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Long-term observations of tropospheric particle number size distributions and equivalent black carbon mass concentrations in the German Ultrafine Aerosol Network (GUAN)

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Abstract

The German Ultrafine Aerosol Network (GUAN) is a cooperative atmospheric observation network, which aims at improving the scientific understanding of aerosol-related effects in the troposphere. The network addresses research questions dedicated to both, climate and health related effects. GUAN's core activity has been the continuous collection of tropospheric particle number size distributions and black carbon mass concentrations at seventeen observation sites in Germany. These sites cover various environmental settings including urban traffic, urban background, rural background, and Alpine mountains. In association with partner projects, GUAN has implemented a high degree of harmonisation of instrumentation, operating procedures, and data evaluation procedures. The quality of the measurement data is assured by laboratory intercomparisons as well as on-site comparisons with reference instruments. This paper describes the measurement sites, instrumentation, quality assurance and data evaluation procedures in the network as well as the EBAS repository, where the data sets can be obtained (doi:10.5072/guan).

1 Introduction

Atmospheric aerosol particles, or particulate matter (PM), are essential constituents in the atmosphere, influencing issues such as atmospheric visibility, global climate, and human health. A climate-relevant effect is their interaction with solar shortwave radiation (Ramanathan et al., 2001). Two major aerosol effects influencing the terrestrial radiation budget have been distinguished: Direct radiative forcing – scattering and absorption of upwelling and down-welling radiation in the absence of clouds (Haywood and Boucher, 2000) and indirect radiative forcing – the modification of cloud radiative properties through the activation of additional particles as cloud condensation nuclei (Lohmann and Feichter, 2005). The magnitude of direct radiative forcing depends, in general, on various properties of the aerosol particles including particle diameter and

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chemical composition (Bohren and Huffman, 1998), but also shape, state of mixture, and hygroscopicity (Hänel, 1976; Zieger et al., 2013). Black carbon (BC) is among the species contributing to light absorption and, thus, atmospheric warming. According to a recent survey, the radiative forcing due to BC can, at present, only be bound to the uncertainty of a factor of two (Bond et al., 2013). The particle number size distribution and the light absorption coefficient are useful parameters to predict the direct radiative forcing on the basis of in-situ measurements.

On the other hand, ambient aerosol particles have been recognised to affect human health (e.g., Dockery and Pope, 1994; Pope et al., 2004; Dockery and Stone, 2007). Recent projections of health effects yield drastic numbers of morbidity and premature deaths due to particulate pollution worldwide (Lelieveld et al., 2015). In the European Union, the mass concentration of PM_{10} and $PM_{2.5}$ (particles smaller than 10 and 2.5 µm in aerodynamic diameter, respectively) currently serve as legal metrics to assess a population's exposure to ambient particles (European Council, 2008/50/EC). A rationale for using PM_{10} and $PM_{2.5}$ has been the large body of epidemiological evidence of adverse health effects based on these metrics.

Some studies, however, have suggested that the mass-based metrics might not be the most favourable parameter to characterise PM-induced health effects (HEI Review Panel on Ultrafine Particles, 2013). Some epidemiological studies have associated health endpoints with the number of ultrafine particles or the particle surface area rather than particle mass (Ibald-Mulli et al., 2002; Franck et al., 2011; Rückerl et al., 2011). Ultrafine particles are ubiquitous in urban atmospheres (Kumar et al., 2014), and their ability to penetrate deep into the human body after inhalation has been forwarded as a rationale for their adverse health effects. A recent overview by the world health organisation WHO (Janssen et al., 2012) also counted atmospheric soot particles (BC) among the relevant environmental risk factors for human health. While there seems little doubt about the potential adverse health effects of ultrafine particles and BC, their relatively low mass concentration makes them hardly accessible by total mass-based measurements. Particle number size distribution and BC mass con-

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centration have consequently been recommended as exposure parameters for future epidemiological studies (HEI Review Panel on Ultrafine Particles, 2013).

International observation networks for in-situ atmospheric aerosol measurements include WMO-GAW (World Meteorological Organization Global Atmosphere Watch) and EMEP (European Monitoring and Evaluation Programme). European research infrastructure programmes have contributed to the systematic collection of in-situ atmospheric aerosol data as well: EUSAAR (European Supersites for Atmospheric Aerosol Research) and ACTRIS (Aerosols, Clouds, and Trace gases Research InfraStructure Network). In the early 2000's the Nordic Aerosol Network implemented particle number size distribution measurements in a number of rural locations in Scandinavia (Tunved et al., 2003). The nature of most of these networks, however, has been to measure aerosol abundance and characteristics on a continental and global scale. Accordingly, the measurement sites are predominantly located in rural settings where direct anthropogenic influence is weak. Observation networks including urban sites are, for example, the Black Carbon and Particle Numbers and Concentrations Networks in the UK, operated by the National Physical Laboratory (Jones et al., 2012).

Government air quality networks in Europe operate many stations that collect data on PM_{10} and $PM_{2.5}$ mass concentrations, which are relevant to European air quality legislation. In the view of limited financial resources, however, there is usually limited incentive to measure aerosol and PM metrics that go beyond legal requirements, although such activities might provide enhanced scientific insights into climate-relevant or health-related processes.

In 2008, the German Federal Environment Agency (UBA) and the Leibniz Institute for Tropospheric Research (TROPOS) founded a new Germany-wide network for the characterisation of fine and ultrafine particles in the atmospheric aerosol. Several of UBA's manned background monitoring stations and numerous other legal and research institutions with their personnel and existing infrastructure have been involved. Notable institutions have included the Saxon State Office for Environment, Agriculture and Geology (LfULG), Helmholtz Zentrum Munich (HMGU), the Institute of Energy and En-

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vironmental Technology (IUTA), and the German Meteorological Service (DWD). As a result, continuous measurements of sub-µm particle number size distributions and equivalent BC mass concentrations have been installed and maintained at a total of seventeen observation sites. This paper serves to describe in detail the characteristics of the measurement sites, the instrumentation deployed for continuous particle measurements, and the location and properties of the data files.

2 Concept for long-term measurements

To date, there are wide experimental options to characterise atmospheric aerosol particles in much physical and chemical detail (e.g., McMurry, 2000; Baltensperger and Prévôt, 2008; Laj et al., 2009). The requirements of long-term deployment in a network, however, reduce these options to experimental methods that are sufficiently stable, reproducible, but also financially viable. When designing the German Ultrafine Aerosol Network (GUAN) in 2008, it was decided to implement a limited number of aerosol parameters measurements only, but with enhanced spatial coverage and operational reliability (Birmili et al., 2009a). The measurements include, in particular:

- Sub-µm particle number size distributions.
- Sub-µm particle number size distributions of non-volatile particles.
- Equivalent black carbon (eBC) mass concentrations.

Number size distributions of particles at dry conditions are measured by mobility particle size spectrometers. Depending on their individual set-up, these instruments are called Scanning Mobility Particle Sizer (SMPS), Twin Differential Mobility Particle Sizer (TDMPS), or Twin Scanning Mobility Particle Sizer (TSMPS). Number size distributions of non-volatile particles are measured after passage through a thermodenuder at a temperature of 300 °C. (Thermodenuders remove particulate compounds that are volatile at this temperature.) Equivalent BC (eBC) mass concentrations are measured

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by Multi-Angle Absorption Photometers (MAAP) mainly. During GUAN's first operation phase (2009–2014) these instrumental methods proved robust and yielded reproducible results so that they warrant successful deployment in an observation network.

3 Atmospheric observation sites

Figure 1 illustrates the location of GUAN's seventeen ground-based atmospheric measurement sites in Germany. Figure 2 provides maps and illustrations of the immediate surroundings of each measurement. Several Tables supply detailed information: Table 1, the characteristics of the measurement sites; Table 2, instrumental features; Table 3, co-location with other particle and air pollutant measurements; Table 4, associations with other networks, infrastructure, and research projects; Table 5, the list of institutions involved. The following text gives a brief description of each GUAN site, its measurement programme and references for published results.

3.1 Annaberg-Buchholz

Annaberg-Buchholz is a site of the Air Quality Monitoring Network of Saxony (AQMNS) run by the Saxon State Office for the Environment, Agriculture and Geology (LfULG, Dresden). Like all AQMNS sites, technical operations are conducted by the State Department for Environmental and Agricultural Operations in Saxony (BfUL, Radebeul). The site is located in the city of Annaberg-Buchholz (population ca. 21 000) in the Ore Mountains (Erzgebirge), distant about 10 km from the German–Czech border. Particle number size distributions and eBC mass concentrations have been measured continuously since 2012 in the framework of UltraSchwarz, a research project dedicated to ultrafine particles and health research in the German–Czech border region (Schladitz et al., 2015). The measurements are complemented by a suite of basic particulate and gaseous pollutants (cf. Table 3).

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3.2 Augsburg

Augsburg is an urban background monitoring station operated by the Helmholtz Center Munich (HMGU), Institute of Epidemiology II, and the University of Augsburg. The site was established in 2004 in the city of Augsburg (population ca. 270 000) in southern Germany with a main purpose to provide input to epidemiological studies of respiratory and cardiovascular disease (KORA, cooperative health research in the Augsburg region). The station is located on the University premises, about 1 km southeast of the city centre. Particle number size distributions and eBC mass concentrations have been collected continuously since 2004 (Pitz et al., 2008; Birmili et al., 2010a; Gu et al., 2012).

3.3 Bösel (Südoldenburg)

Bösel (Südoldenburg) is a regular site in the government air quality monitoring system of Lower Saxony operated by the Labour Inspectorate of Lower Saxony (Staatliches Gewerbeaufsichtsamt Hildesheim, GAA). To the south, the sampling site borders agricultural areas while to the north, it is adjacent to residential areas of the village of Bösel (population ca. 7400). The station is situated in an area where livestock production is most intense. The gaseous ammonia and organic emissions related to these activities are anticipated to have an impact on the regional budget of secondary aerosols. Bösel is distant about 100 km from the North Sea, so that maritime air masses can be sampled with a relatively minor impact of continental sources. Particle number size distributions and eBC mass concentration measurements were carried out on a continuous basis between 2008 and 2015.

3.4 Dresden-Nord

Dresden-Nord is another AQMNS station, located at roadside in the city of Dresden (population ca. 500 000). Continuous particle number size distribution measurements

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since 2001 have confirmed a pronounced diurnal cycle of traffic-related pollutants (Löschau et al., 2010). Around 36 000 vehicles pass by the site per day including 3.5 % heavy duty vehicles. The horizontal distances to the traffic flows range between 7 m (minor traffic flow; southerly direction) and 80 m (major traffic flow, westerly direction). The site borders the railway station Dresden-Neustadt, with mainly electrified trains passing by at a distance of 200 m to the north. Basic features of the particle number size distributions are presented in Gnauck et al. (2012) and Birmili et al. (2013).

3.5 Dresden-Winckelmannstrasse

Dresden-Winckelmannstrasse is another AQMNS station, located in the urban background of Dresden, about 1.7 km south the city centre. The next major road passes by the site at a distance of 100 m in easterly direction. Particle number size distributions measurements were established in 2010, eBC measurements in 2012. A major purpose of the site has been the provision of representative urban background concentrations for particle size distributions and particle mass within the UFIREG project (Table 4). The measurements are complemented by a suite of basic particulate and gaseous pollutants (Table 3).

3.6 Hohenpeissenberg

The Meteorological Observatory Hohenpeissenberg (MOHp) has a long tradition of meteorological and climatological observations. It is operated by the German Meteorological Service (DWD), and contributes to WMO-GAW, ACTRIS and EMEP. MOHp is located on a solitary hill in the rural countryside of southern Bavaria (980 m a.s.l.), approximately 40 km north of the Alpine mountain range. The observatory resides around 300 m above the surrounding rural areas that are composed mainly of agricultural pastures (70 %) and forests (30 %). MOHp hosts a very wide range of atmospheric aerosol and gas phase measurements. Some highlights include continuous observations of volatile organic compounds (VOCs), gaseous sulphuric acid (H_2SO_4), and numerous

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remote sensing parameters. For the basic characteristics of particle number size distributions and their relation to trace gas and meteorological parameters see Birmili et al. (2003).

3.7 Langen

Langen is an urban background measurement site located on the premises of the Federal Environment Agency of Germany (UBA) in Langen. The site is located 15 km south of the city of Frankfurt/Main and 5 km southeast of Frankfurt's Rhein-Main airport. Aerosol particles are sampled at a height of 14 m on the rooftop of the UBA building. Continuous particle number size distribution measurements started in 2008, complemented since 2009 by measurements of total particle number concentrations (UCPC, TSI model 3776) and lung disposable surface area (Nanoparticle Surface Area Monitor, TSI model 3550) (Gerwig et al., 2014).

3.8 Leipzig-Eisenbahnstrasse

Leipzig-Eisenbahnstrasse is a roadside observation site in the city of Leipzig (population ca. 500 000), operated by TROPOS since 2002. The site is located in a street canyon within a densely built-up residential area, characterised by multi-storey period buildings. The street canyon is regular in that its aspect ratio is close to unity (height: 18 m, width: 20 m), and no building gaps are present. The street carries about 12 000 motor vehicles per workday. Ambient aerosol is sampled 6 m above street level on the northern side of the street. Due to the formation of a vortex inside the street canyon, northerly winds have been identified as the condition that favours high particle number concentrations (Voigtländer et al., 2006). For an account of the spatial and temporal variability particle of number size distributions in this area of Leipzig, see Costabile et al. (2009).

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3.9 Leipzig-Mitte

Leipzig-Mitte is another AQMNS station, located at roadside in the city of Leipzig. The site borders the inner-city ring road, in close vicinity to the central train station. Immediately north of the site, three main roads merge at an intersection with daily average traffic volumes around 44 000 vehicles (48 000 on workdays). Among all GUAN sites, Leipzig-Mitte exhibits the greatest exposure to traffic-related pollutants. PM_{10} mass and ultrafine particle number concentrations were discussed by Engler et al. (2012) and Ma and Birmili (2015), respectively. Leipzig-Mitte was added to GUAN for the purpose of monitoring possible changes in UFP number and eBC mass concentrations along with the introduction of the low emission zone (*Umweltzone*) in Leipzig (Rasch et al., 2013; Löschau et al., 2014). Particle number size distribution and eBC measurements started in 2010. The measurement portacabin borders a tributary road connected to the ring road by traffic lights. Construction activities in the vicinity of the site have occasionally disturbed the measurements between 2010 and 2012. Days on which the impact of construction works was significant were documented in Löschau et al. (2012, 2013).

3.10 Leipzig-Tropos

Leipzig-Tropos (simply called "Leipzig" in certain data bases) is an atmospheric research station operated by TROPOS since 1997. The station is situated on the roof of the TROPOS institute building. Aerosol particles are sampled at a height of 16 m above the ground. Highly-trafficked roads touch the premises at distances of at least 100 m. Comparisons of particle number size distributions at multiple sites in Leipzig have confirmed Leipzig-Tropos as an urban background station (Costabile et al., 2009; Ma and Birmili, 2015). A cross-sectional study suggested a total particle number concentration mean of 9400 cm^{-3}, which proved to be higher than at comparable sites in Helsinki and Copenhagen (von Bismarck-Osten et al., 2013). During the heating season, the site can be influenced by a gas heating stack 50 m south of the aerosol inlet. A screening of the data showed that a perturbation of the measurements is likely under

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southerly winds, and at temperatures below 0 °C. The size distribution profile of the gas heating stack affects particle number concentration below 30 nm only.

3.11 Leipzig-West

Leipzig-West is another AQMNS site in Leipzig, located in the western suburbs of Leipzig. The purpose of the site is to provide a second measurement for urban background concentrations in Leipzig. The distance to Leipzig-Mitte is about 6 km, to Leipzig-TROPOS about 10 km. The residential area around the station consists of multi-storey apartment blocks that are heated by district heating. The measurement container is installed in a park area on the premises of a hospital. A minor road passes by the station around 30 m west of the site but has shown to be of negligible influence on the measurements. Particle number size distribution and eBC measurements started in 2010 (Rasch et al., 2013).

3.12 Melpitz

Melpitz is an atmospheric research station operated by TROPOS since 1992. The station is located in eastern Germany near the city of Torgau, and ca. 50 km northeast of Leipzig. The site is surrounded by flat and semi-natural grasslands without any obstacles in all directions. Besides GUAN, Melpitz contributes to WMO-GAW as a regional background site, ACTRIS, and EMEP. Measurements at Melpitz can be taken as representative for the Central European background atmosphere (Asmi et al., 2011). Melpitz hosts a wide suite of physical and chemical measurements of atmospheric aerosols (Birmili et al., 2008). The site has proved useful to detect long-range pollution transport from continental areas in easterly direction (Spindler et al., 2010). The distance to the North Sea is about 400 km in north-westerly direction, and to the Atlantic Ocean about 1000 km in westerly direction. An account on the basic aerosol characteristics can be found in Engler et al. (2007), Spindler et al. (2010) and Ma et al. (2014). The effects of hygroscopic particle properties on atmospheric light scattering were discussed in

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(Zieger et al., 2014). The formation of new atmospheric particles from gaseous precursors has been analysed here since the 1990s (Birmili and Wiedensohler, 2000; Größ et al., 2015).

3.13 Mülheim-Styrum

Mülheim-Styrum is a regular observation site in the air quality monitoring network of North Rhine-Westphalia (LUQS). The site is operated by the State Agency for Nature, Environment, and Consumer Protection (LANUV). IUTA Duisburg operates an additional portacabin for research measurements. The site is situated within a residential area but is also within reach of a motorway (around 250 m to the north), a national road (B223, around 400 m to the west), and industrial premises (around 600 m to the east and south). Overall, the site qualifies as an urban background monitoring station (Quass et al., 2004) and has been used for exposure assessment in health related studies (ESCAPE, European Study of Cohorts for Air Pollution Effects) and source apportionment studies (Beuck et al., 2011). Mülheim-Styrum was added to GUAN as a representative for the Ruhr Area, the largest urban agglomeration in Germany. Continuous particle number size distribution measurements started in 2008. Lung disposable surface area has been measured by a Nanoparticle Surface Area Monitor (TSI model 3550) as an additional measure for quality assurance.

3.14 Neuglobsow

Neuglobsow is one of the permanently manned stations within UBAs regular observation network (UBA, 2013). The sampling site is surrounded by lakes and forested areas in all directions, and is therefore only very little influenced by local sources. Neuglobsow contributes to EMEP. Measurements here can be taken representative for the atmospheric background in north-eastern Germany. Particle number size distribution and eBC measurements started in 2010.

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3.15 Schauinsland

Schauinsland is another of UBA's manned observatories. The station is located at 1205 m a.s.l. near the Schauinsland peak in the Black Forest in south-western Germany. Measurements started as early as 1965 as part of research programmes funded by the German Science Foundation (DFG). The station is well-suited to characterise air masses that approach Central Europe from westerly directions. In winter, the site tends to reside in relatively clean air above the Rhine valleys inversion layer. Observations at Schauinsland have been focused on the detection of long-term trends. The site hosts, for instance, the longest continuous observation of carbon dioxide (CO_2) observations in Europe, since 1972 (Schmidt et al., 2003). Particle number size distribution measurements started in 2005, eBC measurements in 2008.

3.16 Waldhof

Waldhof is another manned station within UBAs regular observation network. The sampling site is surrounded by forest in all directions, and therefore only very little influenced by local sources. Measurements here can be taken representative for the background in the North German lowlands. Waldhof is the only German atmospheric station contributing to GMOS (Global Mercury Observation System) (Weigelt et al., 2013). Particle number size distribution and eBC measurements started in 2008.

3.17 Zugspitze (Schneefernerhaus)

Zugspitze (Schneefernerhaus) is part of the global WMO-GAW station Zugspitze/Hohenpeissenberg and jointly operated by the Federal Environment Agency of Germany (UBA) and the German Meteorological Service (DWD). The observatory is located at 2670 m a.s.l., about 300 m below the Zugspitze summit, and on the southern slope of the corresponding mountain massif. The high altitude leads to a significant annual cycle in observed aerosol particle number and mass

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concentration, caused by different boundary layer heights in summer and winter (Birmili et al., 2009b) The station's elevated position allows to sample air masses that had only little contact with the local boundary layer; especially in the cold season from October to March. Zugspitze occasionally receives lofted aerosol layers from remote source regions, such as North America (Birmili et al., 2010b), or the Eyjafjallajökull volcanic eruption (Schäfer et al., 2011). Besides the GUAN measurements, the Karlsruhe Institute of Technology conducts long term measurements of total particle number concentration (CPC), particle number size distribution (OPC; APS), and primary biological aerosol particles. Particle number size distribution measurements started in 2004, eBC measurements in 2008.

4 Instrumental

4.1 Particle number size distributions

Particle number size distributions are recorded on a continuous basis using mobility particle size spectrometers. Depending on the specific configuration, these can be Twin Differential Mobility Particle Sizers (TDMPS; typical particle size range 5–800 nm), Twin Scanning Mobility Particle Sizers (TSMPS; size range 5–800 nm), or Scanning Mobility Particle Sizers (SMPS; typical size range 10–800 nm). The core component of each mobility particle size spectrometer is a Differential Mobility Analyser (DMA).

4.1.1 SMPS (TROPOS-design)

The principle of the custom-built SMPS is described in detail in Wiedensohler et al. (2012). Briefly, the instrument uses a Vienna-type DMA (electrode length 28 cm) with a condensation particle counter (CPC model 3772, TSI Inc.) to measure particles between 10 and 800 nm. Before entering the DMA, ambient aerosol is conveyed to a bipolar charge equilibrium using a Kr^{85} neutraliser. Sheath flow rate is 5 $L\,min^{-1}$ at an aerosol flow rate of 1 $L\,min^{-1}$. The high voltage supply provides positive polarity,

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thus collecting negatively charged particles. The sheath air is generally circulated in a closed loop. Both the aerosol sample flow and the sheath air flow are actively dried in this instrument, thus ensuring a relative humidity during size classification below 40 % at most times. Temperature, relative humidity, and pressure inside the instrument are continuously monitored. The typical time resolution for one combined upscan and downscan is 5 min. TROPOS-designed SMPS instruments are currently deployed at the GUAN stations 1, 3, 5, 6, 14, 15, and 16 (see Table 2).

4.1.2 SMPS (other designs)

Commercial SMPS, the Scanning Mobility Particle Sizer Spectrometer (model 3936, TSI Inc., Shoreview, USA) are deployed at the GUAN stations 7, 13, and 17. The sheath air to aerosol flow ratio is 5 : 1 L min^{-1} at the stations 7 and 17 – yielding a particle size range 10–600 nm, and 3 : 0.3 L min^{-1} at station 13 – yielding a particle size range 14–750 nm. Time resolution is 5 min. The instruments at all stations were upgraded to meet the quality criteria recommended for ambient aerosol measurements by the EUSAAR and ACTRIS initiatives (Wiedensohler et al., 2012). The upgrades concern dryers for the aerosol sample flow and sheath air, as well as additional sensors for temperature, relative humidity, and pressure. At station 17, the high voltage supply has been changed to positive polarity and a range up to 12.5 kV. Thus, negatively charged particles are collected, which exhibit a higher charging probability than positively charged particles.

4.1.3 TDMPS/TSMPS

Mechanically, the TDMPS and TSMPS are dual DMA versions of the SMPS described in Sect. 4.1.1 (Birmili et al., 1999). The first subsystem combines an ultrafine Vienna-type DMA (electrode length 11 cm) with an ultrafine condensation particle counter (UCPC model 3025, TSI Inc., Shoreview (MN), USA) to measure particles across the range 3–80 nm. The second subsystem combines another DMA (electrode length

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28 cm) with a condensation particle counter (CPC model 3010 or 3772, TSI Inc.) to measure particles between 10 and 800 nm. Due to enhanced measurement uncertainties below 5 nm, only the diameter range 5–800 nm is further analysed and fed into the EBAS data base. Alike the SMPS, sheath air is circulated in a closed loop at relative humidities ranging mostly between 10 and 40 %. The typical time resolution of the instrument is 10 min. TDMPS instruments are deployed at the sites 2, 4, and 12 while TDMPS instruments are operated at the sites 8–11. TSMPS and TSMPS have, by principle, no major differences in hardware. In software, TSMPS use a continuous ramping of the high voltage, like in the SMPS, rather than the step-wise change in the TDMPS.

4.2 Non-volatile size distributions (thermodenuder)

Upstream of some mobility particle size spectrometers, a thermodenuder (TD) is deployed as an option to remove volatile aerosol components. The standard operation mode for these extended instruments is to record size distributions upstream and downstream of the TD in alternating sampling intervals (Birmili et al., 2010a). This procedure provides a steady flow of size distributions both with and without the TD, and an effective time resolution of half the original instrumental time resolution.

One thermodenuder type follows the design of Wehner et al. (2002). Volatile particle material is evaporated at a temperature of 300 °C, and subsequently removed with the assistance of active carbon in a cooling section. The temperature of 300 °C was selected with the aim of evaporating the overwhelming mass of volatile and semi-volatile material, particularly ammonium nitrate, ammonium sulphate and most organic carbonaceous compounds. Major compounds not removed include elemental carbon, crustal material, and sodium chloride (cf. Engler et al., 2007). Meanwhile, 300 °C is a temperature at which charring (i.e. the incomplete combustion of oxygenated hydrocarbons) of organic compounds is avoided. A mass closure for the non-volatile particle fraction at the research station Melpitz suggested that the non-volatile fraction at 300 °C

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contains not only refractory black carbon, but also a comparable share of low-volatility organic aerosol compounds (LVOCs; Poulain et al., 2014).

The second, simplistic thermodenuder type consists just of a simple steel tube that is heated to 300 °C by a laboratory furnace. Excess vapours are only adsorbed by the tube walls. Such a simplistic TD has been used, for example, by Ehn et al. (2007). Experiments with ambient aerosol in Leipzig suggested that both thermodenuder designs produced equivalent results for non-volatile particle size distributions across the diameter range 10–800 nm. The tubing acting as a TD is usually cleaned once per year.

In the TD, the aerosol sample is subject to enhanced particle losses. One can think of, e.g., diffusional as well as thermophoretic losses, which are both a function of particle size. In the Wehner-type thermodenuder, part of the sample flow passes through the active carbon filter. Therefore, this instrument was calibrated for particle losses using spherical silver particles (solid at 300 °C), and by measuring the particle counts upstream and downstream of the TD. The penetration of solid particles through the Wehner-type thermodenuder was about 0.34 at 3 nm, 0.66 at 10 nm and 0.85 for particles bigger than 100 nm at a flow rate of 2.5 $\mathrm{L\,min^{-1}}$. All data collected from this thermodenuder type were corrected for these losses.

4.3 Aerosol absorption and equivalent black carbon (eBC)

The aerosol absorption coefficient is measured by Multi-Angle Absorption Photometers (MAAP). The MAAP converts the light attenuation and reflection by a particle-laden quartz fibre filter into an absorption coefficient by calculating the radiative transfer through this two-layer system (Petzold and Schönlinner, 2004). The wavelength (λ = 637 nm) corresponds to the region of the solar spectrum where black carbon (BC) is the prime absorber, thus minimizing interferences with "brown carbon" and mineral dust. Brown carbon and mineral dust tend, in fact, to absorb light more efficiently towards the ultraviolet wavelengths (Sun et al., 2007; Müller et al., 2009).

Because there is no unique relationship between aerosol absorption and black carbon mass concentration, black carbon is typically reported as equivalent black car-

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bon (eBC) (Petzold et al., 2013). For this purpose, the aerosol absorption coefficient σ_{abs} is converted into an eBC mass concentration using an experimentally determined mass absorption cross section (MAC). The manufacturer of the MAAP instrument reports a MAC value of $6.6\,m^2\,g^{-1}$, which is automatically applied on all data records. An assessment of aerosol absorption using Raman spectroscopy and elemental carbon (EC) measurements as reference methods yielded a mean MAC value of $5.3\,m^2\,g^{-1}$ with a range of variability between 3.9 and $7.4\,m^2\,g^{-1}$ across a selection of seven GUAN sites (Nordmann et al., 2009, 2013). An intercomparison of multiple instruments showed that different MAAP instruments produce comparable results with less than 5 % inter-device variability (Müller et al., 2011). Besides eBC mass concentrations, the MAAP yields the raw signals of loaded and blank filter material at scattering angles of 0, 135 and 165°, the sample flow rate, temperature and pressure, which may be stored internally in a format called "scientific data format". To provide accurate and comparable measurements under dry sample conditions, the MAAP aerosol flow is usually conditioned by a membrane dryer.

At Augsburg, an aethalometer (Type 8100, Thermo Fisher Scientific Inc.) is deployed using a cut-off of 2.5 µm. This instrument yields mean eBC mass concentrations that are comparable to those from a MAAP instrument. Details on this instrument can be seen in the Appendix A.

Unfortunately, not all eBC measurements in GUAN use the same inlet configuration, and factually, inlets for PM_{10}, $PM_{2.5}$ and PM_1 are used throughout the network (Table 2). In order to harmonise the eBC values, the data recorded downstream of the PM_1 cyclone inlets may be adjusted to the corresponding level of a PM_{10} inlet using suitable correction factors. The multiplication factors recommended here are 1.10 for rural sites, 1.08 for urban background sites, and 1.05 for roadside sites. These values were determined by a direct intercomparison of the readings of two MAAP instruments using a PM_{10} and a PM_1 inlet, respectively, at the sites Leipzig-Eisenbahnstrasse (roadside), Leipzig-TROPOS (urban background), and Melpitz (rural background). Because very high correlations were found during those intercomparison experiments ($R^2 = 0.99$), a post correction appears justified (Löschau et al., 2012).

5 Quality assurance

Quality assurance (QA) in GUAN includes a number of measures that ensure measurement data to stay on a comparable level to each other and, beyond this issue, on a defined level compared to a standard. The following paragraphs describe the state-of-the-art QA procedures for mobility particle size spectrometers, which were obeyed at the majority of the stations. Due to the different degree of access and availability of man-power, they may not be valid in all details at every single station and for all historic parts of the data collection. Most of these measures were co-developed within the framework of previous infrastructure projects, such as WMO-GAW, EUSAAR, ACTRIS, and research projects initiated by the Saxon State Office for Environment, Agriculture and Geology (LfULG).

5.1 Maintenance

5.1.1 Weekly or bi-weekly inspection

At unmanned GUAN stations, the mobility particle spectrometers and MAAPs are inspected personally at least every two weeks, preferably every week. At the manned GUAN stations (sites 6 and 14–17), the instrumentation is usually inspected more often, up to once per workday. At unmanned sites, remote data access has helped to check instrumental performance. Unfortunately, remote data access to unmanned stations is currently available only for a few stations (e.g., 1, 2, 4, 5, 12). The weekly or bi-weekly inspection of mobility particle size spectrometers includes visual checks whether all instrumental components are switched on and working correctly: High voltage power supply, sheath air flow and aerosol flow; condensation particle counters

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(CPCs); supply of CPC working liquid (butanol or water); data acquisition program; flow status and operation of the MAAP.

5.1.2 Monthly maintenance

A full maintenance is typically made with mobility particle size spectrometers every four weeks. Here, instrumental flow rates are verified using an external reference flow meter, usually a bubble flow meter: Aerosol flow rate, sheath air flow rate, flow rate of the aerosol dryer's counter flow, flow rate of the sheath air dryer counter flow. Aerosol and sheath air flow meters in the mobility particle size spectrometers are recalibrated if they deviate by more than 5 and 2 % from their set point values, respectively. Instruments are also checked for leaks using a total particle filter. The performance of the mobility particle size spectrometers is deemed satisfactory if the total particle number collected by the instrument after a waiting time of 15 min does not exceed 10 particles cm^{-3}.

The high voltage supply for each DMA is checked with a digital multimeter, involving a verification of the high voltage between 0 and 1000 V. Re-calibrations are made if the voltages exceed defined thresholds at 0 V (±3 V), 6.25 V (±25 %), 100 V (±10 %), and 1000 V (±1 %). Frequently checking the high voltage is essential to provide a correct sizing of the DMA particularly at the lower end of the particle size distribution. A NIST (National Institute of Standards and Technology, US Department of Commerce) certified particle size standard is used to verify the exact sizing of the instrument. Currently, the most popular standard are 203 nm Polystyrene latex (PSL) spheres, which are certified within 2.5 % of the nominal particle diameter. The particles are nebulised from aqueous suspension using a jet nebulizer (e.g., PariBoy, Pari GmbH, Starnberg, Germany). If the sizing of the mobility particle size spectrometer deviates more than 2 % from the standard size (i.e. is outside the interval 200–206 nm), the sheath air flow is adjusted until the DMA matches the certified diameter of the PSL particles.

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5.1.3 Annual maintenance

The annual maintenance event includes more extensive instrumental checks and care. It is preferentially performed in a central laboratory. Many GUAN instruments are returned to the World Calibration Centre for Aerosol Physics (WCCAP; Wiedensohler et al., 2012) at TROPOS Leipzig once a year. The annual maintenance includes: Check and calibration of the humidity sensors; calibration of the sheath air flow rate zero offset; check and calibration of the pressure transducer; disassembly and cleaning of the DMA(s); check of the saturator sponge inside the CPC; check and calibration of the CPC(s) against a particle number concentration standard. It has also been found important to check the activity of the charge neutralizer. A Kr^{85} beta source, for example, degrades substantially in its ion production rate after about 10 years. The annual maintenance event is also used to perform hardware improvements and software updates.

5.2 Comparison to reference instruments

Intercomparisons with reference instruments are essential in establishing a relationship towards to a defined standard. In case of mobility particle size spectrometers, we use CPCs of a specific type (model 3010 and 3772, TSI Inc., Shoreview, USA) and an electrometer (model 3068B, TSI Inc.) as an intermediate standard for particle number concentration. It is also a standard procedure to compare a mobility particle size spectrometer once per year against a reference instrument, which will reveal possible deviations with respect to the size-dependent instrument response. In GUAN, these intercomparisons are made in the central laboratory, within calibration workshops, or in the field within a “round robin test”.

5.2.1 Laboratory intercomparisons

Intercomparisons against the TROPOS reference mobility particle size spectrometers can be made at the WCCAP “on the fly”, i.e. on short notice at most times. However,

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it is a common practice to collect a pool of instruments, which are then examined collectively against one or two reference instruments (Wiedensohler et al., 2012). Because of their state close to the bipolar charge equilibrium, we generally prefer ambient aerosols for all instrumental intercomparisons, even if the experiments are conducted in the central laboratory. Three reference instruments are provided by WCCAP for quality assurance, which are checked regularly against each other, and against total particle counters. (Reference instruments need to agree within ±5 % for all particle sizes between 20 and 300 nm.) Laboratory intercomparisons are preferentially accompanied by a total particle counter (CPC). The deviation between the total particle number concentration derived from any mobility particle size spectrometer and the total particle counter must not exceed ±10 % for ambient aerosols. On the basis of such laboratory intercomparisons, we estimate the accuracy of the particle number concentration measured by mobility particle size spectrometers to be ±10 % for the diameter range 20–300 nm. Outside this range, larger uncertainties are possible. Below 20 nm, deviations between SMPS instruments may amount up to 50 %, above 300 nm up to 30 %. TDMPS and TSMPS instruments exhibit a higher accuracy at the lower particle size end, ±10 % down to 10 nm, and ca. ±30 % at 5 nm.

5.2.2 Round-robin intercomparisons

Ideally, every mobility particle size spectrometer undergoes at least one calibration experiment per year. To bridge gaps between the scheduled WCCAP calibration workshops, the QA measures may include a round-robin test on its measurement site using a reference mobility particle size spectrometer. In practice, such field intercomparisons usually involves (a) setting up a reference instrument in parallel to the mobility particle size spectrometers on site, (b) perform parallel size distribution measurements of ambient aerosol with the reference instrument during one night (introductory performance test), (c) maintain and/or improve the instrument in case of problems, and (d) repeat the ambient aerosol comparison for several days as a final performance test. The round-robin test usually involves checks of particle sizing and parallel measurements by a to-

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tal particle counter (CPC) as well. As in the case of the laboratory intercomparisons, an agreement between the total number concentration of a mobility particle size spectrometer and the reading of a total particle counter ±10 % is required. In most practical cases, we also found a good agreement between the test and reference instruments within ±10 % for the diameter range 20–300 nm.

5.2.3 Enhanced quality assurance

For three stations in Saxony (sites 1, 4 and 5), enhanced quality assurance measures have been developed (Schladitz et al., 2014). At these sites, the mobility particle size spectrometers are equipped with an automatic function control unit that performs unattended instrumental comparisons. First, a leak check using a total particle filter is performed every three days. A dedicated total particle counter (TSI model 3772, "transfer CPC") is moved to each of the three stations at a frequency of eight weeks. During a two-week presence of the transfer CPC, the control unit will perform comparison measurements between this CPC and the particle size spectrometer every 23 h. To avoid uncertainties due to nucleation mode particles present in the lower cut-off region of the CPC, particles at the lower end of the number size distribution are removed by a diffusion screen. For traceability, the transfer CPC is checked every eight weeks against a dedicated reference CPC at WCCAP, and once a year against a calibrated electrometer. This measure allows an even closer tracking of the actual performance of the mobility particle size spectrometer than under less infrequent intercomparisons. According to our experience, this measure can narrow down the uncertainty with respect to particle number concentration across the diameter range of 20–300 nm from ±10 to ±5 %.

At several sites, additional instrumentation has been used as a support for quality assurance and control: At the sites 2, 6, and 17 CPCs have been deployed on a continuous basis to measure total particle number concentration. As total particle concentration can also be computed from the particle number size distribution, a divergence of the two readings can help identify instrumental problems. At the sites 7 and

13, Nanoparticle Surface Area Monitors (NSAM, TSI Inc.) have been deployed as a QA tool, because the lung deposited particle surface area (LDSA) can be computed from the particle number size distribution as well. At site 13, significant deviations between the measured and calculated LDSA over several consecutive days have indicated failure of one of the instruments which, in practice, almost always turned out to be the SMPS or its CPC.

6 Data processing and validation

6.1 Particle number size distribution processing

Particle number size distributions from all stations except station 13 were processed using the TropINV software package, written in LabVIEW (Version 8.5, National Instrument, Austin, USA). This program evokes a linear multiple charge inversion algorithm (Muchain, Multiple Charge Inversion; Pfeifer et al., 2014), and performs various additional corrections before yielding the final particle number size distributions. TropINV performs the following steps:

1. Reformatting the raw concentration and instrument diagnostics data (optional).
2. Condensing the number of size bins of the raw data to a fraction of its original number (optional).
3. Automatic flagging of periods exhibiting diagnostics data outside the nominal range.
4. Assimilation of UDMA and DMA branches (only for TDMPS and TSMPS instrument).
5. Multiple charge inversion (Pfeifer et al., 2014).

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6. Adjustment of particle number concentrations for non-ideal aerosol flow rates (optional).

7. Size-dependent corrections of particle number concentration (Wiedensohler et al., 2012).

   - CPC counting efficiency.
   - Differential mobility analyser (DMA) transfer function.
   - Bipolar charger.
   - Aerosol dryer.
   - Connecting tubes inside and outside of the instrument.

8. Normalisation of ambient concentrations to 0 °C, 1013 hPa (optional).

9. Re-binning the size channels to a pre-defined standard set of standard channels.

Step 7 yields technically correct data that are ready for scientific use (dubbed "Level-1" in Sect. 7.2). Step 8 yields data that, after averaging to one hour resolution, form "Level-2" data (Sect. 7.2). GUAN has followed a practice that during step 9, all particle number size distributions are re-binned to a uniform set of 40 channels between 10 and 800 nm in the case of SMPS instrument, and 46 channels between 5.1 and 800 nm by linear interpolation. It is expected that this way, the data processing will be facilitated for the scientific end users. The data at station 13 was processed using the Aerosol Instrument Manager software (TSI Inc., Revision G, October 2006), which performs all the necessary steps 5–7 in a manner equivalent to the TropINV software (Pfeifer et al., 2014).

6.2 Technical and manual data validation

The first step of quality control of mobility particle size spectrometer data involves the automatic flagging of data records that are associated with invalid instrument diagnostics data. For example, the relative humidity in the instrument needs to remain below

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40 %. Instrumental temperature needs to be within a range of +10 to 30 °C. Aerosol sampling flows are not permitted to deviate from their set point by more than ±10 %, and the sheath air flows must not diverge more than ±5 % from their set point. These thresholds are motivated in Wiedensohler et al. (2012).

Further quality control involves additional visual checks by the scientists processing the data. One procedure is to carefully inspect contour diagrams of the particle number size distribution. An example for such contour diagrams at the highest available time resolution is given in Fig. 3. The human eye is rather sensitive to unusual features in such two-dimensional structures, and according to our experience, this visual inspection has proved very effective in detecting irregularities in the data, such as:

- Flash-over of high voltage between the two DMA electrodes. (This unwanted effect tends to produce artificial particles at the upper end of the size distribution.)
- Decrease in the counting efficiency of a CPC, as a result of laser diode power degradation.
- Contaminations, such as leaks of the instrument against room air
- Disturbing particle sources inside and outside of the measurement station.

A second tool is the screening of the time series, spanning one half year or more, for the all-time maxima and minima. We look, for example, at the total particle number and volume concentrations calculated from number size distributions, and review their time series and histograms for suspicious outliers. Individual values far outside the main frequency distribution are then inspected more closely and, if judged as the result of technical faults, deleted from the subsequent processing.

7 Data repository and format

7.1 EBAS (World Data Center for Aerosols)

Particle number size distributions and aerosol absorption coefficients from all GUAN stations are stored at the World Data Center for Aerosols EBAS data base, where they are publicly available and free of charge. The GUAN data can be accessed by following the persistent identifier doi:10.5072/guan, or by visiting the EBAS user interface directly (http://ebas.nilu.no). EBAS is a database infrastructure operated by NILU (Norwegian Institute for Air Research) with basic funding secured through the EMEP protocol of the Convention on Long-range Transboundary Air Pollution. EBAS handles, stores and disseminates atmospheric composition data generated by international and national frameworks on long-term monitoring programmes and research projects and has, for instance, been used by more than 50 European Union research projects. Dedicated staff (ca. 10 persons) secure the reliability for long-term data access.

Data integrity, interpreted as quality assurance and protection of the data holdings, is secured by the following measures: Each data submission to EBAS undergoes manual quality assurance for correctness of the accompanying metadata, as well as an extra check of the data records for outliers and implausible values. Errors are iterated with the data originators until the submission passes quality assurance. Once inserted into the database, the data is backed up together with the whole database at least twice per week (2 full and 2 incremental back-ups per week) at an off-site location.

With regard to copyright, any scientific user is free to download, copy, distribute, transmit, and adapt the data sets as long as he/she gives credit to the original authors (equivalent to the Creative Commons Attribution License). Data users accept to make an offer of co-authorship through personal contact with the data providers or owners whenever substantial use is made of their data. It is desired that the data users get in touch with the data originators, in order to create positive feedback, and foster scientific communication. In all cases, an acknowledgement must be made to the data originators and to the project name when these data are used within a publication. Tørseth

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et al. (2012) illustrate the possibilities of a cross-sectional study that makes extensive use of data stored at EBAS.

7.2 Data format

At EBAS, aerosol data are stored in three levels, reflecting an enhanced data submission protocol for particle number size distribution and BC mass concentration data. The objective of a three-level protocol is to enhance traceability of measurements and data processing, ranging from instrumental raw data up to final hourly averages.

Level-0 data contain raw data, i.e. electrical particle mobility distribution data and instrumental diagnostic parameters, as they are measured directly by the instrument. The electrical particle mobility distribution is provided as an array of measured particle number concentrations vs. particle diameters (electrical particle mobility for singly charged particles).

Level-1 data are scientifically correct particle number size distributions after multiple-charge inversion and the correction for particle losses. In the case of particle number size distributions, the data refer to the original conditions during the measurement, i.e. ambient pressure at the station, and typically 20 °C laboratory temperature. In the case of the aerosol absorption coefficient, the data refer to standard temperature (273.15 K) and pressure (1013.25 hPa) already. In any case, the time resolution refers to the original instrumental time resolution.

Level-2 data are given as hourly averages, and adjusted to a uniform standard temperature (273.15 K) and pressure (1013.25 hPa). Moreover, most of the particle number size distribution data sets have been rebinned to the same standard set of 40 or 46 diameter channels (cf. Sect. 6.1). Level-2 data are presumed to be the main point of interest to most data users. Level-2 data is available through EBAS user interface (see Supplement) while Level-0 and Level-1 data are available at request from the administrators of the data base.

Formally, data at EBAS are stored in the NASA-AMES format, which is based on the ASCII text NASA-Ames 1001 format, but contains additional metadata specifications

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ensuring instrumental documentation. For more information on the format, we refer the reader to Wiedensohler et al. (2012) and the EBAS website. An extract of NASA-Ames 1001 text is provided in the Supplement.

7.3 State of data submission

Although data are collected continuously, the body of GUAN data currently requires some degree of manual quality control and semi-manual data post processing. Data parcels encompassing half a calendar year are currently submitted to EBAS at regular intervals. Feeding the data from all seventeen GUAN stations and also non-volatile particle number size distributions into EBAS has not been completed, and will require, especially in the case of some retrospective data sets, additional time. It has been our first priority to transfer all recent GUAN data spanning the years 2009–2014 into EBAS. The Tables provided on GUAN's download page doi:10.5072/guan presents the current state of the data submission and availability. (A snapshot of that page is also shown in the Supplement to this article.) To date, the most complete year of data availability is 2012, with particle number size distributions available at 14 sites. The longest continuous time series are available for particle number size distributions in Dresden-Nord and Melpitz.

It needs to be mentioned that certain data sets prior to 2009 were still processed under former data processing standards. This is a result of the EBAS repository emerging from projects that go back to the early 2000s. These older particle number size distribution data sets lack, particularly, the corrections for the losses in the internal tubing of the mobility particle size spectrometers. These losses come into effect mainly for particle diameters below 30 nm, and inconsistencies might occur when older and modern data sets are directly compared. The older data sets are clearly marked on the download page. It is planned that the old data sets will be updated to modern standards.

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8 Conclusions

The co-operative German Ultrafine Aerosol Network (GUAN) delivers atmospheric particle number size distributions and equivalent black carbon concentrations. Data are transferred at regular intervals into the World Data Center for Aerosols repository EBAS (doi:10.5072/guan). These continuously measured data are expected to provide the basis towards a better scientific understanding of sub-µm aerosol processes in the troposphere addressing questions related to both, human particle exposure and climate-relevant effects. Particle number size distributions down to 5 nm allow to study the emission and formation processes of atmospheric ultrafine particles (e.g., Ma and Birmili, 2015). Black carbon, on the other hand, is an essential parameter that is linked to atmospheric particulate light absorption (Bond et al., 2013; Nordmann et al., 2014). Number size distributions of refractory particles (300 °C) represent particle cores that are of likely relevance for health studies, and which have been associated with harmful soot particles (Nordmann et al., 2009; Poulain et al., 2014).

The selection of GUAN's measurement sites covers a continuum of exposure situations between roadside sites, urban background sites, rural background sites, and a high Alpine mountain site. In association with partner projects, GUAN has implemented a high degree of harmonisation of instrumentation, operating procedures, and data evaluation procedures. The GUAN data have already proved suitable for the validation of atmospheric dispersion and process simulations involving atmospheric aerosols. Examples for their successful use include the validation of a global chemical transport model (Reddington et al., 2011), a regional-scale radiative transfer model (Nordmann et al., 2014; Chen et al., 2015), a street canyon aerosol dynamics model (Toenges-Schuller et al., 2015), and statistical prediction tools for particle number size distributions in rural and urban environments (Mølgaard et al., 2013; von Bismarck-Osten et al., 2015).

The high standards of the GUAN data make them suitable for use in cross-sectional air quality and health studies, particularly alleviating the lack of health studies using BC

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and ultrafine particles as exposure variables. In the future, the data set also might serve as an excellent basis for the future discussion of legislative regulation of (ultrafine) particle number and/or BC mass concentrations in ambient air.

Appendix A: Correction formula for the aethalometer in Augsburg

At Augsburg, an aethalometer (Type 8100, Thermo Fisher Scientific Inc.) is deployed using a cut-off of 2.5 µm. Similarly to the MAAP, the Type 8100 aethalometer evaluates the attenuation of transmitted light, although at a different wavelength ($\lambda = 880$ nm) and using a different mass absorption cross section ($16.6\,m^2\,g^{-1}$). To validate the aethalometer's performance, we compared the instrument in Augsburg during one month (9 September–8 October, 2008) against a MAAP instrument (Birmili et al., 2010a). The intercomparison suggested that the aethalometer values are biased by a value around $+0.3\,\mu g\,m^{-3}$ at the zero end, while the mean concentrations during the period matched rather precisely (MAAP: $1.89\,\mu g\,m^{-3}$, aethalometer: $1.87\,\mu g\,m^{-3}$). The relation to determine standardised (i.e. MAAP-based) black carbon concentrations from the aethalometer is as follows:

$$BC_{MAAP} = 1.111 \cdot BC_{aeth} - 0.18 \quad (A1)$$

For aethalometer concentrations below $1.36\,\mu g\,m^{-3}$, a squared fit represents the data more accurately:

$$BC_{MAAP} = -0.033 \cdot BC^2_{aeth} + 1.335 \cdot BC_{aeth} - 0.43 \quad (A2)$$

Both fits show a high measure of determination (R^2) of 0.96.

The Supplement related to this article is available online at doi:10.5194/essdd-8-935-2015-supplement.

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Table 1. Atmospheric measurement sites in GUAN, in alphabetic order.

No.	Name	EBAS Code	Site operator	Type	Altitude	Location	Site and/or data set description
1	Annaberg-Buchholz	DE0061B	LfULG	urban background	545 m	50°34′18″ N, 12°59′56″ E	Schladitz et al. (2015)
2	Augsburg	DE0062B	HMGU/UA	urban background	485 m	48°21′29″ N, 10°54′25″ E	Pitz et al. (2008); Gu et al. (2012)
3	Bösel (Südoldenburg)	DE0056R	GAA	rural	17 m	52°59′53″ N, 07°56′34″ E	Asmi et al. (2011)
4	Dresden-Nord	DE0063K	LfULG	roadside	116 m	51°03′54″ N, 13°44′29″ E	Löschau et al. (2010); Birmili et al. (2013)
5	Dresden-Winckelmannstr.	DE0064B	LfULG	urban background	120 m	51°02′10″ N, 13°43′50″ E	Löschau et al. (2012)
6	Hohenpeissenberg	DE0043G	DWD	rural (mountain)	980 m	47°48′06″ N, 11°00′34″ E	Birmili et al. (2003)
7	Langen	DE0065B	UBA	urban background	130 m	50°00′18″ N, 08°39′05″ E	Gerwig et al. (2014); UBA (2013)
8	Leipzig-Eisenbahnstr.	DE0066K	TROPOS	roadside	120 m	51°20′45″ N, 12°24′23″ E	Voigtländer et al. (2006); Costabile et al. (2009)
9	Leipzig-Mitte	DE0067K	LfULG	roadside	111 m	51°20′39″ N, 12°22′38″ E	Engler et al. (2012); Rasch et al. (2013)
10	Leipzig-Tropos	DE0055B	TROPOS	urban background	126 m	51°21′10″ N, 12°26′03″ E	Costabile et al. (2009); Ma and Birmili (2015)
11	Leipzig-West	DE0068B	LfULG	urban background	122 m	51°19′05″ N, 12°17′51″ E	Löschau et al. (2012); Rasch et al. (2013)
12	Melpitz	DE0044R	Tropos	rural	84 m	51°31′32″ N, 12°55′40″ E	Engler et al. (2007); Ma et al. (2014)
13	Mülheim-Styrum	DE0069B	LANUV/IUTA	urban background	37 m	51°27′17″ N, 06°51′56″ E	Beuck et al. (2011)
14	Neuglobsow	DE0007R	UBA	rural	70 m	53°08′28″ N, 13°01′52″ E	UBA (2013)
15	Schauinsland	DE0003R	UBA	rural (mountain)	1205 m	47°54′49″ N, 07°54′29″ E	UBA (2013); Asmi et al. (2011)
16	Waldhof	DE0002R	UBA	rural	75 m	52°48′04″ N, 10°45′23″ E	UBA (2013); Asmi et al. (2011)
17	Zugspitze (Schneefernerhaus)	DE0054R	UBA/DWD	Alpine mountain	2650 m	47°25′00″ N, 10°58′47″ E	Birmili et al. (2009b, 2010b)

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Table 2. Technical features of GUAN instrumentation. Mobility particle size spectrometers follow the TROPOS design (Sect. 4.1.1) unless mentioned otherwise. Two types of thermodenuders are used to measure non-volatile size distributions are used (Sect. 4.2).

No.	Name	Type	Inlet height above ground	Particle mobility size spectrometer type	Size range	Thermodenuder	eBC instrument	eBC cut-off size
1	Annaberg-Buchholz	portacabin	4 m	SMPS	10–800 nm	–	MAAP	PM_1
2	Augsburg	portacabin	4 m	TSMPS	5–800 nm	Wehner et al. (2002)	Aethalometer	$PM_{2.5}$
3	Bösel (Südoldenburg)	portacabin	4 m	SMPS	10–800 nm	simplistic type	MAAP	PM_{10}
4	Dresden-Nord	portacabin	4 m	TSMPS	5–800 nm	–	MAAP	PM_1
5	Dresden-Winckelmannstr.	portacabin	4 m	SMPS	10–800 nm	–	MAAP	PM_1
6	Hohenpeissenberg	building	12 m	SMPS	10–800 nm	simplistic type	MAAP	PM_{10}
7	Langen	portacabin	14 m	SMPS (TSI 3936)	10–600 nm	–	–	PM_1
8	Leipzig-Eisenbahnstr.	building	6 m	TDMPS	5–800 nm	Wehner et al. (2002)	MAAP	PM_1
9	Leipzig-Mitte	portacabin	4 m	TDMPS	5–800 nm	–	MAAP	PM_{10}
10	Leipzig-Tropos	portacabin	16 m	TDMPS	5–800 nm	Wehner et al. (2002)	MAAP	PM_{10}
11	Leipzig-West	portacabin	4 m	TDMPS	10–800 nm	–	MAAP	PM_{10}
12	Melpitz	portacabin	4 m	TSMPS	5–800 nm	Wehner et al. (2002)	MAAP	PM_{10}
13	Mülheim-Styrum	portacabin	4 m	SMPS (TSI 3936)	14–750 nm	–	–	PM_{10}
14	Neuglobsow	building	6 m	SMPS	10–800 nm	–	MAAP	PM_{10}
15	Schauinsland	building	6 m	SMPS	10–800 nm	simplistic type	MAAP	PM_{10}
16	Waldhof	building	6 m	SMPS	10–800 nm	–	MAAP	PM_{10}
17	Zugspitze (Schneefernerhaus)	building	6 m	SMPS (TSI 3936)	10–600 nm	simplistic type	MAAP	PM_{10}

Table 3. Co-location of GUAN measurements with other continuous aerosol and air pollutant measurements. The core of GUAN measurements include: sub-µm particle number size distributions (PNSD), sub-µm particle number size distributions of non-volatile particles (NV-PNSD), and equivalent black carbon mass concentrations (eBC). Additional continuous measurements may include: total particle number concentration (TNC) – measured by condensation particle counters, coarse particle number size distribution CPNSD – using an aerodynamic particle sizer (APS) or an optical particle counter (OPC), Nanoparticle Surface Area (NSA) – using a NSAM monitor, aerosol scattering coefficient (σ_p) – using a nephelometer, PM_x particle mass concentrations, and basic meteorological parameters (Meteo) including T, RH, wind speed, wind direction, global radiation, precipitation. These additional data need to be obtained directly from the operator of the respective measurements.

No.	Name	PNSD	NV-PNSD	eBC	TNC	CPNSD	NSA	σ_p	PM_{10}	$PM_{2.5}$	NO_x	SO_2	CO	O_3	CO_2	Meteo
1	Annaberg-Buchholz	•		•					•	•	•	•		•		•
2	Augsburg	•	•	•	•	•			•	•						•
3	Bösel (Südoldenburg)	•	•	•					•		•	•	•	•		•
4	Dresden-Nord	•		•					•		•			•		•
5	Dresden-Winckelmannstr.	•		•					•	•	•	•		•		•
6	Hohenpeissenberg	•	•	•	•	•		•	•	•	•	•	•	•	•	•
7	Langen	•			•		•									•
8	Leipzig-Eisenbahnstr.	•	•	•												•
9	Leipzig-Mitte	•		•		•			•	•	•	•				•
10	Leipzig-Tropos	•	•	•							•	•		•		•
11	Leipzig-West	•		•					•	•	•					•
12	Melpitz	•	•	•		•		•	•	•	•	•		•		•
13	Mülheim-Styrum	•					•		•	•	•	•	•	•		•
14	Neuglobsow	•		•					•		•	•	•	•		•
15	Schauinsland	•	•	•		•			•		•	•	•	•	•	•
16	Waldhof	•		•					•		•	•	•	•	•	•
17	Zugspitze (Schneefernerhaus)	•	•	•	•	•		•	•	•	•	•	•	•	•	•

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Table 4. Associations of GUAN stations with other air quality networks, infrastructure and research projects. For further abbreviations, see Table 5.

No.	Name	WMO-GAW[a]	EMEP[b]	UBA	ACTRIS[c]	AQMNS[d]	UFIREG[e]	LLEZ[f]	More
1	Annaberg-Buchholz					•		UltraSchwarz[g]	
2	Augsburg						•		KORA[h]
3	Bösel (Südoldenburg)	regional station							LÜN[i]
4	Dresden-Nord					•		•	
5	Dresden-Winckelmannstr.					•	•	•	
6	Hohenpeissenberg	global station	•		•				DWD, VAO[j]
7	Langen			•					
8	Leipzig-Eisenbahnstr.							•	
9	Leipzig-Mitte					•		•	
10	Leipzig-Tropos	regional station						•	
11	Leipzig-West					•		•	
12	Melpitz	regional station	•		•			•	
13	Mülheim-Styrum								LUQS[k]
14	Neuglobsow	regional station	•	•					
15	Schauinsland	regional station	•	•					
16	Waldhof	regional station	•	•					GMOS[l]
17	Zugspitze (Schneefernerhaus)	global station		•					VAO[j]

[a] WMO-GAW: World Meteorological Organization/Global Atmosphere Watch (http://www.wmo.int/pages/prog/arep/gaw/gaw_home_en.html, http://gaw.empa.ch/gawsis).
[b] EMEP: European Monitoring and Evaluation Programme (http://www.emep.int).
[c] ACTRIS: Aerosols, Clouds, and Trace gases Research InfraStructure Network (http://www.actris.eu).
[d] AQMNS: Air quality monitoring network of Saxony (Luftgütemessnetz Sachsen), coordinated by LfULG, operated by BfUL.
[e] UFIREG: Ultrafine particles – an evidence based contribution to the development of regional and European environmental and health policy (http://www.ufireg-central.eu).
[f] LLEZ: Leipzig low emission zone studies (Löschau et al., 2012, 2013, 2014; Rasch et al., 2013).
[g] UltraSchwarz: Ultrafine particles and health in the Ore Mountains district and the region of Usti (*Ultrafeinstaub und Gesundheit im Erzgebirgskreis und Region Usti*) (http://www.ultraschwarz-ziel3.de).
[h] KORA: Cooperative health research in the Augsburg region (http://www.helmholtz-muenchen.de/kora).
[i] LÜN: Air quality monitoring system in Lower Saxony (Luftüberwachungssystem Niedersachsen), operated by GAA.
[j] VAO: Virtual Alpine Observatory, coordinated by the Bavarian Research Alliance (http://www.bayfor.org).
[k] LUQS: Air quality monitoring network of Northrhine-Westfalia (Kontinuierliches Luftmessnetz), operated by LANUV.
[l] GMOS: Global Mercury Observation System (http://www.gmos.eu).

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Table 5. Institutions involved in GUAN, in alphabetic order.

Acronym	Name	Location (all in Germany)
BfUL	State Dept. for Environmental and Agricultural Operations in Saxony (Betriebsgesellschaft für Umwelt und Landwirtschaft)	Radebeul
DWD	German Meteorological Service (Deutscher Wetterdienst)	Hohenpeissenberg
GAA	Labour Inspectorate of Lower Saxony (Staatliches Gewerbeaufsichtsamt)	Hildesheim
HMGU	Helmholtz Zentrum Munich, Institute of Epidemiology II	Neuherberg
IUTA	Institute of Energy and Environmental Technology e.V.	Duisburg
LANUV	State Agency for Nature, Environment and Consumer Protection in North Rhine-Westphalia (Landesamt für Natur, Umwelt und Verbraucherschutz Nordrhein-Westfalen)	Essen
LfULG	Saxon State Agency for the Environment, Agriculture and Geology (Sächsisches Landesamt für Umwelt, Landwirtschaft und Geologie)	Dresden
TROPOS	Leibniz Institute for Tropospheric Research	Leipzig
UA	University of Augsburg	Augsburg
UBA	Federal Environment Agency (Umweltbundesamt)	Dessau-Rosslau

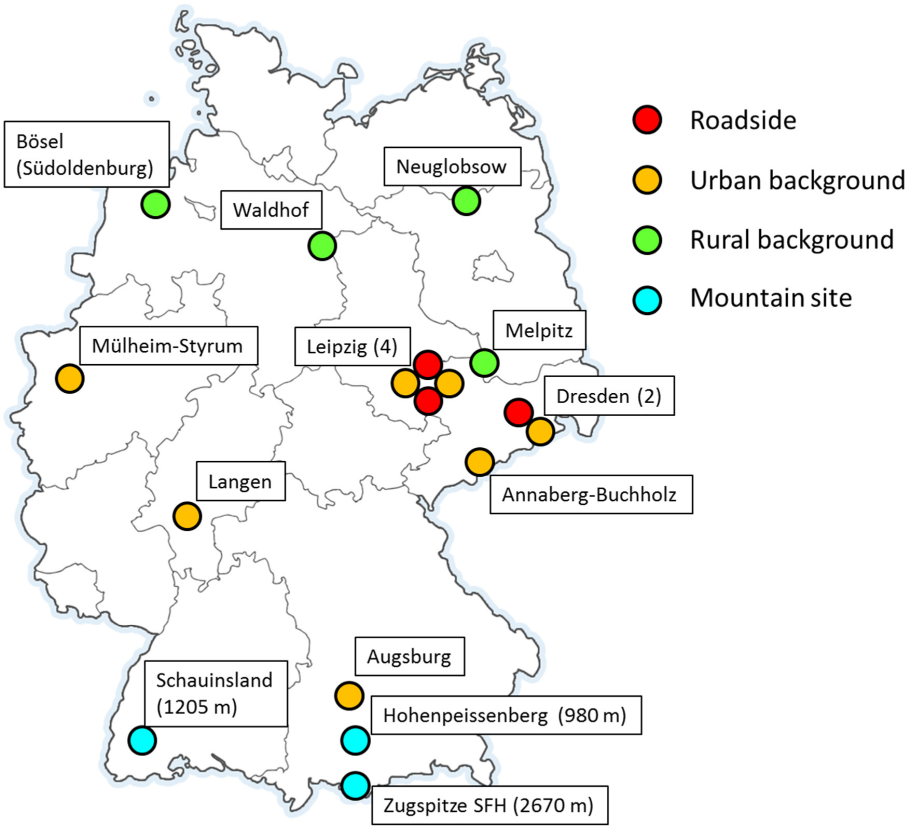

Figure 1. Location of the atmospheric observation sites in the German Ultrafine Aerosol Network (GUAN), currently consisting of seventeen sites. See Table 1 for the full names and characteristics of the sites.

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Site 1: Annaberg-Buchholz (LfULG)

Site 2: Augsburg (HMGU/UA)

Figure 2.

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Site 3: Bösel/Südoldenburg (GAA)

Site 4: Dresden-Nord (LfULG)

Figure 2.

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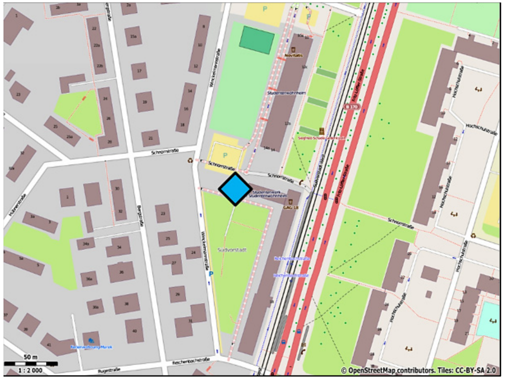
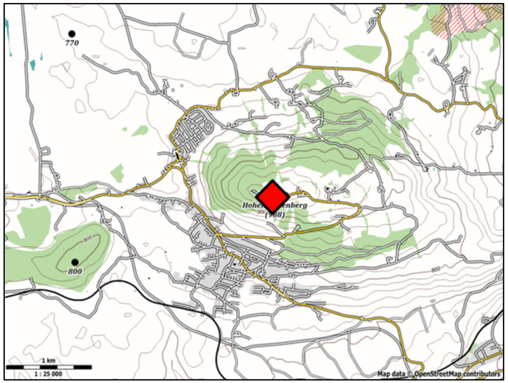


Site 5: Dresden-Winckelmannstrasse (LfULG)



Site 6: Hohenpeissenberg (DWD)

Figure 2.

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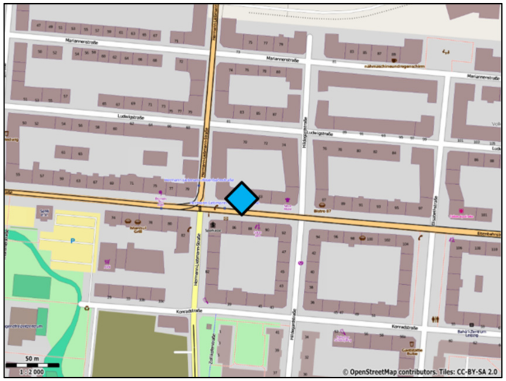
Site 7: Langen (UBA)



Site 8: Leipzig-Eisenbahnstrasse (TROPOS)

Figure 2.

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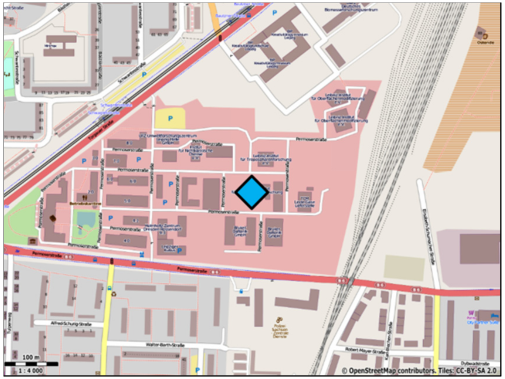
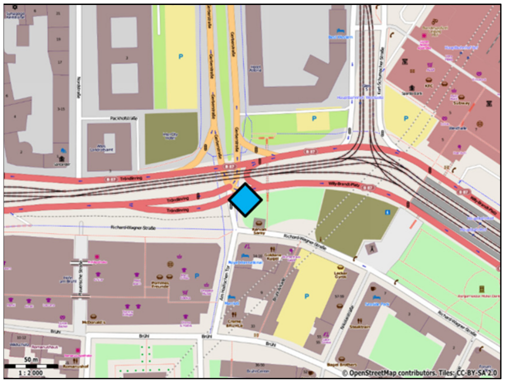


Site 9: Leipzig-Tropos (TROPOS)



Site 10: Leipzig-Mitte (LfULG)

Figure 2.

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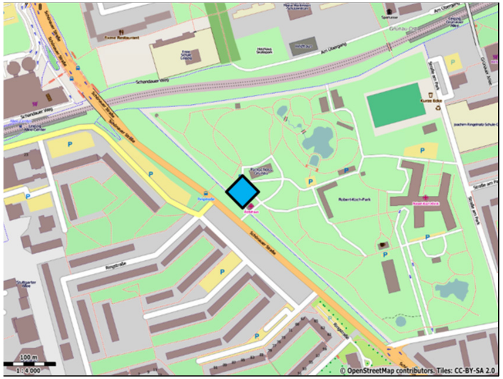
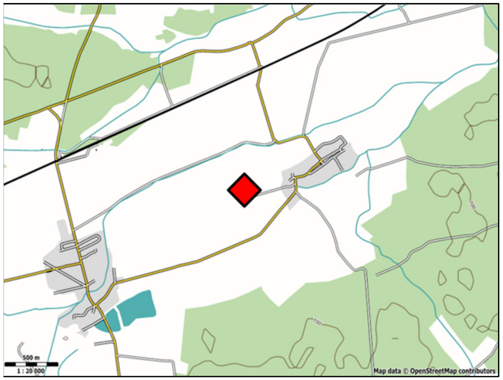


Site 11: Leipzig-West (LfULG)



Site 12: Melpitz (TROPOS)

Figure 2.

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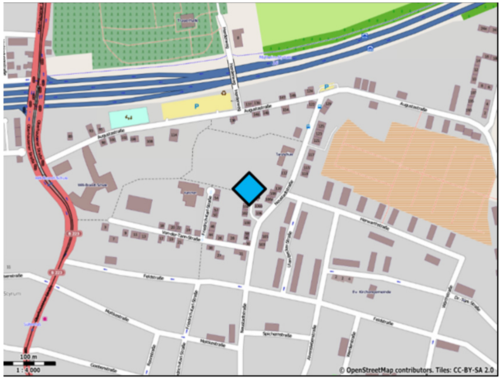
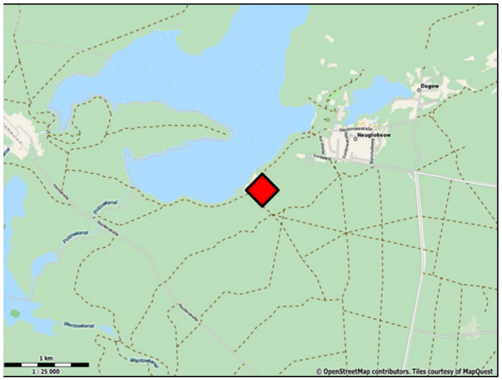


Site 13: Mülheim-Styrum (IUTA/LANUV)



Site 14: Neuglobsow (UBA)

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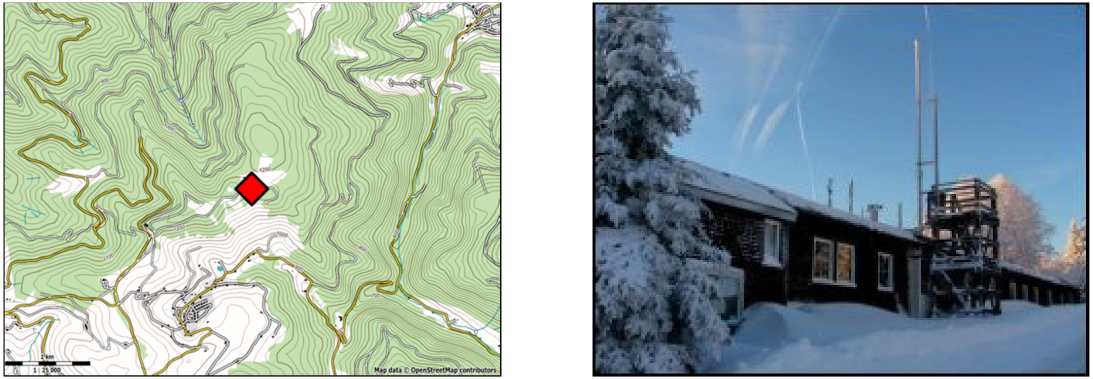

Site 15: Schauinsland (UBA)

Site 16: Waldhof (UBA)

Figure 2.

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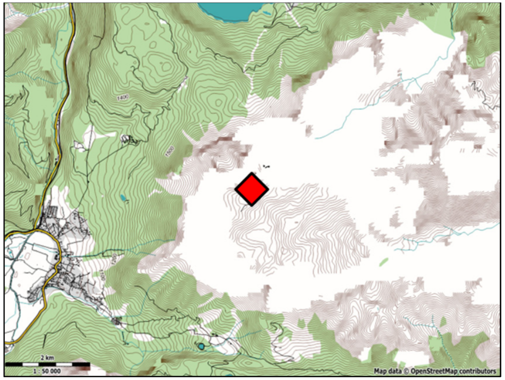

Site 17: Zugspitze/Schneefernerhaus (UBA/DWD)

Figure 2. Illustration of the GUAN measurement sites. The maps show the immediate surroundings of the measurement sites, illustrating the major types of land use (Source: OpenStreetMap, processed by Maperitive V.2.3.22). The location of the measurement site is marked by a diamond. The pictures illustrate the immediate surroundings of the station.

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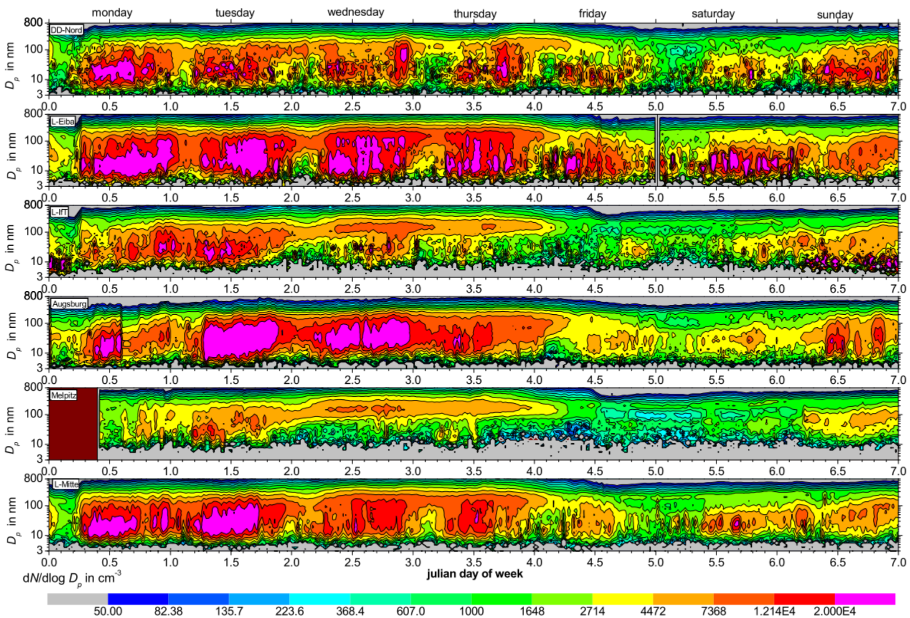

Figure 3. Exemplary contour diagrams of particle number size distributions at six mainly urban GUAN sites (20–26 December 2009). Such diagrams assist the visual quality control of the data.

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