REVIEW

of a Thesis work for obtaining the scientific degree "Doctor "
in the professional field 4.1 Physical sciences, Meteorology,
by defense procedure at the Faculty of Physics (FoP)
Sofia University "St. Kliment Ohridski ”

Reviewer: Assoc. Prof. **Dr Tatiana Stoycheva Spassova,** National Institute of Meteorology and Hydrology (NIMH), Sofia, member of the scientific jury according to Order № РД 38-188 / 25.04.2023 of the Rector of Sofia University and according to the decision of the scientific jury taken at its first meeting on 03.05.2023 (Protocol No1)

Topic of the dissertation: ”***A Complex Approach to the Research of Atmospheric Aerosols***”

Author of the dissertation: ***VIKTORIA LYUBOMIROVA KLESHTANOVA***

1. General description of the submitted materials

The doctoral candidate Viktoria Kleshtanova has provided all the necessary documents: Dissertation, Abstract (in Bulgarian and English), professional CV, as well as the mandatory tables for the Faculty of Physics, Academic Staff Development Act (ZRASRB), the Regulations Act for the Implementation of ZRASRB (PPZRASRB), the Rules of Procedure and the Regulations on the Terms and Conditions for Acquisition of Scientific Degrees and Academic Positions at Sofia University “St. Kliment Ohridski” (PURPNSZADSU). All this gives grounds for the dissertation to be accepted for review.

Electronic copies of published articles and reports, as well as one paper accepted for publication, supporting the candidate's achievements are also provided. Missing, although as a preprint version, is the publication in journal Crystals (MDPI), on which the Chapter 8 of the thesis is based.

2. Information about the Candidate

Viktoria Kleshtanova obtained her Master's degree in Meteorology at the Faculty of Physics, Sofia University. Since January 10, 2019 she was a full-time PhD student for a period of 3 years in the professional field 4.1. Physical Sciences - Meteorology, at the Department of Meteorology and Geophysics. By a decision of the Faculty Council of the FoP, her full-time PhD was transformed into a part-time one, starting from 01.04.2021, with a deadline 01.04.2022. Another extension of the deadline followed – until 01.04.2023.

Since January 2019, V. Kleshtanova is working at NIMH as a weather forecaster and since April 2022, as an assistant at the Department of Forecasting and Information Services, Meteorological Forecasting Section.

3. General characteristics of the candidate's scientific achievements

The dissertation submitted for peer-review conforms in type, volume and content to the specific requirements of the primary unit in which it was prepared. Its total volume is 111 pages, 85 of which are substantive. Bibliographical references consist of 129 titles. The abstract in Bulgarian is 57 pages long, the one in English - 53 pages and their contents correspond to that of the dissertation. The graphical material comprises 50 figures and 7 tables. Some of the figures are not of very good quality.

The dissertation is mainly devoted to the study of aerosol concentrations in the atmosphere by different methods and approaches. The topic is undoubtedly important and actual, since aerosols influence weather and climate both by directly scattering and absorbing incoming radiation from the sun and by trapping longwave radiation emitted from the Earth's surface (effective radiative forcing), and by changing the optical properties of clouds, affecting cloud formation and precipitations. The influence of aerosol particles on the above processes is not well studied yet and its incorporation into the parameterization schemes of numerical weather prediction and climate models at different spatial and temporal scales is under constant refinement.

The aim of the dissertation is, for the first time in Bulgaria, to make a comprehensive study of cloud condensation nuclei (CCN) obtained from measurements with the CCN counter (CCNC) at the Basic Environmental Observatory (BEO) on Moussala peak. The period under consideration covers the whole year 2016. The tasks related to this objective are:

1) Finding specific regularities in the distribution of CCN;

2) Determine and study of extremes (minima and maxima) of the concentration of CCN;

3) Linking the distribution of CCN with different synoptic conditions and circulation features in the country;

4) Description of heterogeneous nucleation in specific electrochemical conditions using three known crystallization models.

Publications on the subject of the dissertation are 5 in total. Four of them are in journals with ISI impact factor (one in press), and one - in a journal without IF. In all publications V. Kleshtanova is the first author. These scientific metrics exceed the recommended requirements of the Faculty of Physics for obtaining the scientific degree “Doctor”.

B. Kleshtanova is a project leader of 3 research projects funded by Bulgarian Ministry of Education and Science, under 3 calls of the National Program "Young Scientists and Postdoctoral Fellows" - in 2019, 2021 and 2022. All three projects are related to the thesis topic. She has participated in 2 research projects with Bulgarian funding and in 1, funded by international sources (COST, CA16202). Six presentations at national scientific forums on the thesis topic are mentioned.

There is no proven plagiarism in the submitted dissertation and abstract (attached are two similarity reports, prepared by her scientific supervisor Assoc. Prof. Dr V. Tonchev), with conclusions that there is no plagiarism.

4. Analysis of the scientific and scientific-applied achievements of the candidate related to this application.

The dissertation is summarized in 8 chapters, conclusions, contributions of the candidate and list of publications and presentations.

The first three chapters of the dissertation can be characterized as overview chapters, introducing basic concepts and terms and summarizing the results of research presented in about 100 publications. The first chapter argues the relevance of the presented research and formulates the main objectives of the dissertation.

Chapter 2 introduces basic concepts such as "aerosol", "cloud condensation nuclei", types of interactions. It also describes the two main methods for determining and predicting the CCN concentrations – using Köhler's theory and Twomey's empirical power law. A fairly comprehensive review of CCN studies worldwide is made, in more details for the Balkan Peninsula, especially in Greece. Concerning studies in Bulgaria on this topic, only 1 publication based on master's degree thesis of the candidate is mentioned, published in 2019 by AIP Publishing LLC. This may be explained by the fact that the CCN counter at BEO Moussala started working in late 2015 and there are few scientists with access to these specific data.

Chapter 3 is a description of the data and methodologies used in this dissertation to study CCN: 1) computing back trajectories of air masses with the HYSPLIT model to determine their origin and identify the underlying surface over which they pass; 2) description and classification by weather types based on circulation features; 3) analyses of synoptic conditions using Global Forecast System (GFS) maps and NCEP/NCAR reanalysis maps; 4) description of the Jenkinson-Collison-Types classification. The way of description of these methodologies shows the understanding and comprehension of their essence by the PhD student, which is a necessary prerequisite for their correct application in practice.

Chapter 4 is devoted to the study of the CCN concentrations under two synoptic conditions in 2016, indicative for the winter and summer half-year, respectively. The measured concentrations are analysed, extreme values are determined and episodes with concentrations above a certain value (over 400 CNNs per cubic meter for January and over 2500 for July) are investigated. The diurnal changes for one winter and one summer episodes are presented. A detailed analysis of the synoptic processes occurring during these periods (pressure field, wind, relative humidity) and the corresponding reported changes in CCN concentrations has been made. It is clearly highlighted that the CCN concentrations during the summer month are much higher compared to the winter month. The Ph.D. student shows good skills in both graphical representation and descriptive analysis of synoptic situations.

Results from this chapter have been published in the NIMH's Bulgarian Journal of Meteorology and Hydrology.

Chapter 5 is devoted to the study of backward trajectories of the air masses (AM) reaching Moussala Peak in July and December. Here, a new normalized variable n_d is introduced, which represents the percentage of the number CCNs of a given size/diameter *d* to their total number in the same volume. Figs. 5.1 and 5.2 show the dependence of this new variable from the diameter of CCNs at 6 values of air supersaturation for each day of the two months. *It is to note the similarity of the curves for the same values of supersaturation observed during the two months*. I am curious *what the explanation might be*. The variations of the average daily CCN concentrations over the two months are also presented. I do not find appropriate the idea to use one and the same scale for the Y-axis for July and December. It is mentioned in the text that some possible reasons for the higher concentrations of CCN in the summer month might be: " (i) More pollutants are reported in July, including pollen and also as a result of large fires in agricultural areas; (ii) Saharan dust transport is more intense in July".

Question: *Has any reference been made to whether there was more pollen and/or Saharan intrusion in the days with the found high CCN concentrations in July 2016? If a reference has been made and such episodes were reported, did the days with measured higher concentrations coincide with the days of episodes with more pollen and/or dust intrusions?*

Based on the results of the HYSPLIT model, a relationship between the measured CCN concentrations and the height of the layer through which the air masses (AMs) have passed in the last 72 h is made, by counting and grouping the cases with AMs from the high, middle and low atmospheric layers. Statistical evaluation is performed and results are presented in the form of box-plots for the two months. A grouping of the CCN concentrations according to the types of weather influencing the atmospheric circulation over Bulgaria has been made as well. The third type of grouping is according to the period of time from these 72 h, spent by AMs over a certain type of underlying surface and the results are presented as box-plot again.

NCEP/NCAR reanalyses for the period 1981-2010 are used as meteorological data and maps of the geopotential and the temperature anomaly at the 700 hPa level are presented for each of the two months. The days with recorded extremes in mean daily concentrations (Fig. 5.3) are superposed to the meteorological data from the reanalyses and some relationships are reported.

Results from this chapter are published in the Journal of Atmospheric and Solar-Terrestrial Physics.

Chapter 6 investigates the relationship between automatic classification of atmospheric processes and the extremes of daily averaged CCN concentrations at supersaturation S=0.43%. Days with extreme low and extreme high daily averaged concentrations have been counted for each month of 2016. It appears that the highest number of days with extreme low values are in March, May, and October and the highest number of days with extreme high values occurred in May. The days with extremely low and extremely high CCN concentrations are grouped according to the JCT atmospheric circulation types and the results are presented graphically. The conclusions from the analyses performed are that extremely low concentrations are most

frequently reported for C, NW and W, and extremely high concentrations for SW, W and NW types of JCT circulation.

The number of days with extreme low and extreme high reported concentrations of CCN by month is examined again for the hourly averaged concentrations at S=0.43%. It is concluded that, in general, the concentration of CCN is higher in summer than in winter. The uncertainty in the extreme low values is greater than in the extreme high values.

Here again, as in Chapter 5, the HYSPLIT model is applied for each day with reported minimum or maximum of the CCN concentrations. The selected trajectory duration is 72 hours backward, and the trajectories reach Moussala Peak at 12 UTC each day. The model provides information on the location, geographic coordinates and altitude, of the air mass at each hour. Two types of grouping are used: 1) according to the surface the air masses pass over - sea (S) or continental (C); and 2) according to the altitude the air masses pass trough – low, medium and high. Comparing the altitude of air masses with their origin, it is concluded that for the extremely low concentrations of CCNs no specific dependence could be found, while for the extremely high concentrations, two most common dependencies are detected: (1) air masses coming from the low layers and purely continental (Cc) and (2) AM coming from the middle layers of the atmosphere that have spent more than 75% of the time over the continent (Cb). Both results are presented as two dimensional (bivariate) histograms.

Results from this chapter have been published in the journal Comptes rendus de l'Académie bulgare des Sciences.

Chapter 7 discusses results from the application of Twomey's law. The CCN distribution data are approximated by Twomey's law for all 6 supersaturations used by CCN counter. Four months of 2016, representative for the four seasons, are selected. The data are presented graphically, the values of the parameters C and k from Twomey's law are obtained, the standard error R^2 as well as other statistical characteristics are calculated. A large part of the data described by Twomey's law has R^2 above 0.95% throughout the period considered, and its lowest obtained value is 0.81%.

Relationships between the values of the parameters C, k and basic meteorological characteristics such as temperature, wind direction and speed, relative humidity, atmospheric pressure have been found and studied. A comparison of the relationships between C, k and the temperature at the synoptic station Moussala and at the climatic station Borovets is made.

Similar trend in the behaviour of the parameter C and the temperature is found, results obtained for different time periods for the four months considered at both stations. Finding a physical explanation of the behaviour of these parameters and subsequent separation into characteristic time periods, rises new research questions, subject to future research, according to the plans of the PhD student.

The Pearson correlation coefficients between the parameters of the Twomey's equation and the corresponding minimum, maximum, and daily mean C, k, and temperatures, as well as the JCT atmospheric circulation types, are tabulated. Relationships between the parameter C and the temperature at the highest correlation coefficients are determined.

Chapter 8 of the thesis is devoted to a reanalysis of data from experiments on heterogeneous nucleation under electrochemical conditions conducted by other authors. An analogy is made between the studies on CCN and vapor deposition on solid surfaces, once conceptually and once, in defining supersaturation in the experimental system and that of supersaturated vapor in the atmosphere. The data are approximated by three models: a two-, three- and four-parameters model, respectively.

The studies in this chapter are presented in a publication in the MDPI journal Crystals.

The conclusion summarizes the research done in the previous chapters and the results obtained.

The formulated by the PhD student contributions correspond to the objectives of the dissertation set at the beginning, namely:

- A correlation is established between the maxima in the CCNs concentration, the backward trajectories of the air masses and the synoptic conditions in 2016;

- Relationships between the extremes of the CCN concentration and the Jenkinson-Collison-Types circulation types are found;

- High correlation coefficients between a parameter of the Twomey equation and temperatures at Moussala and Borovets are found;

- New analyses of previously published data on heterogeneous nucleation in electrochemical conditions by three models are made.

The contributions could be identified as building on existing knowledge and obtaining new one, a basis for future research in applied science. The relationships between cloud condensation nuclei concentrations and atmospheric circulation types obtained in the dissertation, can be applied in operational weather forecasting, precipitation forecasting and especially hazardous phenomena such as hailstorms.

5. Critical remarks and recommendations

The work for data collection and processing and the results obtained is undoubtedly time-consuming and requires versatile proficiencies and skills that the PhD student possesses. The data has been studied from different aspects, with different techniques and approaches, which really justifies the title of the thesis "A Complex Approach to the Study of Atmospheric Aerosols". However, in many places the results are just shown and described verbally, but not analysed. I assume that with the experience and skills accumulated so far, V. Kleschanova will continue the research on the subject by paying more attention to the analyzes of the reasons for the changes in the CCN concentrations under different synoptic conditions, different types of underlying surfaces over which the AM has passed, the height of the stream, etc.

A more precise formulation of the Bulgarian text and some additional clarifications are needed.

The above-mentioned remarks in no way detract the work done by V. Kleshtanova and the significance of the obtained results.

6. Personal impressions of the candidate

I have almost no personal impressions of V. Kleshchanova. I listened her presentation at a COST CA16202 inDust meeting where she introduced to the audience cases with Saharan dust intrusions over Bulgaria. I was very impressed with the content, the way of presenting the topic and the confidence with which it was delivered. These are valuable skills.

7. Conclusion

Having read the submitted dissertation, Abstract and other materials, and on the basis of the analyses of their significance and scientific and applied contributions contained therein, I confirm that the scientific achievements of the candidate fully meet the requirements of the ZRASRB and Regulations for its application and the relevant Regulations of the Sofia University "St. Kliment Ohridski" for acquiring the educational and scientific degree "Doctor". In particular, the candidate satisfies the minimum national requirements in the professional field and no plagiarism has been found in the dissertation, abstract and scientific papers submitted for the competition.

I give my positive assessment of the dissertation work.

OVERALL CONCLUSION

Based on the above, **I recommend** the scientific jury to award **the scientific degree "Doctor"** in the professional field 4.1 "Physical Sciences" - Meteorology to **Viktoria Kleshtanova**.

25.06.2023 Reviewer: .....

(Assoc. Prof. Dr Tatiana Spassova)