

EXTREMES IN THE CONCENTRATIONS OF CCN
AT MT. MUSALA AND SYNOPTIC CLASSIFICATIONS

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Abstract

According to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change the aerosols are having generally a negative but decreasing in magnitude contribution to the effective radiative forcing (ERF) and, therefore, they act against the global warming. Leading role in this ERF balance play the aerosols involved in aerosol-cloud interaction, named the cloud condensation nuclei (CCN). In the present article we study the distribution of CCN as monitored by the Basic Environmental Observatory Musala in 2016 and their performance in different atmospheric circulations. As first step, we determine the days with extrema in the concentration and sort them separately for minima and maxima using the circulation scheme of Jenkinson–Collinson Types that comprises 26 types of circulations. Thus we determine the most frequent types of circulation occurring in the studied period. Consequently, for each of these days we obtain the backward trajectories using the Hybrid Single-particle Lagrangian Integrated Trajectory model. This permits to match the prevailing types of circulation with the origin of air masses, and, on the other hand, their origin with respect to prevailing height of the trajectory.

Key words: Cloud Condensation Nuclei (CCN), Mt. Musala, HYSPLIT, JCT, IPCC

Introduction. Human activity, and mostly the burning of bio-mass leads to a considerable increase of the aerosol mass in the atmosphere [1]. Aerosols

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influence the radiative forcing of the atmosphere in two ways: directly, through scattering and absorbing radiation and, indirectly, through changing the micro-physical properties of the clouds, their lifetime and albedo. Aerosol particles that take part in the aerosol-cloud interactions are called CCN. The ability of the aerosols to play the role of CCN depends on many factors – concentration, size-distribution, solubility, morphology and chemical composition [2]. A thermodynamic treatment of CCN is provided by KÖHLER in [3].

Knowledge of the spatial and temporal distribution of CCNs in the atmosphere is of utmost importance in order to incorporate their effects into numerical meteorological models at all scales, from regional to global [4].

In general, model simulations report larger effective radiative forcing (ERF) than observational (experimental) studies [5]. However, in the latter, human error also plays a role, so it is not clear whether observation-based estimates are more accurate than numerical models. According to the Sixth Assessment Report of the Intergovernmental Panel on Climate Change (IPCC) [6] aerosols have a negative total contribution to the ERF of -1.1 [-1.7 to -0.4] W/m^2 over the years 1750–2019. About 75–80% of this ERF is due to aerosol–cloud interactions, and the remaining contribution is due to aerosol–radiative interactions. Statistical relationships and effects on clouds due to aerosols show that human activity leads to an increase in the concentration of cloud droplets. However, during the 2014–2019 period considered in the Sixth Assessment Report of the IPCC, the aerosol ERF decreased in magnitude and was estimated at $+0.2$ W/m^2 . This is due to the reduction in global mean aerosol optical depth and the reduction of aerosol emissions and their sources through a number of state and world decrees [7]. Based on these estimates, instead of suppressing warming, clouds will lead to an increase in the global radiation balance in the future, due to the release of more greenhouse gases and fewer aerosols into the atmosphere by human activities.

In the last few years, research on CCN continues by employing different methods. GENZ et al. [8] make an evaluation of CCN and a comparison with the data from Melpitz station, Germany and those from lidars. During the NASA Observations of Aerosols above Clouds and their interactions (ORACLES) project, a strong linear relationship was found between the data from in-situ eruptions and the airborne High Spectral Resolution Lidar 2 measurements [9]. Upper atmospheric CCN measurements are often representative of free troposphere conditions. Therefore, the distribution of CCN along the mountain peaks [10–14] is indicative of the transfer of air masses influencing the respective region.

In this study, the data on the distribution of CCN in 2016, measured at the Basic Ecological Observatory (BEO) Musala ($42^\circ 10' 45''\text{N}$, $23^\circ 35' 07''\text{E}$) were used. BEO is located on Mount Musala – the highest peak in Bulgaria, as well as on the entire Balkan Peninsula, with an altitude of 2925 meters, therefore the site can be considered to be in the free troposphere and far away from the regional contamination for most of the time. Days with minimum and maximum concen-

tration of CCN were identified. They are grouped by the atmospheric circulation of Jenkinson–Collinson Types (JCT) [15], described in detail in STOEV's dissertation [16]. The most common types were separated from the classification and they were compared with data from the backward trajectories of air masses obtained from the Hybrid Single-particle Lagrangian Integrated Trajectory (HYSPLIT) [17] model of the National Oceanic and Atmospheric Administration (NOAA).

Materials and methods. The number concentration of CCN ($\#/cm^3$) was obtained from a continuous-flow streamwise thermal-gradient CCN counter (CCNC, Droplet Measurement Technologies (DMT) Inc., CCN 100) [18] for each day of 2016. It is read directly from the counter every second, at six different supersaturations: 0.13; 0.23, 0.43, 0.53, 0.73 and 1.13%, which switch automatically every 10 minutes from any hour. In the present study, the data from the concentration at 0.43% supersaturation, chosen as representative out of the 6 supersaturations available from the instrument, were used, and it was hourly averaged and then averaged on a diurnal scale. A statistical function, low and upper 95% confidence interval of the mean was used to separate the days in which concentration values below and above the confidence interval of concentrations were reported in the respective month. After that, only the most clearly expressed and significant days of these cases were subjectively separated, taking into account that the concentration change was at least twice compared to the beginning of the increase or decrease and that the concentration on the corresponding day was not statistically similar to the adjacent days. After identifying the days with minima and maxima of CCN concentration, they were mapped to the JCT atmospheric circulation types. The classification includes: 1) eight main directions, west (W), southwest (SW), northwest (NW), north (N), northeast (NE), east (E), southwest (SE) and south (S), 2) one cyclonic type (C), and 3) one anticyclonic type (AC). There are also two additional groups: 1) eight anticyclonic types (AW, ASW, ANW, AN, ANE, AE, ASE, and AS), and 2) eight cyclonic types (CW, CSW, CNW, CN, CNE, CE, CSE, and CS). After grouping, the most common weather types were identified and compared with data extracted from the back trajectories obtained by HYSPLIT. The backward trajectories that reach Mt. Musala every day at 12 UTC are followed back in time for duration of 72 h, a period that describes well the path of air masses over the continent of Europe and is related to the development of atmospheric processes affecting the Balkan Peninsula.

Results. CCN concentration on a mean diurnal scale in 2016. For the needs of the present study, the concentration of CCN was averaged on a mean diurnal scale at a supersaturation of 0.43% for each day of 2016. Then, using a statistical function (lower/upper 95% confidence interval of mean) and after subjective separation of the cases of interest for the present study, the days with extremely high and extremely low concentrations were separated. Their number is summarized in Fig. 1a. The least number of days in a month with extremely low values is in December (zero days), and with extremely high values

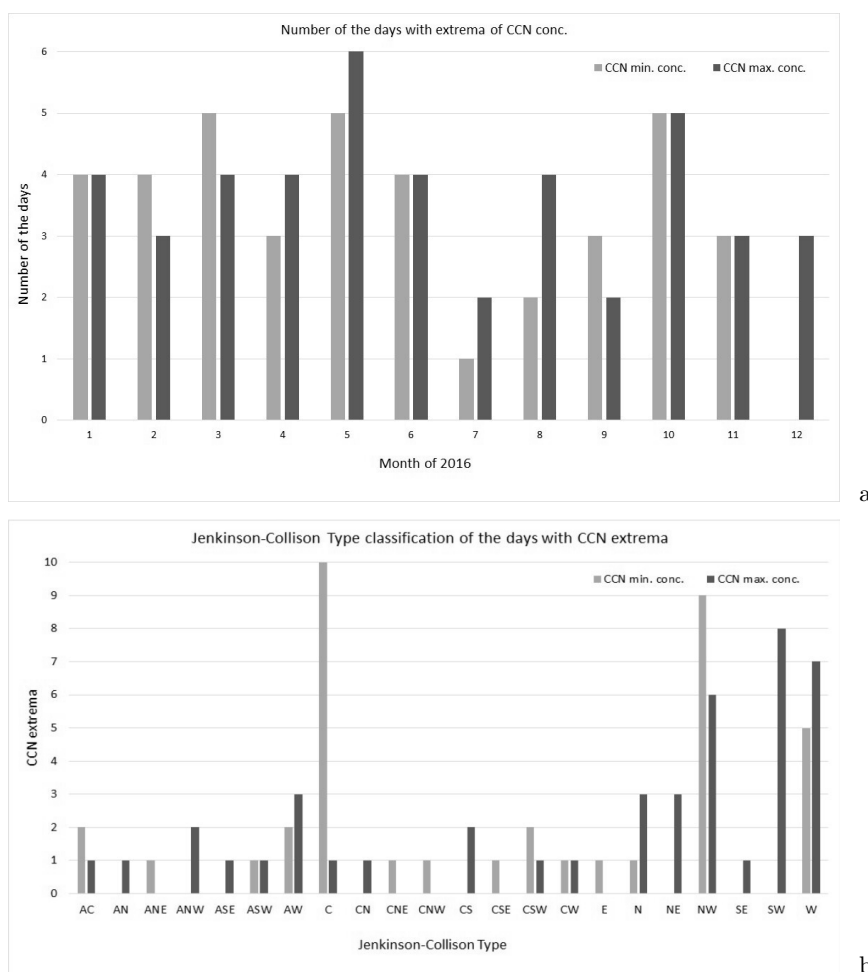


Fig. 1. Monthly distribution of the number of days in 2016 with (a) extremely low (light grey bars) and high (dark grey bars) values of CCN concentration and (b) JCT circulation types

– in July and September (two days for each of the two months). The most cases with extremely low values are in March, May and October (five days each), and with extremely high values – in May (six days). These days, with extremely low and extremely high concentrations of CCN are grouped according to the JCT atmospheric circulation types shown in Fig. 1b. It can be seen that extremely low concentrations of CCN are reported most often in C, NW and W, respectively, 10, 9 and 5 cases, and extremely high concentrations in SW, W and NW, respectively, 8, 7 and 6 cases. I.e. both types of extremes are observed in W and NW JCT types, but differ in the third most commonly reported JCT atmospheric circulation type. This third type is the absolute maximum for the respective type of extremum, C for the maximum low CCN concentration values and SW for the maximum high.

Furthermore, in the cases with extremely high CCN concentration values, only one case of C type (maximum at the extremely low values) was reported, and at the extremely low CCN concentration values, no case of SW type circulation was reported (maximum many times reported at the extremely high values). At the maximum concentrations, in addition to the three main types of atmospheric circulation, three minor types – AW, N and NE – can be determined, each with three reported cases.

2016 hourly mean CCN concentration. In this study, the CCN concentration at $SS = 0.43\%$ was averaged hourly for each reported minimum and maximum. The days with maxima are represented by a box plot in Fig. 2, noted on the legend to the right. In December, there are no extreme minimum values of CCN concentration.

During the considered period, 2016, 38 cases with extremely low concentrations of CCN and 44 cases with extremely high concentrations were reported. In general, the concentration of CCN is higher in summer than in winter. Uncertainty at extremely low values is greater than at extremely high values. At extremely low values, the lowest average hourly concentration was recorded at $9.11 - 0.76 \text{ \#}/\text{cm}^3$, and the highest at $10.09 - 420 \text{ \#}/\text{cm}^3$. At the extremely high values (Fig. 2), the lowest average hourly concentration was recorded at $12.01 - 124 \text{ \#}/\text{cm}^3$, and the highest at $12.07 - 1602 \text{ \#}/\text{cm}^3$.

Distribution of the days with extrema by using HYSPLIT. For each day with registered minimum or maximum of the concentration of CCN is drawn the backward trajectory by using HYSPLIT. The chosen time span of the backward trajectories is 72 h since this is the time scale which is typical for the description of the processes that influence the development of the atmospheric processes over the Balkan Peninsula. The backward trajectories reach Mt. Musala each day in 12 UTC. The model yields information for the location – geolocation and altitude of the air masses for each hour from the 72 back. Two types of trajectories grouping are made. On the one hand, with respect to the Earth's surface – sea (S) or continental (Cont), over which the air masses spend most of the time. For that purpose is counted for each hour over what surface the corresponding air mass is passing and three sub-groups are identified:

- a) When the air mass is more than half and less than 75% of the time, $\geq 36 < 54$ h, over the continent or over the ocean is marked with “a”;
- b) When the air mass is for 75% or more hours of the time, ≥ 54 h, over the continent or over the ocean is marked with “b”;
- c) When the air mass is all the time, 72 h, over the continent or over the ocean is marked with “c”.

With the help of this division, the most common classification of atmospheric circulation according to JCT, obtained in item 3.2, are compared. At the ex-

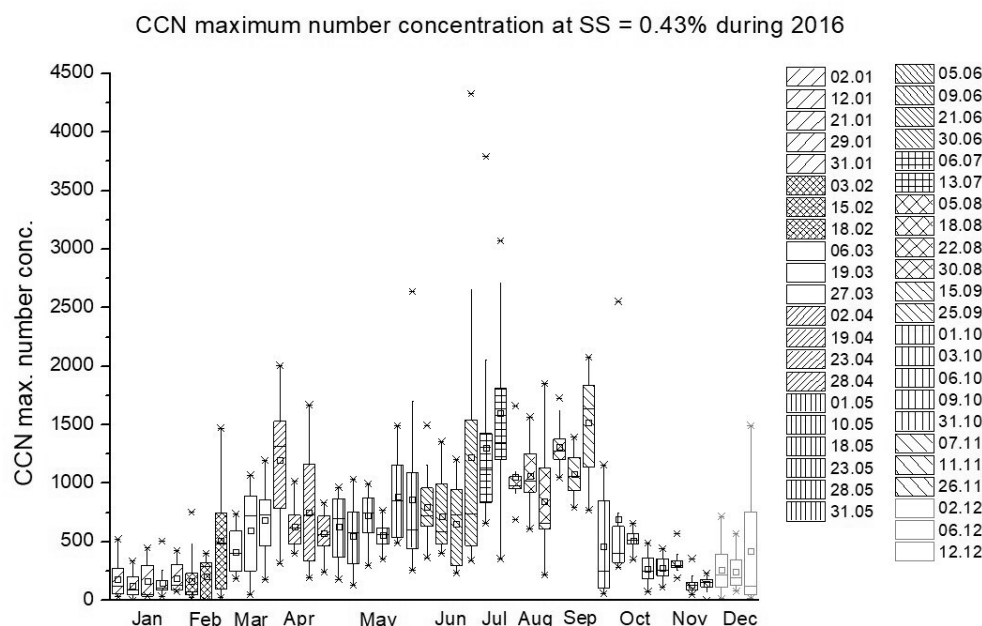


Fig. 2. CCN concentration on an hourly average scale for all days with recorded maxima. The legend on the right shows the specific day. Different patterns correspond to different months represented by x. In each box, the distribution is reported by: central line – median, square – mean, the two ends of the box plot correspond to the 25th and 75th percentiles, and the interquartile range is enclosed between them. The 5th and 95th percentile are marked with a dash, and the minimum and maximum value of the distribution with an x

tremely low values of the CCN concentration these are C, NW and W, and at the extremely high values – C, NW and W. It can be seen in Fig. 3a that at the extremely low values of CCN concentration in atmospheric circulations of type C, NW and W, a predominant type of air mass origin can be determined, respectively S_a (marine air mass that spends between 36 and 54 h over the ocean, and the rest over the continent), Cont_c (continental air mass that is all the time, 72 h, over the continent) and Cont_a (continental air mass that spends between 36 and 54 h over the continent, and the rest over the ocean). In W-type atmospheric circulation, there is no predominant origin of the air mass. The same applies to the extremely high values of the CCN concentration.

Another classification of the air masses that comes again from the backward trajectories as computed with HYSPLIT is based on the altitude. It is also compliant with basic atmospheric layers as used in the synoptic. Three groups of air masses are determined with respect to their elevation above the sea level:

- a) Low (L) – up to 3000 m (=700 hPa) – an altitude that is very close to the altitude of Mt. Musala;

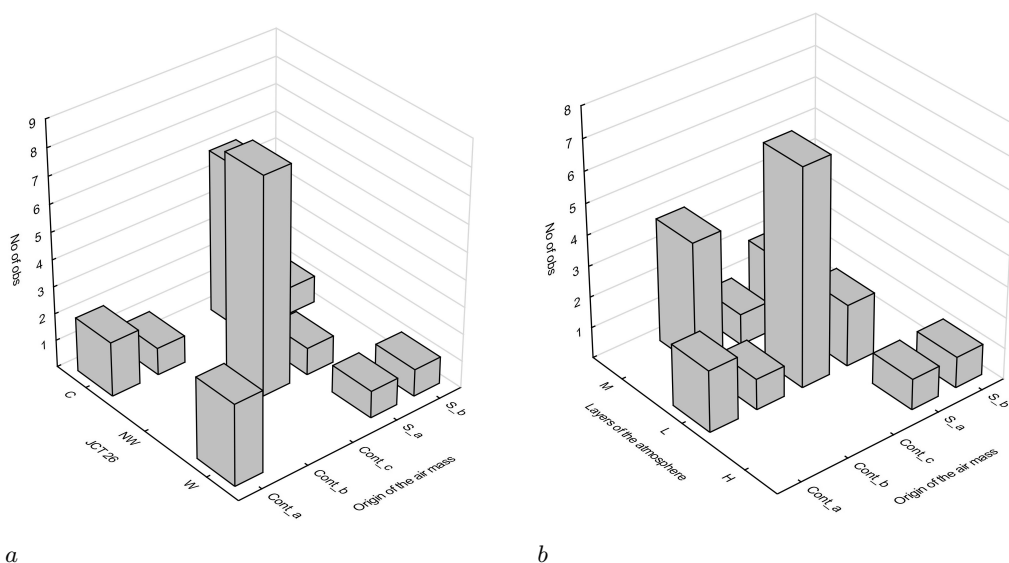


Fig. 3. Bivariate Histograms of (a) the origin of the air mass against JCT 26 on the days with minimum concentration and (b) Bivariate Histogram of Layers of the atmosphere against Origin of the air mass on days of maximum concentration

- b) Medium (M) – above 3000 up to 5500 m (=500 hPa, mid-tropospheric layer);
- c) High (H) – above 5500 m.

When comparing the altitude of the air masses with their origin as determined above it becomes clear that the extremely low concentrations of the CCN cannot be linked to a concrete dependence between their altitude and their origin. At extremely high concentrations of CCN the most frequent air masses are two types (Fig. 3b): these coming from the low layers – L, they are purely continental, and these coming from the medium layers – M, and spending more than 75% of their time over the continent (Cont_b).

Discussion and conclusion. The distribution of the CCN on top of the mountains is representative for the transfer of the air masses that influence the corresponding region. We study the distribution of the CCN during 2016 at BEO Musala. We determine the days with extrema of the CCN concentration and we use the classification at Materials and methods section [16]. In these days the prevailing atmospheric circulations according to the atmospheric circulation of JCT with 26 types at the minima of the CCN concentration are cyclonic type (C), northwest (NW) and west (W). At these types of weather the prevailing type of the origin of air masses was S_a, C_c and C_a, correspondingly. At the maxima the prevailing atmospheric circulations are southwest (SW), west (W) and northwest (NW) (with the most prevailing in both cases – the first mentioned). At the extremely high values also three secondary types occur – anticyclonic (AW),

north (N) and northeast (NE). When the extremely high concentration of CCN was observed the most frequently occurring air masses are two types: these coming from the low atmospheric layers and purely continental, and 2) these coming from the medium layers of the atmosphere (M) and spending more than 75% of the time over the continent (C_b).

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