably lower percentage of SO$_4^{2-}$ and NH$_4^+$ in the coarse fraction. The Ca$^{2+}$, Na$^+$ and Cl$^-$ percentages are much higher in PM$_{10}$ than in PM$_{2.5}$, owing to the presence of dust and sea salt in the coarse fraction. SO$_4^{2-}$, NH$_4^+$ and NO$_3^-$ represent about 90% of the total mass of inorganic ions measured in PM$_{2.5}$. The SO$_4^{2-}$ percentage is in accordance with values reported for the fine fraction in the Eastern Mediterranean region including urban and urban background (e.g. Theodosi et al., 2011; Mantas et al., 2014). It is known that sulfate levels above Greece are also heavily influenced by long-range transport and processes evolving at a large spatial scale (Koulouri et al., 2008; Theodosi et al., 2011).

Regarding the total volume of aerosol size distribution up to 1 μm as derived from SMPS and OPC data at the DEM site (Fig. 8), it seems that maximum values were reached during the late evening hours (~180 μm$^{-3}$ cm$^{-3}$) due to possible accumulation of small particles (with diameters of ~50–60 nm) of anthropogenic origin (e.g. car traffic and other anthropogenic activities) under a shallow PBL (<1–1.2 km height). Aerosol backscatter values retrieved from lidar and the nephelometer showed a good agreement with each other and indicated similar late evening peak (Fettatzis et al., 2015).

In Fig. 9 we present the time series of the hourly averaged values of OC, EC and EBC. The EBC and EC concentrations covaried throughout the measurement period. OC exhibited low concentrations during May, when the transported air masses reaching the DEM station originate from SW-NW and higher concentrations later in the campaign when the Etesians dominated. This indicates regional transport of polluted air masses from the NE. Our findings showed that the GAA exhibited

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**Table 2**

<table>
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<th>Date</th>
<th>Cloud formation</th>
<th>Cloud vertical extent (m)</th>
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<td>1.88 ± 0.15</td>
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</table>
similar EC values to Chicago and New York (USA), as well to Tokyo (Japan), but much lower values compared to those reported for other major European major metropolitan areas and megacities (Istanbul, London, Paris and Barcelona) (Beekmann et al., 2015).

Fig. 10 represents the time series of the CCN concentrations measured at the five different levels of supersaturation, along with concentration of NO2, which generally followed similar trends for the period from 18 to 22 June. Throughout the measurement period, CCN concentrations at each supersaturation level varied by up to a factor of five. CCN concentrations and CCN activity and hygroscopicity are modulated by atmospheric processing and aging. During days with a shallow (< 1–1.2 km height) and stable PBL (on 17–18 June) with poor ventilation aloft, CCN concentrations were highest (~3500–5000 cm\(^{-3}\) at 1% supersaturation). During those days, local pollution was dominant and CCN concentrations track the NO2 variability very well (cf. Fig. 10). During days with a higher PBLH (cf. Table 2 on 20–21 June), dispersion and vertical transport of elevated CCN concentrations can interact with the cloud-topped PBL. The presence of high CCN concentrations during those days (20–21 June) are likely associated with long-range-transport, as CCN are more “aged” and peak concentrations do not always follow the trend of locally emitted NO2.

Fig. 5. Diurnal evolution of the mean horizontal wind speed (in m s\(^{-1}\)) (upper plot) and direction (in °) (lower plot) within the PBL during the period 11 May to 15 June 2014, as obtained by the HALO wind lidar.

Fig. 6. Daily integrated mean values of the aerosol optical depth (AOD) at 380 and 500 nm (black and red, respectively), single scattering albedo (SSA) at 440 nm (blue) and the 440–870 nm Ångström exponent (AE) (green), all obtained from the Cimel sun photometer from 15 May to 14 June 2014.
CCN concentrations at the lowest supersaturation (0.2%) correspond mostly to processed aerosol, where particles are larger in size and contain significant amounts of soluble material (mostly inorganic salts) making them more efficient CCN (Petäjä et al., 2007). This is also reflected in their hygroscopicity values (κ varying between 0.33 and 0.45 for the 0.2% supersaturation), which are close to the proposed global average of 0.3 (Andreae and Rosenfeld, 2008; Pöschl et al., 2009). The activation diameter that corresponds to the 0.2% supersaturation (110–120 nm) is often near a peak in the number size distribution, thus there might be a significant fraction of the total particles that do not activate at the given diameter. This is further supported by the fact that at the lowest supersaturation the measured CCN particles account for around a quarter of the total particles. As supersaturation increases, smaller particles begin to activate and hygroscopicity diminishes. Smaller particles often consist mainly of organics (Bougiatioti et al., 2015) and the obtained κ values are close to typical values found for oxidized organics (e.g., Engelhart et al., 2008; Asa-Awuku et al., 2008, 2009). As expected, activation fraction increases with increasing supersaturation, and their diurnal variability exhibits maximum values during nighttime, possibly because of additional anthropogenic, less CCN-active sources during daytime and aged, processed particles, during nighttime (Bougiatioti et al., in preparation).

For each of the tropospheric aerosol layers (up to 5 km height) identified by lidar within HyGrA-CD, we performed an individual 9-day backward trajectory analysis for the base, center and top of the layer at the lidar measurement times in order to define the air mass origins. The number of fire hotspots, for nine days prior to each event, were given by Moderate Resolution Imaging Spectroradiometer (MODIS) (Giglio et al., 2010; Zhang et al., 2015) to confirm the presence of smoke particles that were identified by the lidar classification scheme (discussed below) over our site. The DREAM (Pérez et al., 2011) and WRF-FLEXPART models were also used to identify those cases where Saharan dust was advected above the measurement site.

The height region of the aerosol layer, in relation to the air mass dominant type (mixed dust, continental, mixed biomass burning, mixed Arctic-continental-biomass burning, mixed continental-marine, and Asian dust-biomass burning-continental), the S values at 355 and 532 nm, the S532nm/S355nm and the corresponding AE-related to extinction coefficient values based on Raman EOLE measurements are portrayed in Fig. 11a. (nighttime cloud-free days). The air mass classification was based on the air mass origin and on the respective values of S (355 and 532 nm), AE-related to extinction coefficient and δp using literature data (e.g. Groß et al., 2011, 2013; Wiegner et al., 2011; Mamouri et al., 2013; Papayannis et al., 2014; Mamouri and Ansmann, 2015; Giannakaki et al., 2016). The mixed BB air masses (shown in light green) dominate between 10 and 14 June (1–3 km height), while the mixed dust ones dominate on 20 and 26–27 May and 17–18 June (1–5 km).

Very occasionally, air masses originating from the Arctic region (traveling very fast, within a few days), exhibiting a lower lidar ratio (shown in yellow), were sampled at ~3 km height, just above the PBL on 14 and 16 May and 22 June. In Fig. 11b the height region of the aerosol layer is related to the air mass dominant type (upper panel, same as in Fig. 11a) and to the δp at 532 nm (lower panel), based on the AERI measurements (daytime cloud-free days). Here the dominant presence of mixed dust is marked by high δp values (20–30%), while all other mixed aerosols present δp values below 10%, in accordance with literature data (Groß et al., 2013).

Fig. 12a presents in a 3-dimensional plot the particle lidar ratios and the δp at 532 nm, along with the AE-related to extinction (355 nm/532 nm) for the aerosol types that were identified. Different aerosol types occupy different areas in this figure. Significantly larger linear particle depolarization ratios were found for the mixed Saharan dust aerosols. Larger S values were found for mixed biomass burning aerosols (in the range between 54 and 60 sr), as well as for the anthropogenic aerosols. The eventual contribution of Arctic aerosols to the mixture of biomass burning and anthropogenic aerosols makes S lower, with a mean value of 41 ± 6 sr. The mixture of marine with anthropogenic aerosols gives one of the largest AEs, possibly due to the dominance of the marine factor and associated smaller particle sizes. The observed large variability of AEs and S values for all aerosol types studied can be attributed to the dominant contribution of each air mass to the observed aerosol mixture.

In Fig. 12b we compare our findings on the relation between S and δp at 532 nm (full squares) with the results of other studies for characteristic aerosol types. Our results are generally in good agreement with the literature values. For example, the aerosol properties of the mixed...
anthropogenic and mixed marine aerosol type in this study were found to lie between the pure marine aerosols and pure anthropogenic aerosols on both axes, in compliance with the values reported by Gros et al. (2013). However, $S$ and $\delta_p$ values found for mixed Saharan dust particles in this study are at the lower limit of results previously published for this aerosol type (Gros et al., 2013, 2016), and mixed biomass burning aerosols sampled in HygrA-CD have similar intensive aerosol properties as anthropogenic aerosols reported in Gros et al. (2013).

Concerning the aerosol micro-physical properties (NC, $r_{\text{eff}}$, RI, SSA), they were derived for selected days within specific layers aloft, using lidar inversion techniques (Müller et al., 2001). Typical derived $r_{\text{eff}}$ values are in the range of 0.22–0.4 $\mu$m, while SSA (532 nm) varies between 0.80 and 0.95. More details can be found in Papayannis et al. (in preparation-a).

As for cloud properties, the sensitivity of our X-pol radar allowed the detection of relatively dense elevated non-precipitating clouds with a reflectivity of $-20$ dBZ or higher, up to a distance of 5 km from the radar, passing over the DEM station. The method proposed by Frisch et al. (1995) was used to estimate from radar reflectivity data the parameters of the clouds detected at a distance of 2 km. Cloud droplets are too small to use polarimetric radar variables like differential reflectivity, which has an accuracy limit of 0.1–0.2 dB and can be only used for droplets larger than 0.5 $\mu$m. Liquid water path estimations from the microwave radiometer data were not reliable; instead a constraint suggested by Wang (2013) was used, utilizing mean number density concentration of droplets ($N_d$ in $\text{cm}^{-3}$) and modal diameter ($D_o$ in $\mu$m), as follows: $N_d = 3604(2D_o)^{-1.11}$. The size distribution of cloud droplets was assumed to be a lognormal distribution with 0.4 logarithmic width. Similar results were obtained when a Gamma distribution was used with a shape parameter value of 4.5.

The results of this analysis (concerning average values of scarce sporadic clouds observed in the period from 6:00 to 18:00 UTC for each day in correspondence with the lidar daily average results shown in Fig. 11a and b), are shown in Fig. 13a and indicate generally clouds with small vertical extent (<50–500 m), low density and relatively large droplet diameters (mean values of 16–20 $\mu$m). The vertical extent of the cloud in each profile was estimated as the ratio of integrated liquid water path to mean liquid water content in cloud. The base of the clouds varied during the day in the range 1500 to 2500 m depending on the cloud type (stratus clouds or cumulus clouds at the rising top of the atmospheric boundary layer). It should be noted that the detection of non-precipitating clouds by the radar only occurred on days with biomass burning particles in the atmosphere (cf. Fig. 11a,b), even though the lidars detected clouds at heights of about 2 km or the top of the atmospheric boundary layer during most of the days of the campaign. The radar also detected some elevated clouds at the limit of its noise level for short time periods in many of those days. This implies that the non-precipitating clouds detected by the lidars in the rest of the days when the radar was operating
were characterized by smaller droplets. The radar was operating continuously from the afternoon of 26 May until 23 June, except during some time periods due to technical problems (cf. Fig. 13a, b).

The detected rain clouds and their average parameters estimated using polarimetric radar variables and the algorithm proposed by Kalogiros et al. (2012) are shown in Fig. 13b. The observed rain rates and median volume diameters of an assumed Gamma droplet size distribution (about 1.6 times the modal diameters of the corresponding lognormal droplet size distribution) were small, mostly in the range of drizzle values. Higher values were observed during the days with no non-precipitating cloud detection from the radar.

Based on the classification of air masses (Section 4) the dominant flows during the HygrA-CD were of continental origin. This flow (SW-W-NW) is characterized by relatively low total aerosol concentrations (~20 μg m⁻³) corresponding to an AOD <0.1 in the near-IR. Fine particles were the dominant mode of the total aerosol concentration (contribution of 65%). Also, low OC and EC concentrations were recorded. During continental flow, clouds were present at the top of the PBL, with a significant vertical development (300–500 m). Finally, moderate to high horizontal wind speeds within the PBL were observed, which were inversely proportional to the PBLH during local noon, with high wind speeds (>16 m s⁻¹) associated with shallow PBL (~1–1.2 km height) and moderate speeds associated with deeper PBLs (>2 km height).

During Etesians the total aerosol concentrations measured at the DEM surface station were quite high (~35 μg m⁻³), with an AOD (at 1064 nm) >0.1 and an even higher contribution of the fine mode (75%) than during continental flows. OC concentrations were highest (2–4 μg m⁻³) during Etesians, possibly due to the influence of Istanbul and the area surrounding the Black Sea, in accordance with Koçak et al. (2011). Once again, clouds were present with significant vertical development often resulting in precipitation, and these clouds had high water vapour content (>20 kg m⁻²). Moderate to high wind speeds within the PBL were recorded (>10 m s⁻¹) along with increased PBL height, which in extreme cases reached up to 2.6 km above ground level.

Finally, during periods of Saharan dust influence, the total aerosol concentrations at the surface were the highest observed (>42 μg m⁻³) with the lowest contribution of the fine mode (48%). Constituents of crustal origin, such as calcium and magnesium contribute significantly, especially in the coarse mode. OC and EC levels were within the values reported for Athens in May, during periods of high occurrence of dust transport (Paraskevopoulos et al., 2014). AOD (at 1064 nm) showed the highest values (>0.2) and the PBL height was quite low (~1 km). These were the only cases where no clouds were vertically developed and during these periods the lowest wind speeds within the PBL (<10 m s⁻¹) were observed, along with low water vapour content (<15 kg m⁻²).

Fig. 11. a. From top to bottom: the height region of the aerosol layer in relation to the dominant air mass type, the lidar ratio (S) values at 355 and 532 nm, the S₆₅₃₉⁵₃₂ and the corresponding Ångström exponent (AE)-related extinction coefficient (355 nm/532 nm) values, all based on the nighttime EOLE measurements, from 15 May to 16 June 2014. b. Upper panel: the height region of the aerosol layer, in relation to the dominant air mass type (same as in Fig. 10a); lower panel: the particle linear depolarization ratio δₚ (%), based on daytime AIAS measurements, from 15 May to 16 June 2014.
6. General conclusions

Air masses arriving over the GAA were classified into three main synoptic flow patterns: continental (72.31%), Etesians (14.36%) and Saharan (13.33%). The mean horizontal wind speed was of the order of 6–10 m s\(^{-1}\), reaching maximum values of about 20–22 m s\(^{-1}\), with a N-NE direction of the prevailing wind within the PBL. The development of local-scale flows and the diurnal variability of the near surface wind are prominent for the city of Athens due to both the complexity of the landscape and the sea breeze. The effects of these processes on the monitoring site are examined with the use of high resolution modeling systems (WRF and RAMS) that are capable of resolving such meso-γ scale wind circulations (Solomos et al., in preparation).

We found that under continental and Etesians wind flow, the PBLH was quite deep (≈2–2.5 km), while under Saharan wind flow conditions, the PBLH remained shallow (≈1–1.2 km). Concerning the evaluation of the daytime PBLH using model simulations we found ambiguous results of the comparisons between the WRF model and lidar data, dependent upon the PBL scheme and synoptic flow type. Differences as large as 300% were found between PBL heights from the lowest and highest PBL schemes, respectively. WRF model-simulated PBL heights with the ACM2 and QNSE schemes performed better than other schemes during Continental and Etesians synoptic flow types. In turn, the YSU and MYNN2 PBL schemes were better performers during the Saharan synoptic case examined. In general, a non-local PBL scheme such as ACM2 provided the most consistent results (Banks et al., 2016). Finally, direct radiative effects of dust particles on climate were investigated by Solomos et al. (in preparation) using the WRF model for the GAA.

The ionic composition of PM\(_{2.5}\) and PM\(_{10}\) revealed that hygroscopic particles contained high concentrations of SO\(_4^{2-}\) forming a significant percentage of their mass: 17 and 10% for PM\(_{2.5}\) and PM\(_{10}\), respectively. The sum of SO\(_4^{2-}\), NO\(_3^-\) and NH\(_4^+\) represented about 26 and 17% of the PM\(_{2.5}\) and PM\(_{10}\) mass, respectively. Peaks in concentrations of these largely anthropogenic source ions occurred during times of continental air masses arriving from N-NE directions. Hygroscopic particles from marine aerosol (Na\(^+\) and Cl\(^-\)) were present in much lower percentage (~3% in PM\(_{10}\) mass) their maxima being associated mostly with S-SW directions.

The ground-level aerosol RI values determined from in situ instruments (OPC and SMPS) were in good agreement (within 10–20%) with the daily-averaged retrieved dry RI estimates from PM\(_{2.5}\) measurements (Vratolis et al., in preparation). Furthermore, good agreement with the lidar-calculated RI values at the lowest height was observed during days with a deep PBL and relative humidities lower than 60%. RIs showed higher values in May (≈1.6) when SW winds are more frequent, and lower values in June (≈1.5), when the Etesian winds are more frequent (Vratolis et al., in preparation).

In this work we also investigated the synergy of aerosol in situ optical and microphysical measurements at the DEM station with lidar data. Such a comparison provided a deeper insight into efforts attempting to couple in situ and lidar aerosol measurements near the ground. Closure...
The information on the aerosol volume concentration is sufficient to convert the bare elastic and a Doppler lidar by calculating the cospectra of the vertical wind component and bare (Engelmann et al., 2008). The combination of CCN and lidar measurements demonstrated that CCN concentrations were inversely related to PBL height owing to a combination of ventilation and local production of aerosol. The analysis of the activation fraction distribution for a given level of supersaturation indicates that the hygroscopicity tends to increase with particle size, but particles exhibit a large degree of external mixing. Because of low updraft velocities (\(0.5 \text{ m s}^{-1}\)) associated with shallow PBL depths, cloud droplet number potentially developing in clouds remains low (\(<200 \text{ cm}^{-3}\)) despite the very high CCN number. The strong competition for water vapour that arises because of the low updrafts and high CCN also make clouds in shallow PBL highly insensitive to aerosol perturbations.

In contrast, increased updraft velocities (1–2 m s\(^{-1}\)) associated with Etesian flows, in combination with lower CCN levels increase cloud supersaturation by about a factor of 4. Because of this, cloud droplet numbers were much higher (\(<800 \text{ cm}^{-3}\)) even though CCN levels were not.

The Raman lidar data analysis revealed a wide range of geometrical and optical properties for different aerosol types. This was attributed to the external mixing of different aerosols (from near- or far-range aerosol sources) and the continuous influence of urban aerosol load in the region (Papayannis et al., in preparation-a, in preparation-b).

The aerosol mass upward flux was investigated with the synergy of an elastic and a Doppler lidar by calculating the cospectra of the fluctuations of the vertical wind component and bare (Engelmann et al., 2008). We found that the frequencies contributing to the vertical flux cover a wide range from \(1 \times 10^{-3}\) to \(1.8 \times 10^{-2}\) Hz which corresponds to eddy lengths of up to few hundred meters, depending on the wind speed (Argyrouli et al., in preparation-a). The time-collocated lidar and sun-photometer measurements allowed us to retrieve the vertical profile of aerosol volume concentration (considering both fine- and coarse-mode aerosols) using the LIRIC inversion code (Chaikovsky et al., 2015).

The inversion of data from our multi-wavelength Raman lidar enabled the retrieval of aerosol size spectra, which could then be used to retrieve CCN spectra as a function of supersaturation. Such retrievals, combined with cloud-scale vertical velocity from a Doppler lidar enabled the determination of cloud droplet number concentration in the vicinity of the PBL top. Lognormal size distribution was used for the CCN spectra estimation and a maximum fractional difference of \(\sim 30\%\) was found. The retrieval is more unstable for supersaturations below \(0.1\%\) and in particular the largest discrepancy was observed for supersaturations around \(0.06\%\). This discrepancy was reduced to \(20\%\), when an optimal maximum likelihood estimator was applied to weight the retrieved aerosol spectra. This new approach, proposed in Argyrouli et al. (in preparation-b), carries the potential to extend our knowledge of aerosol to cloud droplet population by means of synergistic lidar remote sensing.

A comparison between two methods of retrieving the vertical profiles of the relative humidity based on a combination of a Raman water vapour lidar with temperature profiles obtained from microwave radiometer or WRF simulations is presented by Labzovskii et al. (in preparation). The same study showed that combining lidar with microwave radiometer data drastically improves the accuracy of the retrieved relative humidity profiles (resulting in a 3.9\% accuracy in the retrieved mean value).

The inversion of data from our multi-wavelength Raman lidar enabled the retrieval of aerosol size spectra, which could then be used to retrieve CCN spectra as a function of supersaturation. Such retrievals, combined with cloud-scale vertical velocity from a Doppler lidar enabled the determination of cloud droplet number concentration in the vicinity of the PBL top. Lognormal size distribution was used for the CCN spectra estimation and a maximum fractional difference of \(\sim 30\%\) was found. The retrieval is more unstable for supersaturations below \(0.1\%\) and in particular the largest discrepancy was observed for supersaturations around \(0.06\%\). This discrepancy was reduced to \(20\%\), when an optimal maximum likelihood estimator was applied to weight the retrieved aerosol spectra. This new approach, proposed in Argyrouli et al. (in preparation-b), carries the potential to extend our knowledge of aerosol to cloud droplet population by means of synergistic lidar remote sensing.

The combination of CCN and lidar measurements demonstrated that CCN concentrations were inversely related to PBL height owing to a combination of ventilation and local production of aerosol. The analysis of the activation fraction distribution for a given level of supersaturation indicates that the hygroscopicity tends to increase with particle size, but particles exhibit a large degree of external mixing. Because of low updraft velocities (\(0.5 \text{ m s}^{-1}\)) associated with shallow PBL depths, cloud droplet number potentially developing in clouds remains low (\(<200 \text{ cm}^{-3}\)) despite the very high CCN number. The strong competition for water vapour that arises because of the low updrafts and high CCN also make clouds in shallow PBL highly insensitive to aerosol perturbations.

In contrast, increased updraft velocities (1–2 m s\(^{-1}\)) associated with Etesian flows, in combination with lower CCN levels increase cloud supersaturation by about a factor of 4. Because of this, cloud droplet numbers were much higher (\(<800 \text{ cm}^{-3}\)) even though CCN levels were not.

between these two types of measurements allows combination of in situ data with lidar aerosol profiles, hence providing a better understanding of local mixing in the lower atmosphere over complex terrain, under katabatic wind conditions (Fetfatzis et al., 2015).

We found that the frequencies contributing to the vertical flux cover a wide range from \(1 \times 10^{-3}\) to \(1.8 \times 10^{-2}\) Hz which corresponds to eddy lengths of up to few hundred meters, depending on the wind speed (Argyrouli et al., in preparation-a). The time-collocated lidar and sun-photometer measurements allowed us to retrieve the vertical profile of aerosol volume concentration (considering both fine- and coarse-mode aerosols) using the LIRIC inversion code (Chaikovsky et al., 2015).

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Fig. 13. a. Mean properties of elevated non-precipitating clouds detected and estimated from XPol radar over the DEM station from 6:00 to 18:00 UTC from 26 May to 23 June 2014. The time periods of radar operation are shown with horizontal bars. b. As in Fig. 13a, but for rain clouds.
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