Science of the Total Environment 511 (2015) 723–737

Contents lists available at ScienceDirect

Science of the Total Environment

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

A long-term study of new particle formation in a coastal environment: Meteorology, gas phase and solar radiation implications

M. Sorribas [a,b,*], J.A. Adame [c], F.J. Olmo [a,b], J.M. Vilaplana [c], M. Gil-Ojeda [c], L. Alados-Arboledas [a,b]

[a] Department of Applied Physics, University of Granada, Granada, 18071, Spain
[b] Andalusian Institute for Earth System Research (IISTA), University of Granada, 18006, Spain
[c] 'El Arenosillo' — Atmospheric Sounding Station, Atmospheric Research and Instrumentation Branch, National Institute for Aerospace Technology (INTA), Mazagón, Huelva, 21130, Spain

HIGHLIGHTS

- New particle formation (NPF) was observed over 4-years in the South-western Spain.
- New particle formation events showed a frequency of 24% of the total days analysed.
- NPF was more frequent in cold seasons and less frequent in summer time.
- NPF parameters were different under synoptic or regional-scale atmospheric patterns.
- Formation and growth rates were dependent on the solar radiation and gas levels.

ARTICLE INFO

Article history:
Received 25 April 2014
Received in revised form 28 November 2014
Accepted 2 December 2014
Available online 23 January 2015

Editor: Lidia Morawska

Keywords:
New particle formation
Meteorological variables
Solar radiation
Trace gases
Formation and growth rates

ABSTRACT

New particle formation (NPF) was investigated at a coastal background site in Southwest Spain over a four-year period using a Scanning Particle Mobility Sizer (SMPS). The goals of the study were to characterise the NPF and to investigate their relationship to meteorology, gas phase (O_3, SO_2, CO and NO_2) and solar radiation (UVA, UVB and global). A methodology for identifying and classifying the NPF was implemented using the wind direction and modal concentrations as inputs. NPF events showed a frequency of 24% of the total days analysed. The mean duration was 9.2 ± 4.2 h. Contrary to previous studies conducted in other locations, the NPF frequency reached its maximum during cold seasons for approximately 30% of the days. The lowest frequency took place in July with 10%, and the seasonal wind pattern was found to be the most important parameter influencing the NPF frequency. The mean formation rate was 2.2 ± 1.7 cm^{-3} s^{-1}, with a maximum in the spring and early autumn and a minimum during the summer and winter. The mean growth rate was 3.8 ± 2.4 nm h^{-1} with higher values occurring from spring to autumn. The mean and seasonal formation and growth rates are in agreement with previous observations from continental sites in the Northern Hemisphere. NPF classification of different classes was conducted to explore the effect of synoptic and regional-scale patterns on NPF and growth. The results show that under a breeze regime, the temperature indirectly affects NPF events. Higher temperatures increase the strength of the breeze recirculation, favouring gas accumulation and subsequent NPF appearance. Additionally, the role of high relative humidity in inhibiting the NPF was evinced during synoptic scenarios. The remaining meteorological variables (RH), trace gases (CO and NO), solar radiation, PM_{10} and condensation sink, showed a moderate or high connection with both formation and growth rates.

1. Introduction

Atmospheric particles contribute to light absorption, scattering of solar radiation and cloud formation, which are key processes of the global climate system. Homogeneous nucleation of supersaturated vapour plays an important role in the atmospheric particle concentration. Although the impact of secondary particle formation on the global climate is highly uncertain, it could be potentially large and opposite of the warming effect of greenhouse gases (IPCC, 2014). Furthermore, the seasonal total concentration cycle is better simulated by including the nucleation mechanism than by increasing emissions from primary sources (Spracklen et al., 2010). In recent years, the potential effect of aerosols on the climate has been corroborated by studies linking new particle formation (NPF) events with modifications to aerosol scattering processes (Shen et al., 2011) and with the aerosol number acting as

* Corresponding author at: Department of Applied Physics, University of Granada, Granada, 18071, Spain.
E-mail address: sorribas@ugr.es (M. Sorribas).

http://dx.doi.org/10.1016/j.scitotenv.2014.12.011
0048-9697/

cloud condensation nuclei (CCN) (Yum et al., 2007; Spracklen et al., 2008).

NPF through homogeneous gas-phase processes, followed by the condensation growth of the freshly nucleated particles to detectable sizes, occurs frequently in the atmosphere (e.g., Kulmala et al., 2004a). The particle formation mechanisms have been studied in detail (e.g., Kulmala et al., 2004a,b, 2006), but the number of global observations is still very limited. These measurements have been performed over short periods (e.g., Jeong et al., 2010; Mahajan et al., 2011; Young et al., 2013) or as part of long-term monitoring programs (e.g., Dal Maso et al., 2005; Hirsikko et al., 2012; Cusack et al., 2013). Although these studies have been conducted, the influence on the NPF processes of the nucleation mechanisms, chemical species or meteorological variables has not been completely clarified. The most previously studied mechanisms examined include binary nucleation by water and sulphuric acid (e.g., Nilsson and Kulmala, 1998), ternary nucleation that involucrates water, sulphuric acid and ammonia (e.g., Korhonen et al., 1999) and ion-induced nucleation (e.g., Mahajan et al., 2011).

Although studies of NPF events based on long-term observations have increased in Europe in recent years, very few studies have been conducted in Spain. A previous study focusing on the daily and seasonal variability of particle number size distribution and the importance of NPF events in the absence of a significant condensation sink was conducted in a regional background area in Northwest Spain (Cusack et al., 2013). Additionally, a climatology study of NPF events in the subtropical North Atlantic free troposphere of Tenerife Island was presented in García et al. (2014), which evinced a clearly marked NPF season from May to August with an occurrence frequency of approximately 55% of total days/months.

A preliminary study of NPF events in Southwest Spain was reported by Sorribas et al. (2011). This work stated that nucleation events segregate depending on the wind direction during the hours preceding the nucleation burst and in terms of the source region. Based on these more recent results, the goals of the present paper are as follows: (1) to examine the long-term occurrence and duration of NPF in this coastal area, (2) to explore the formation and growth rates of newly formed particles, (3) to determine the meteorological variable values and trace gas concentrations under which NPF events occur, and (4) to determine the variables that influence nucleation and growth rates.

2. Experimental setup

2.1. Sampling site

The 'El Arenosillo' Atmospheric Sounding Station is located in Southwest Spain along the Atlantic Coast and close to the Mediterranean Sea and the North African Coast (Fig. 1). This observatory delivers data to the World Ozone and Ultraviolet Radiation Data Centre (WOUDC) and World Data Centre for Aerosols (WDCA) of the Global Atmosphere Watch (GAW) program developed by the World Meteorological Organization (WMO). The most recent results regarding this observatory can be found in Prats et al. (2008), Córdoba-Jabonero et al. (2011), Sorribas et al. (2011), Antón et al. (2012) and Adame et al. (2014) among others.

The measurement site is situated in the protected rural environment of the Doñana National Park and is less than 1 km away from the coastline. The sector lying north-north-west and east-south-east (NNW and ESE) (Fig. 1) has been identified as the 'sector with *P*redominance of *B*iogenic *E*missions' (Adame et al., 2014), and it will be referred to throughout this work as the PBE sector. The majority of the land coverage in the PBE sector is composed of forest (60.2%), followed by crops (35.5%), urban and industrial activities (3.9%) and water environments (0.4%). Additionally, the land coverage beside the PBE sector and Atlantic Coast is also composed of the forest area, but other anthropogenic emissions from Huelva City and the industrial and dock areas surrounding the city could alter the levels of biogenic emissions. The PBE sector was selected based on the requirements of our study, which will be presented in greater detail in Section 3.1.

Regarding the main aerosol types, typical atmospheric synoptic patterns over the sampling site cause the arrival of coastal marine, continental and desert dust air masses (Hernández-Ceballos et al., 2013). Additionally, as the sampling site is located in a coastal area, frequent mesoscale processes can develop, inhibiting air mass renovation and favouring gas and aerosol accumulation. Adame et al. (2010) identified the existence of two sea–land breeze patterns called pure and non-pure breezes. In this study, only the scenarios under pure breeze (a typical coastal recirculation process) have been considered. The criterion applied for identification of these processes is the same as used in the previous mentioned work. This pattern is characterised by wind occurring during the land regime from the PBE sector (land to sea and perpendicular to

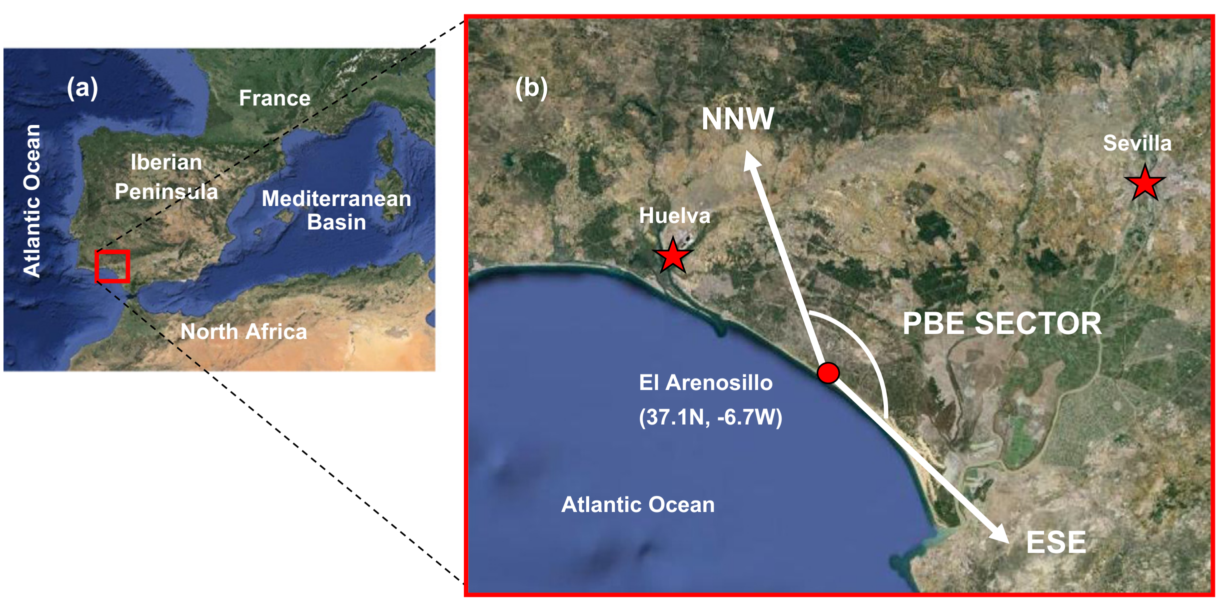

Fig. 1. (a) Sampling area in the Southwest of Europe and (b) El Arenosillo Station and the *P*redominance of *B*iogenic *E*missions (PBE) sector, which covers NNW to ESE in a clockwise direction.