RELAZIONE FINALE PROGRAMMA DI RICERCA

SHORT TERM MOBILITY (STM) 2105

Investigation of new particle formation events and growth of atmospheric aerosol using SMPS measurements

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The main scientific objectives of this study was to identify and characterize the new particle formation events in a suburban background station in the Apulia Region. For this purpose was used a dataset of submicron particle concentrations collected in Lecce during 2015 (January - November) by a Scanning Mobility Particle Sizer (SMPS) realized by Tropos-Leibniz Institute for Tropospheric Research, installed at the Environmental-Climate Observatory of Lecce, regional station of the GAW-WMO network. The identification of new particle formation events was done on a dataset of continuous measurement of particle size distributions, in the range of 10–800 nm, with day-by-day approach, in order to classify each day as either event day or non-event day. In this study were analyzed 262 sampling days and 59 new particle formation (NPF) events were identified.

The intensity and duration of each formation process and growth of the particle was used to classify new particle formation events.

Air mass properties, by means of back trajectories analysis, together with meteorological parameters such as relative humidity and solar radiation were used to explain if and how new particle formation events can be affected by the transport of the air masses of different origin and meteorological parameters.

Data handling und inversion

The TROPOS-SMPS is a mobility particle size spectrometer that measures continuously the particle number size distribution in a size range from 10 to 800 nanometers with a temporal resolution of approximately 5 minutes. Measured data are stored on PC, which is part of the measuring system, as Level_ 0 data set containing all mandatory raw data (00_dia and 00_raw) and system parameters.

Particle number size distributions have to be processed using a linear multiple charge inversion algorithm that performs various additional corrections before yielding the final particle number size distributions (Wiedensohler et al., 2012; Birmili et al, 2015). In this work was applied an evaluation procedure called *DMPS-Inversion-2.13.exe* that has allowed to converted Level_0 data into real particle number size distributions. In this way, the program generated data of the particle number size distribution without technical and final end validation called Level_1 data. The unchecked Level_1 data are stored on a daily basis in the folder *_in2. Data in Level_1 contain processed particle number size distributions with the original time resolution of the instrument. The entire process includes inversion from electrical particle mobility distribution to particle number size distribution (conversion to dN/dlogDp and multiple charge correction), correction for CPC counting efficiency, correction for internal losses due to particle diffusion and correction for particle losses from the aerosol inlet to the instrument. After that, the data were subjected to a technical and final end validation by using the program called *Apply flagging (FLG) to SMPS data.exe.* It converted unchecked *.in2-data into technical and final end validated *.in3–data. Final quality control involved additional visual checks in order to process the data carefully inspecting contour diagrams of the particle number size distribution.

Identifying new particle formation events

The next step has been the identification of new particle formation events (NPF). The classification was done with day-by-day approach, in order to classify each day as either event day or non-event day. The method used in this paper is the procedure suggested by Dal Maso (Dal Maso et al., 2005) where the particle number size distributions were visually inspected determining the occurrence of NPF events.

The day is considered a NPF event day if the formation of new aerosol particles starts in the nucleation mode size range (particles with diameters smaller than 20 nm) and subsequently grows, and the formation and growth is observed over a period >1 hour. In practice, a NPF event can be seen as an increase in the particle concentrations in the smallest channels of the SMPS system. These newly formed particles then experience subsequent growth that can be seen to occur typically at a rate of few nanometers per hour during the rest of the day. If the aerosol size distribution for a given day exhibits these signs, the day can be classified as a typical NPF event day with the typical "banana shape".

New-particle-formation days were classify as class I, class II and undefined event days; the days with no particle formation are classified as non-event days (NE); an example of different classification is shown in Fig.1.

Class I days (Fig. 1a) exhibit new-particle formation and a traceable growth of the nucleation mode to at least 50 nm before the nucleation mode disappears. **Class II** days (Fig. 1b) exhibit new-particle formation and a weak growth; **Undefined events** (Fig. 1c) exhibit particles measured at the smallest sizes of the SMPS but there is either no growth or there is growth followed by shrinking.

The features associated with NPF events are very important for characterizing the NPF. These features include: 1) the start and cut-off (end time) of the particle bursts above the detection limit, 2) the new particle event duration and 3) the NPF rate (cm^{-3} s^{-1}). In addition to NPF days, days with an absence of particles in the nucleation mode are also interesting because they facilitate the study of the cause of NPF through the comparison of the conditions during event and non-event periods.

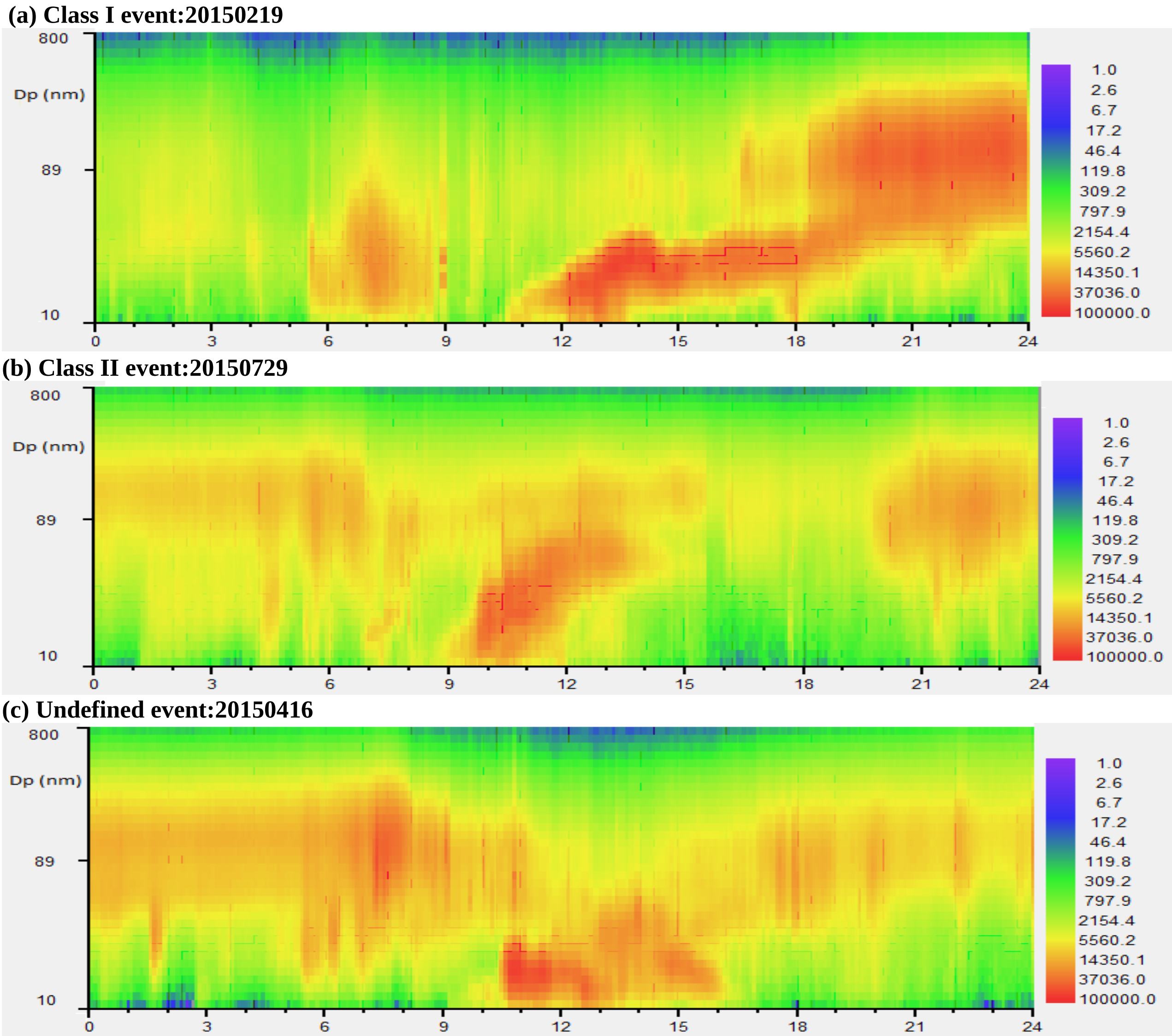

Figure 1. Sample size-distribution time series for a **(a)** class I nucleation day, **(b)** class II nucleation day and **(c)** undefined nucleation day.

In this work a total of 262 days were analyzed; on average, there were 59 events out of which 29 were class I events. On average, the burst was detected by high particle number concentrations between late morning and midday, followed by a subsequent, gradual shift of the new nucleation mode towards larger diameters. Fig. 2 illustrates a new particle formation event. The aerosol number concentration with a diameter of 10-20 nm begin to increase at 10:15 and reaches the peak after just one hour, particles with a diameter 20-30 nm show a sudden increase at 11:10 with an increment of one order of magnitude while the number concentration of aerosols in the diameter of 30-40nm begin to rise at 12:00 reaching to the maximum after 3hours. The aerosol number concentration in the diameter range of 40-50nm shows a small increase at about 14:00 that maintains constant for about 6 hours.

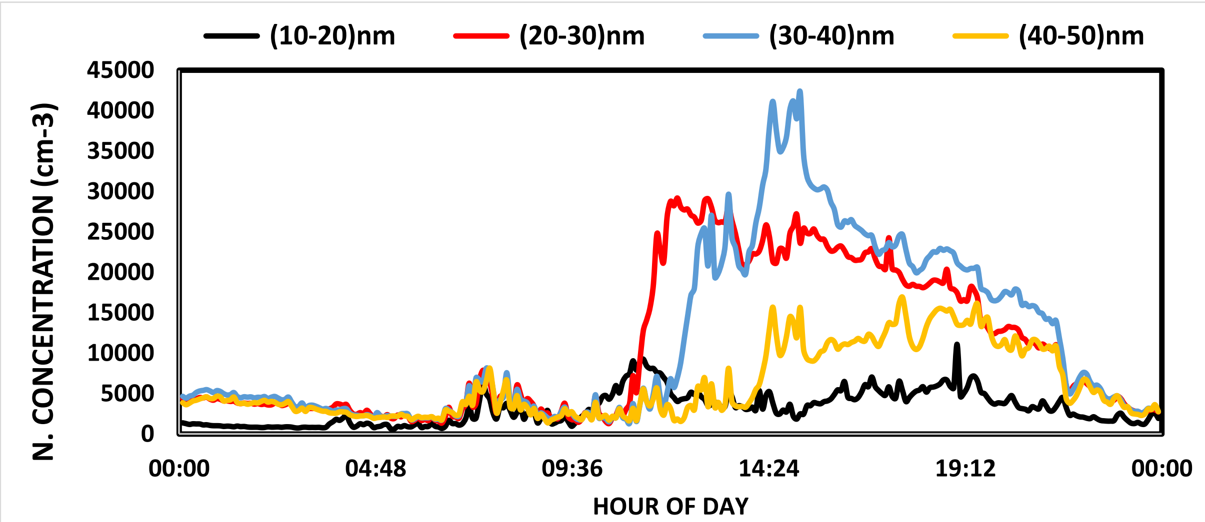

Figure 2. Hourly variation of 4 different sizes aerosol number concentration for a class I nucleation event.

The events classification showed a seasonal trend with higher frequencies of NPF events during summer as we can see in Fig. 3; events fraction distribution has a maximum in May (0.35) and another maximum in September (0.33). The non-event fraction on the other hand is highest in the winter, January and February (0.9–0.8). The undefined day fraction peaked in September (0.3).

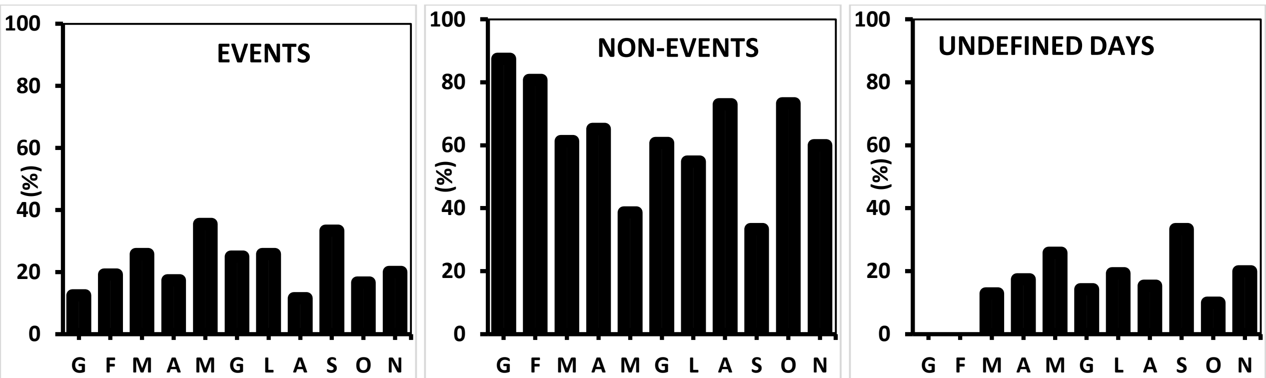

Fig. 3. Mean monthly event, non-event and undefined day fraction during 2015.

This monthly distribution supports statistical studies on NPF events where it was highlighted their connection to days with intense solar radiation (Boy et al., 2002; Birmili et al., 2003). In meteorological terms, solar irradiance causes convection, turbulence, well-mixed boundary layer but it is also used to indicate the reactivity of photochemical reactions. This implies that for the estimation of NPF, data from such well-mixed meteorological situations should be preferred. Fig.4 displays the daily averaged global radiation measured during sampling days.

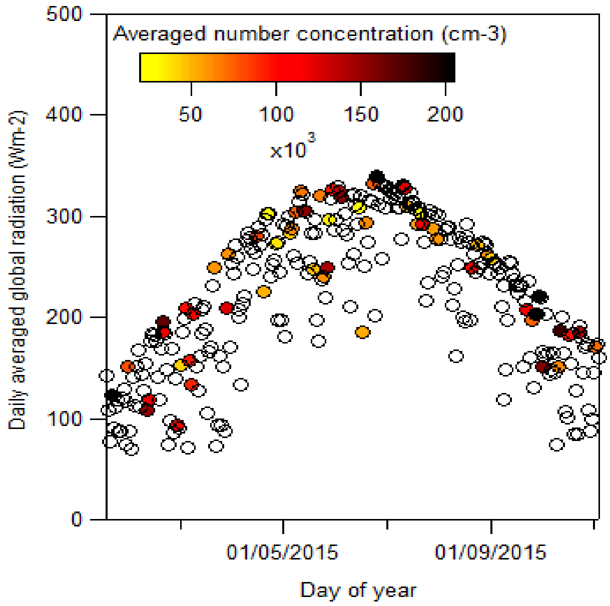

Fig. 4. Daily average global radiation measured during sampling days; days with new particle formation events are marked in color, encoding the daily average number concentration of N [10–20] (cm^{-3}).

Days with new particle formation events are marked in color, encoding the daily average number concentration N considering only particles in nucleation mode size range 10–20 nm. As we can see, NPF events occur almost exclusively on days with a daily average radiation of more than 100 Wm^{-2} and the fraction of event days generally increased with an increasing value of solar radiation even if there are a considerable number of days with high solar radiation not classified as NPF events.

It is well known that there is an anticorrelation between solar radiation and RH, and then an anticorrelation between RH and new particle formation events; indeed high atmospheric relative humidity has proved to be a factor disfavoring NPF (e.g. Birmili and Wiedensohler, 2000; Boy and Kulmala, 2002; Mikkonen et al., 2006; Hamed et al., 2007). In this work, it was not found a significant dependence of NPF events from RH, average values of RH are 62% (45÷87) and 68% (44÷90) for event days and non-event days respectively.

This result underlines that meteorological conditions are not the only key parameters affecting the nucleation events but chemical compounds are needed in order to form a nucleus and contribute to subsequent growth of particles (Conte et al., 2015; Dall'Osto et al., 2015).

Origin of air mass together with meteorological parameters may affect physical and chemical properties of the aerosol (e.g. Van Eijk and De Leeuw, 1992). The probability of occurrence of NPF events at a given location and time depend not only on local emissions, but also on long-range transport (Sogacheva et al., 2007; Tunved et al., 2006). To classify the types of the air masses arriving at sampling site, it was made a back-trajectory analysis using the HYSPLIT single particle Lagrangian transport model developed by NOAA and freely available on the internet (http: //www.arl.noaa.gov/HYSPLIT.php). It has considered one trajectory per day, back trajectories are generated for 24 h prior to their arrival at 500m above ground level of observation site at 12:00 UTC, in the middle of a typical new-particle-formation event. In Fig.5, we can observe that most of back-trajectory (70%) related to NPF events originated from Continental air masses.

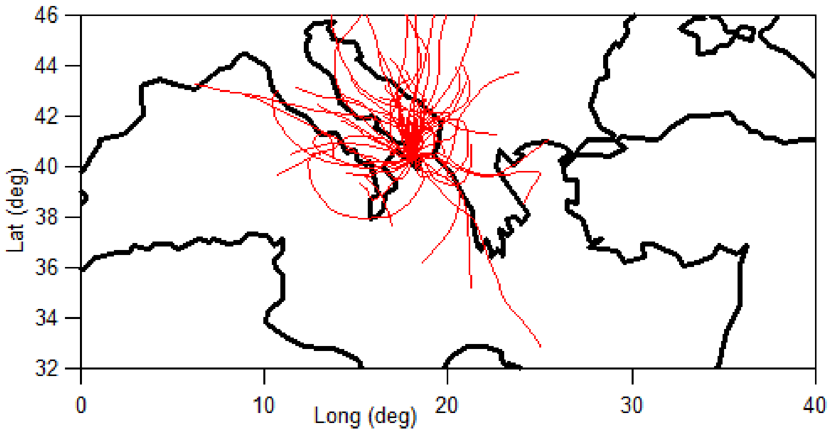

Fig. 5. The 24 h back trajectories arriving during the new particle-formation event day (class I and II), one back trajectory per event.

Before reaching the monitoring site, airflow travelled over Northern and Eastern Europe, densely industrialized and polluted regions. High SO_2 concentration are considered as possible precursors to the formation of new particles by nucleation process. This would seem to suggest that in more polluted continental air masses there are more favorable conditions for new particle formation events.

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