Science of the Total Environment 625 (2018) 752–761

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Long-term observations of the background aerosol at Cabauw, The Netherlands

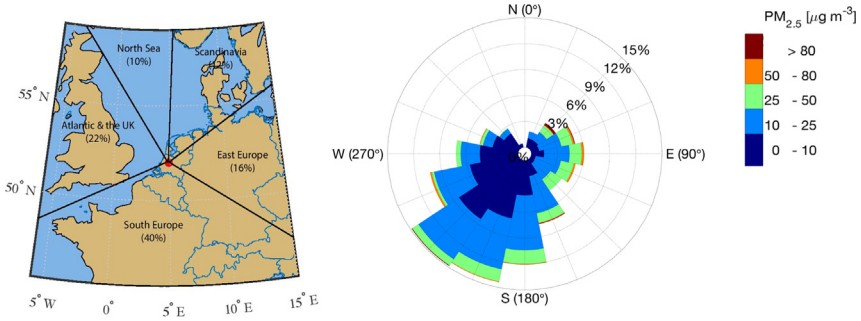
D. Mamali[a,*], J. Mikkilä[b], B. Henzing[c], R. Spoor[d], M. Ehn[b], T. Petäjä[b], H. Russchenberg[a], G. Biskos[a,e,**]

[a] Faculty of Civil Engineering and Geosciences, Delft University of Technology, Delft 2628 CN, The Netherlands
[b] Department of Physics, University of Helsinki, P.O. Box 64, Helsinki 00014, Finland
[c] Netherlands Organisation for Applied Scientific Research TNO, Princetonlaan 6, Utrecht 3508 TA, The Netherlands
[d] National Institute of Public Health and the Environment RIVM, Bilthoven 3720 BA, The Netherlands
[e] Energy Environment and Water Research Center, The Cyprus Institute, Nicosia 2121, Cyprus

HIGHLIGHTS

- We report 8-year-long measurements of that atmospheric aerosol observed at Cabauw.
- $PM_{2.5}$ exhibit an overall decrease with a trend of −0.74 μm m^{-3} $year^{-1}$.
- Particle number concentrations are stable with a mean value of 9.2 × 10^3 # cm^{-3}.
- Highest particle concentrations are observed in winter and spring.
- Particle hygroscopicity shows that the aerosol is often externally mixed.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 20 September 2017
Received in revised form 10 November 2017
Accepted 12 December 2017
Available online 4 January 2018

Editor: Xuexi Tie

Keywords:
Air pollution
Atmospheric particles
$PM_{2.5}$
SMPS
HTDMA

ABSTRACT

Long-term measurements of $PM_{2.5}$ mass concentrations and aerosol particle size distributions from 2008 to 2015, as well as hygroscopicity measurements conducted over one year (2008–2009) at Cabauw, The Netherlands, are compiled here in order to provide a comprehensive dataset for understanding the trends and annual variabilities of the atmospheric aerosol in the region. $PM_{2.5}$ concentrations have a mean value of 14.4 μg m^{-3} with standard deviation 2.1 μg m^{-3}, and exhibit an overall decreasing trend of −0.74 μg m^{-3} $year^{-1}$. The highest values are observed in winter and spring and are associated with a shallower boundary layer and lower precipitation, respectively, compared to the rest of the seasons. Number concentrations of particles smaller than 500 nm have a mean of 9.2 × 10^3 particles cm^{-3} and standard deviation 4.9 × 10^3 particles cm^{-3}, exhibiting an increasing trend between 2008 and 2011 and a decreasing trend from 2013 to 2015. The particle number concentrations exhibit highest values in spring and summer (despite the increased precipitation) due to the high occurrence of nucleation-mode particles, which most likely are formed elsewhere and are transported to the observation station. Particle hygroscopicity measurements show that, independently of the air mass origin, the particles are mostly externally mixed with the more hydrophobic mode having a mean hygroscopic parameter κ of 0.1 while for the more hydrophilic mode κ is 0.35. The hygroscopicity of the smaller particles investigated in this work (i.e., particles having diameters of 35 nm) appears to increase during the course of the nucleation events, reflecting a change in the chemical composition of the particles.

* Corresponding author.
** Correspondence to: G. Biskos, Energy Environment and Water Research Centre, The Cyprus Institute, Nicosia 2121, Cyprus & Faculty of Civil Engineering and Geosciences, Delft University of Technology, Delft 2628 CN, The Netherlands.
E-mail addresses: D.Mamali@tudelft.nl (D. Mamali), G.Biskos@cyi.ac.cy, G.Biskos@tudelft.nl (G. Biskos).

https://doi.org/10.1016/j.scitotenv.2017.12.136
0048-9697/

1. Introduction

Epidemiological and medical studies have established that Particulate Matter (PM) pollution has a strong impact on human health (e.g., US-EPA, 2004; Pope and Dockery, 2006; Baccarelli et al., 2016), with elevated ambient $PM_{2.5}$ concentrations being associated with adverse respiratory effects to chronically ill patients and increased mortality rates for people with respiratory problems (Wichmann and Peters, 2000). In addition, fine particles (i.e., particles with diameters below 1 μm), and especially their smallest fraction that can penetrate deeper in the respiratory tract and reach the alveoli (Asgharian et al., 2014), can enter the blood recirculation system and cause cardiovascular deceases (Zlotkowska, 2015). Model studies have shown that $PM_{2.5}$ concentrations correlate well with morbidity and mortality, causing ca. 3.3 million premature deaths annually around the globe (e.g., Lelieveld et al., 2015).

Apart from being related to adverse health effects, airborne particles emitted from natural and anthropogenic sources can affect the climate of our planet at local, regional and global scales. Air-suspended particles affect climate in a direct way by absorbing and scattering incoming solar radiation (Ramanathan et al., 2001), and indirectly by acting as cloud condensation and ice nuclei (CCN and IN, respectively; Lohmann and Feichter, 2005). Both effects result in an overall cooling of the Earth and together can contribute to a forcing of -1 W/m^2 at the top of the atmosphere with an estimated uncertainty of the order of 100% (Solomon, 2007). This high uncertainty results from the high spatial and temporal variability of the atmospheric aerosol as well as from the poor understanding of key physicochemical transformations that the aerosol particles undergo during their lifetime. To better understand their role on climate, and thus to improve the predictability of atmospheric-climate models, we need long-term measurements of the aerosol properties. Apart from the concentration (by mass and number) of airborne particles, information on the temporal variability of their size and chemical composition is highly required for understanding the processes they are involved in.

The Cabauw Experimental Site for Atmospheric Research (CESAR) in The Netherlands is one of the oldest stations for atmospheric observations in Europe, and one of the core observatories in the global network. CESAR is one of the few observatories where characterization of the atmosphere from the ground up to the top of the atmospheric column is taking place by combining in situ sensors installed at the ground level and at different heights on a 213-m tower, as well as ground-based remote sensing instruments for measuring radiation, wind, turbulence, trace gases, aerosols, and clouds at higher altitudes. Continuous long-term in-situ measurements of the mass concentrations and the number size distributions of atmospheric particles are performed at CESAR since more than a decade. In addition, measurements related to the atmospheric state and various atmosphere-land surface interactions for supporting climate modeling are carried out at CESAR using both in-situ and remote sensing techniques (Russchenberg et al., 2005).

Here we report $PM_{2.5}$ mass concentration and aerosol size distribution measurements conducted at Cabauw from 2008 to 2015, and analyze their trends and seasonal variability. In addition, we provide aerosol hygroscopicity measurements that took place over almost one year (10 months), and link them with the patterns observed in the recorded size distributions. The rest of the paper is organized as follows: Section 2 describes the instrumentation used for the measurements. Section 3 discusses the measurements, including the inter-annual and seasonal variations of $PM_{2.5}$ concentrations and the particle number distributions, as well as the seasonality in particle hygroscopicity. Finally Section 4 summarizes the most important conclusions.

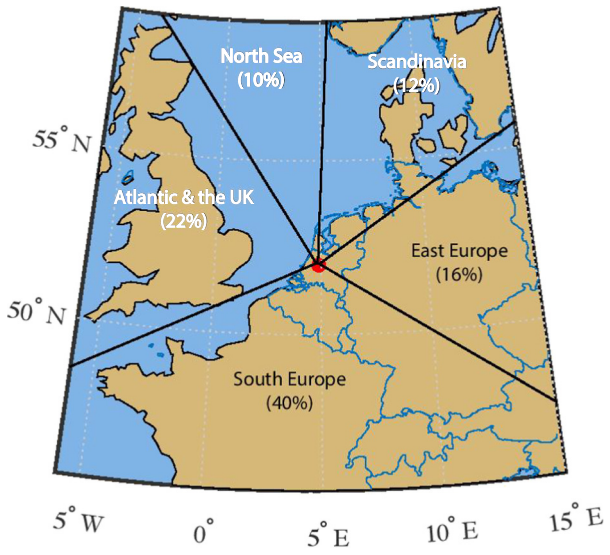


Fig. 1. Location of the CESAR and annual relative frequency of the wind directions observed at the site during the study period.

2. Instrumentation and methods

2.1. The Cabauw Experimental Site for Atmospheric Research (CESAR)

CESAR is a rural site in a region that is one of the most populated areas in Europe (Gallego, 2010). The station, located 50 km away from the coast, is surrounded by grassland and agricultural pastures, and is influenced by maritime and continental environments. The terrain in the region is flat, varying from −3 to 5 m a.s.l. within a radius of 45 km. Fig. 1 shows the location of the station and the frequency of the different wind directions based on measurements recorded between 2008 and 2015 at the tower of the station (cf. Fig. S1 that shows the wind rose diagrams of each year for more details). The site most often receives continental air masses from Southern Europe (40%), with the predominant wind directions being Southwestern (SW), advecting polluted air masses that pass over the neighboring city of Rotterdam. Clean air masses are associated with Northern (N) air originating from the North Sea or Scandinavia only if they do not follow trajectories over Amsterdam and Utrecht; i.e., the other two major neighboring cities to CESAR. Considering that the site mostly receives air masses passing over close-by cities and a dense highway grid located within a radius of 45 km, the station is representative of the wider region.

2.2. Instrumentation

A low-volume sampler operated at a flow rate of 2.3 m^3 h^{-1} and equipped with a $PM_{2.5}$ sampling head (complying with the EN 12341 standards) was deployed to measure the mass concentration of particles smaller than 2.5 μm on quartz filters (Whatman QM-A). All $PM_{2.5}$ measurements were performed by the Dutch National Institute of Public Health and the Environment (RIVM) at the Cabauw Wielsekade station (500 m away from the tower) where air was sampled at a height of 4 m a.s.l. The samples were collected every 24 h, and the mass of the particles deposited on the filters was determined gravimetrically (e.g., Triantafyllou et al., 2016). All the filters were conditioned at 20 ± 1°C and 50 ± 5% RH for at least 48 h before weighing (CEN, 2005). $PM_{2.5}$ measurements were performed on a daily basis during 2009 and 2015, while for the rest of the years investigated in this work, data were available every second day. During days without $PM_{2.5}$ measurements, the quartz filters