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1. INTRODUCTION

The interest on atmospheric new particle formation (NPF) has increased enormously in recent years because of the high importance that ultrafine particles (with <100 nm in diameter) play on the climate system, air quality and human health. Ultrafine particles originate from anthropogenic activity (industries, road traffic, aircraft, ships, and combustion processes) and are also produced in the atmosphere as secondary particles by gas-to-particle conversion. Anthropogenic precursor vapours, after oxidation in the atmosphere, can form small particles or clusters approximately 1 nm in diameter, which may then grow rapidly to larger sizes in the range of 10–50 nm (Kulmala et al., 2012). It is estimated that NPF is an important source of ultrafine particles, which provides more than half of the global cloud condensation nuclei (Gordon et al., 2017). Atmospheric NPF is a global phenomenon, largely observed and documented in a wide variety of environments and climatic conditions. Together with intense solar radiation, low concentrations of preexisting aerosol particles, high concentrations of gaseous precursors (e.g., SO_2 and VOC) may all create the suitable conditions to trigger the formation process and the growth of new particles. In most cases, NPFs happen as regional events covering large spatial areas in background and remote sites, however, in several condition also urban nucleation is observed.
Although numerous results were obtained, many questions remain still open and, in this sense, the statistical analysis of time series that span over long time periods and in different environmental conditions could be a useful approach. The aim of this report is to identify and characterize the conditions that trigger NPF in an urban background area in Southern Italy through a comprehensive long-term study of NPF and the application/development of a semi-automated method for their identification.

2. MEASUREMENTS

The activities are focused on a flat area in Southeastern Italy recognized as a hot-spot region both in terms of climate change and air-quality being a crossroad where pollutants from different sources (natural and anthropogenic) converge (Dinoi et al., 2021). Climatic features, such as frequent conditions of clear skies during the whole year and intense solar radiation in spring-summer seasons, make this site particularly interesting for the purpose of this study and representative of coastal areas of the central Mediterranean. In this report, a comprehensive dataset of number particle size distributions was analysed, together with meteorological parameters and gaseous pollutant concentrations. Moreover, back-trajectories analysis was used to interpret the impact of meteorology and of the origin of air masses on the frequency of observed NPF events.
All data are collected at the Environmental-Climate Observatory of Lecce, ECO, regional station of GAW/ACTRIS Network (Global Atmosphere Watch Programme/Aerosol, Clouds and Trace Gases Research Infrastructure). The observatory (Fig. 1) is located on the roof of the ISAC-CNR Institute, inside the University Campus (40° 20′ 8″ N, 18° 07′ 28″ E) at about 13 m above the ground level and at about 5 km SW of the town of Lecce. The main aerosol sources in the ECO observatory are the emissions of vehicular traffic and biomass combustions, which add to natural and anthropogenic long-range contributions (Conte et al., 2020).

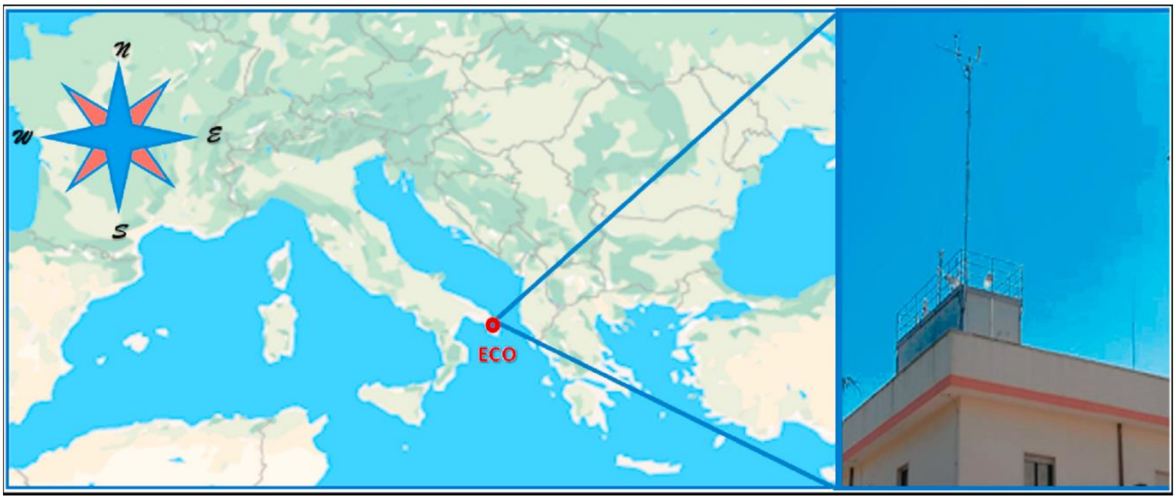

Figure 1: Position of the environmental-climate observatory (ECO) of Lecce.

Aerosol particle number size distributions (PNSD) from 10 to 800 m, with a time resolution of 5 min, were collected using a TROPOS-type custom-built MPSS (mobility particle size spectrometer), designed and manufactured according to EUSAAR/ACTRIS recommendations (Wiedensohler et al., 2012). Classification of NPF events is performed by visual inspection of daily contour plots (Dal Maso et al., 2005). Examining the time evolution of the particle number size distribution, three main classes were detected: NPF events, non-events, and undefined events. NPF events contain cases where a significant increase in the number concentrations of ultrafine particles and growth toward larger diameters was observed for at least 3–4 h continuously, displaying the shape of a "banana". "Non-events" are the days without new particle formation, while "undefined events" group are ambiguous cases.

3. RESULTS

The relative frequency of the different classes (events, non-events, and undefined events) was calculated over 5 years of measurements (2015-2019). In this period, 25 % of days were identified as NPF events, while 4 % as undefined events. The highest frequencies of NPF events were observed in March and September (∼30 %), and the lowest in November and December (12 % −16 %), reflecting a different seasonality of events, more frequent in spring and summer (Fig.2). This is in good agreement with frequencies (10 %–36 %) found in other studies based on long-term measurements and carried out in the Mediterranean area (Hussein et al., 2020; Kalkavouras et al., 2020; Baalbaki et al., 2021). These higher frequnecies in warmer seasons can be ascribed to the higher emissions of biogenic aerosol precursor compounds, to the intensification of the photochemical activity promoted by the higher temperature during the warmer months, and abundance of SO_2, which acts as a precursor of sulfuric acid. (Asmi et al., 2016).

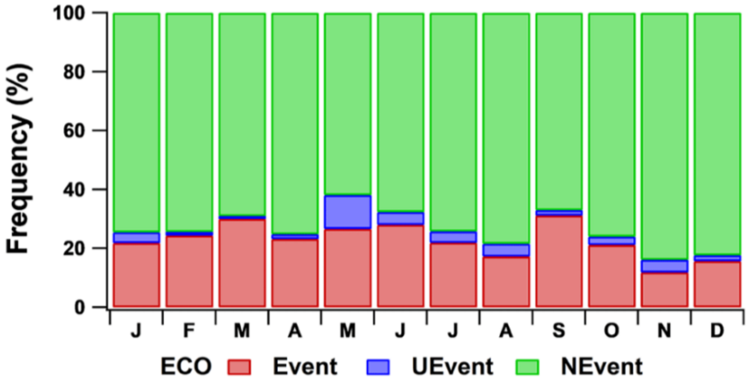

Figure 2: Monthly percentage of occurrence of events (red bars), undefined events (blue bars), and non-events (green bars) related to available measurement days at ECO.

From this perspective, the effects of SO_2 concentrations on the occurrence of NPF were investigated, resulting that NPF days were characterized by higher SO_2 concentrations (~20 %) compared to non-event days. Regarding the meteorological conditions, it was observed that NPF events occurred in conditions of high pressure, moderate wind speed (3–4 m s^{-1}), and low RH (~52 %), about 25 % lower than that on non-event days. This result is not a novelty because lower RH is usually observed during NPF events in both clean and polluted environments (Kerminen et al., 2018).

The study of the back-trajectories associated (Fig. 3) with the NPF events highlighted a prevalent origin of the air masses, both of continental and marine origin, from the north–northwest directions, suggesting that the chemical compounds involved in the NPFs could have been transported by the air masses. In addition, winds from N-NW are generally associated with low relative humidity and this favors NPF events.

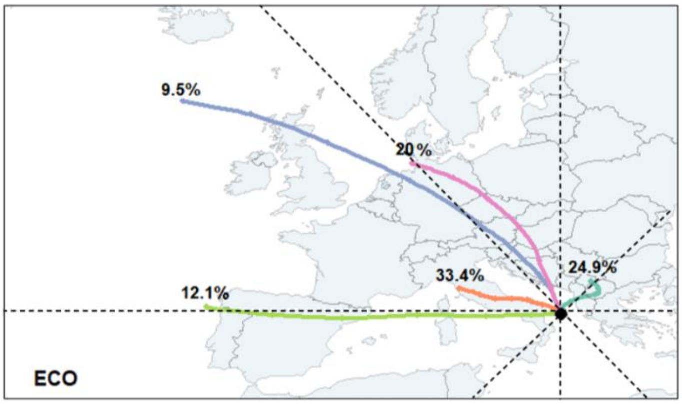

Figure 3: Cluster centroids retrieved from 3 days analytical back trajectories reaching ECO at 500 m a.s.l.

Indeed, the pathways of air masses can contribute significantly to the occurrence of NPF processes (Wonaschütz et al., 2015), because depending on their advection pattern, air masses can be affected by emission sources, and chemical or physical processes able to influence the properties of the pollutants during transportation.

4. AUTOMATIC IDENTIFICATION OF NPF

The traditional approach to individuate and characterise the NPF could be time consuming on long time series and has some limitations. One limitation is that when NPF is relatively weak, or it is influenced by meteorological variability or non-homogeneity of air masses could results in non-classified days (Dal Maso et al., 2005; Kulmala et al., 2012) or incorrectly classified as non-event. Long-term measurements indicate that these cases could be a relevant fraction of all days (Salma and Nemeth, 2019). In addition, the evaluation of the growth rate (GR) can be done only for days during which there is a clear new mode of particles and the size distribution can be followed for several hours with the growth marginally affected by changes in air masses (Kulmala et al., 2023). The recent work of Kulmala et al. (2023) propose a different approach in which, instead of separately evaluating the frequency of NPF and its intensity for a sub-set of days, these two quantities are combined into a probability distribution of the intensity of NPF. This could, in practice, cove all the days. This maybe an approach to be further developed and tested in the project ITINERIS at different observatories.

In this report, in addition to day-by-day visual classification of NPF, a new approach to automatically detect NPF events was tested. Thanks to a collaboration with the EGAR (Environmental Geochemistry and Atmospheric Research) research group, the automatic identification of NPF days is performed using a deep learning model called "ConvNeXt". The model, through convolutional neural network, captures different parameters in the PNSD dataset to process identification and characterization of NPF. In addition to the detection of NPF events, the method can automatically determine the growth rates, and start and end times of NPF events. The first test was performed over a short measurement period of one year and the results were compared with those presented by Dinoi et al. (2021, 2023). Results show that the automatically determined events agree with those visually determined, but essentially only for the events sufficiently strong and homogenous to allow the determination of the specific mentioned parameters. Days during which the NPF is relatively weak or non-homogeneous for various reasons (including weather conditions), were not identified by the model as events. In these cases, the analysis is difficult and those days risk to be classified as undefined days or incorrectly classified as non-event days. This mostly happens with the automatic method but, in some cases, can also happen with the visual inspection.

This difficulty can be partly resolved by making measurements in the size range below 10 nm. Because this will allow to identify better the onset of nucleation that involves small clusters (a few nm) and this size range is less influenced by primary emissions compared to size > 10 nm. Since December 2023, a neutral cluster and air ion spectrometer (NAIS, Airel Ltd., Estonia,) has been operating at the ECO observatory. The instrument measures the mobility distributions of ions and size distribution of aerosol particles in the size range of 0.8–40 nm and 2–40 nm, respectively. This size range is crucial for identifying and characterising NPF events and is very important for studying the beginning of NPF and characterizing the early stages of growth. Thereby, further studies will be conducted during the project on this topic with both visual and automatic identification methods.

5. CONCLUSIONS

In summary, a multi-year dataset was investigated to characterize the new particle formation events at the Environmental-Climate Observatory (ECO) in Lecce. Results allowed to investigate the seasonality and the influence of meteorological conditions, circulation of air masses, and gaseous pollutant concentrations on NPF events frequency and intensity. An automatic method of identification was tested that allowed to retrieve the most intense cases of nucleation. Further, studies will be necessary to investigate longer time periods and other statistical approaches. The new instruments installed for detection of particles with diameter lower than 10 nm will likely help to individuate nucleation events with both manual and semi-automatic methods. The post-processed dataset will be made available on the ITINERIS hub as well as future datasets that will be developed during project implementation.

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