

# **HABILITATION THESIS**

## **Summary**

### **Contributions to Advanced Characterization of Atmospheric Aerosol Optical and Mass Properties through the Synergy of Remote Sensing Techniques and Atmospheric Models**

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## Summary

Atmospheric aerosols remain one of the greatest sources of uncertainty in the assessment of radiative forcing and in climate projections. Variations in net radiative flux, in this context, are produced by changes in external factors of the climate system such as: aerosols, greenhouse gases, surface albedo, etc. A positive variation indicates a surplus of energy flux absorbed by the climate system (warming effect), while a negative variation indicates an energy flux deficit (cooling effect).

According to the IPCC<sup>1</sup> assessment (Chapter 7.3.3), total anthropogenic radiative forcing for the period 1750–2019 is positive, dominated by increasing concentrations of greenhouse gases, indicating a net warming trend in the climate system. The primary contribution comes from carbon dioxide (CO<sub>2</sub>), followed by the remaining well-mixed greenhouse gases (methane: CH<sub>4</sub>, nitrous oxide: N<sub>2</sub>O, and halogenated compounds), while stratospheric ozone and water vapour add additional positive contributions, albeit with minor influence. Changes in surface albedo are attributed to land-use modifications (deforestation, expansion of agricultural areas) and to the deposition of absorbing particles on snow and ice, and likewise represent a modest positive radiative forcing that amplifies warming. Anthropogenic aerosols, by contrast, exert an overall negative radiative forcing (cooling effect), through interactions both with radiation and with clouds, partially offsetting the positive radiative forcing of greenhouse gases. Systematic monitoring through long-term time series measurements and determination of aerosol optical properties (AOD<sup>2</sup>, SSA<sup>3</sup>, lidar ratio, depolarization, etc.) makes significant contributions to reducing uncertainties in the input parameters used in radiative transfer models, thereby improving the calculation of radiative forcing through the direct effect. Reference models recognised in the scientific literature include OPAC<sup>4</sup>, POLIPHON<sup>5</sup>, and AERONET<sup>6</sup>.

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<sup>1</sup> Intergovernmental Panel on Climate Change: <https://www.ipcc.ch/>

<sup>2</sup> AOD - Aerosol Optical Depth

<sup>3</sup> SSA - Single Scattering Albedo

<sup>4</sup> Optical Properties of Aerosols and Clouds

<sup>5</sup> POLarization LIdar PHOtometer Networking

<sup>6</sup> Aerosol Robotic Network, <https://aeronet.gsfc.nasa.gov/>

European research infrastructures such as ACTRIS<sup>7</sup> have the explicit objective of providing data based on long-term standardised time series of observations of aerosol optical properties (from the total atmospheric column and from vertical profiles), for documenting the effects of aerosols, clouds, and short-lived reactive gases (hours, days) on climate and air quality.

In Romania, the context for the proposal of a national lidar network is represented by EARLINET<sup>8</sup>, founded in 2000 as a European Commission research project, with the aim of creating a statistical database of the vertical distribution of aerosols at continental scale. The lidar station (RALI<sup>9</sup>) at Bucharest-Măgurele (INOE<sup>10</sup>) joined EARLINET in November 2005, under the coordination of Dr. Doina Nicolae. ROLINET<sup>11</sup> was implemented between 2008 and 2011 as a national initiative to consolidate Romania's capabilities in atmospheric remote sensing through lidar systems. The project was nationally funded (PNCDI-II) and brought together a consortium of research centres and universities including: INOE, ANM<sup>12</sup>, UPT<sup>13</sup>, UAIC<sup>14</sup>, UBB<sup>15</sup>. The main objective was the creation of the first network of lidar systems in Romania, optimised for continuous monitoring of the vertical distribution of aerosols, with integration into EARLINET, GAW<sup>16</sup>, and GEOSS<sup>17</sup>. Simultaneously, under INOE coordination, the RADO<sup>18</sup> project was initiated, funded through the Norwegian Cooperation Programme with Romania, with an implementation period of 2009–2011.

RADO aimed to create the first 3D atmospheric observatory in Romania (and the only one in South-Eastern Europe at the time), dedicated to the research and monitoring of atmospheric processes in the planetary boundary layer and the free troposphere. AERONET in Romania was implemented through the infrastructure and techniques developed within the ROLINET and RADO projects.

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<sup>7</sup> Aerosol, Clouds and Trace Gases Research Infrastructure: <https://www.actris.eu/>. ACTRIS coordinates remote sensing instruments (lidar, photometers, cloud radars) and in situ measurements, with calibration and quality control procedures recognised in the scientific literature. ACTRIS enforces common operating protocols so that data are directly comparable with those from the rest of Europe.

<sup>8</sup> European Aerosol Research Lidar Network, <https://www.earlinet.org>

<sup>9</sup> Multi-wavelength Raman lidar system with depolarization

<sup>10</sup> National Institute for Research and Development in Optoelectronics – INOE 2000

<sup>11</sup> Romanian Lidar NETWORK

<sup>12</sup> National Meteorological Administration

<sup>13</sup> Politehnica University of Timișoara

<sup>14</sup> Alexandru Ioan Cuza University of Iași

<sup>15</sup> Babeș-Bolyai University

<sup>16</sup> Global Atmosphere Watch

<sup>17</sup> Global Earth Observation System of Systems

<sup>18</sup> Romanian Atmospheric research 3D Observatory

In the present habilitation thesis, the studies carried out after obtaining the doctoral degree are described. These studies focused on identifying the physical phenomena that control how much the solar irradiance is absorbed or scattered by atmospheric aerosols, on the characterization of the optical and microphysical parameters of aerosols at the eastern border of ACTRIS (with direct application in the evaluation of regional climate and air pollution models). Within the framework of the European ACTRIS infrastructure, the central component of the present habilitation thesis is represented by the physics of the interaction of electromagnetic radiation with matter (aerosols, clouds), quantified through measurable optical quantities (e.g.: AOD, AE<sup>19</sup>, lidar ratio, depolarization, single scattering albedo, etc.) using modern remote sensing techniques (lidar, solar photometry), employed to characterize aerosol types (from various sources: Saharan dust, biomass burning, volcanic ash, etc.), to characterize seasonal variability, to determine the role of the PBL structure in aerosol dispersion, and to assess air quality risk (as an operational application of aerosol optical property identification). Furthermore, through the presentation of case studies, new methods are proposed to address the challenge of discerning relatively small variations in column-integrated aerosol optical parameters.

The majority of experiments and measurements (monitoring activities) were carried out in the Laboratory of Atmospheric Optics, Spectroscopy and Lasers (RADO-Iași monitoring station within ACTRIS-RO), Faculty of Physics, “Alexandru Ioan Cuza” University of Iași, and in close collaboration with researchers and academic staff from the ACTRIS and ACTRIS-RO network: the Remote Sensing Department and MARS<sup>20</sup> research centre (RADO-Bucharest monitoring station within ACTRIS-RO) at INOE; ACTRIS-UBB (RADO-Cluj monitoring station), Faculty of Environmental Science and Engineering, Babeş-Bolyai University, Cluj-Napoca; the Climate Change Observation Platform (POSC Fix) at the REXDAN<sup>21</sup> research centre (RADO-Galați monitoring station within ACTRIS-RO), “Dunărea de Jos” University of Galați; the Department of Environmental Engineering and Management, Faculty of Chemical Engineering and Environmental Protection “Cristofor Simionescu”, “Gheorghe Asachi” Technical University of Iași; the Laboratory of Electron Microscopy and Chemical Analysis, Faculty of Materials Science and Engineering, “Gheorghe Asachi” Technical University of Iași; Faculty of Geography and Geology, “Alexandru Ioan Cuza” University of Iași; National

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<sup>19</sup> Ångström exponent

<sup>20</sup> Magurele Atmosphere and Radiation Site

<sup>21</sup> Integrated system for comprehensive research and monitoring of the environment in the Danube river area

Meteorological Administration, Regional Forecasting Centre Bacău; Politehnica University of Timișoara; Laser Remote Sensing Unit, Physics Department, National Technical University of Athens, Greece; Université des Sciences et Techniques de Lille, Laboratoire d'Optique Atmosphérique, Villeneuve d'Ascq, France. Access to certain infrastructures was also made possible through collaboration with: SC INOESY SRL, Iași, Romania; SC EnviroScopY SRL, Iași, Romania; Raymetrics SA, Athens, Greece.

The thematic architecture of the present habilitation thesis is illustrated schematically through the conceptual diagram presented below in Figure 1. The central element of this diagram synthesizes the theoretical and experimental methodological framework described in Chapters 1 and 2, which constitutes the instrumental and procedural foundation of the research carried out. It is important to emphasize that some of the methods and techniques presented in these chapters do not represent exclusively tools adopted from specialized literature, but were developed, adapted, and validated within the studies comprising the present habilitation thesis, thus constituting original contributions reported after obtaining the doctoral degree.

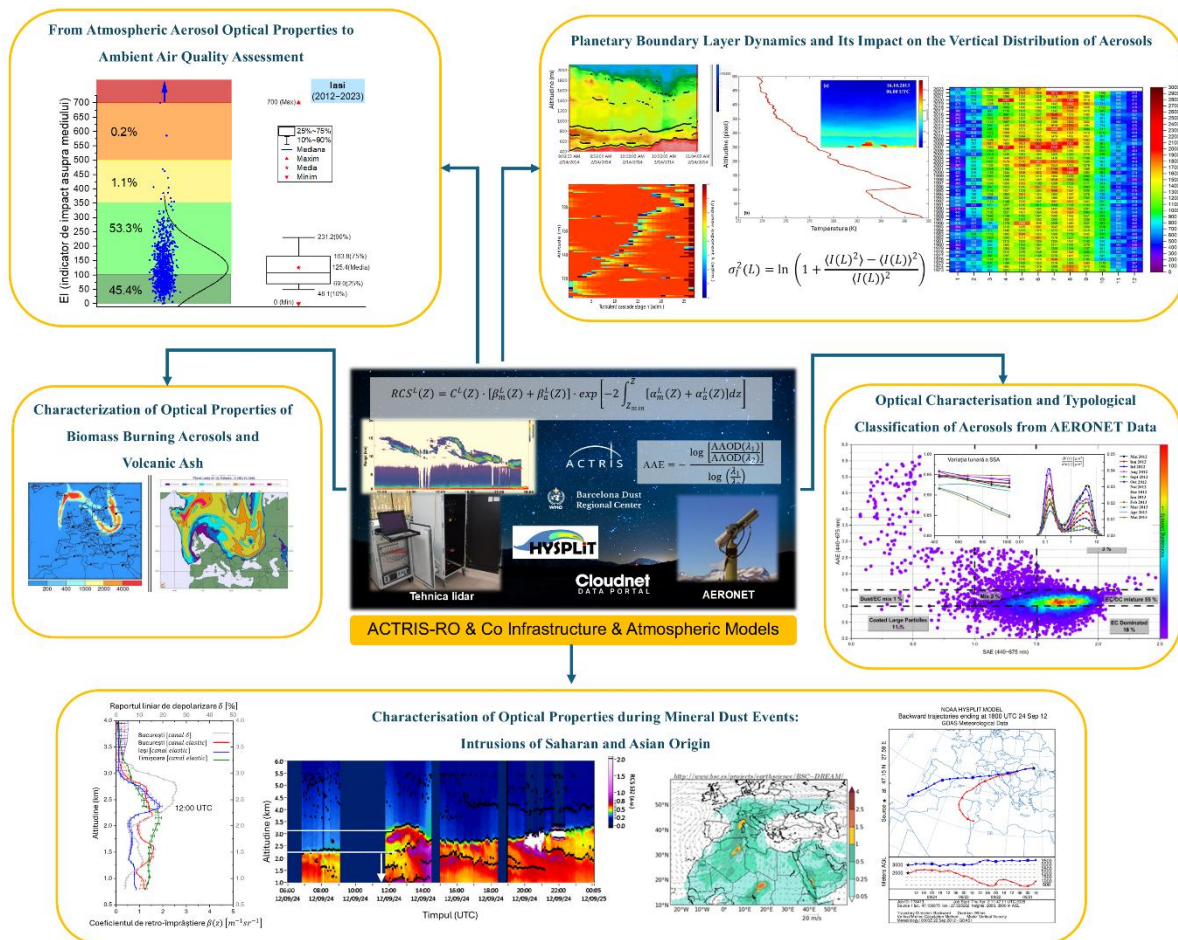


Figure 1: Schematic representation of the thematic architecture of the habilitation thesis

Starting from this methodological core (the remote sensing infrastructure of both ACTRIS and certain institutional partners, together with the atmospheric models employed), the diagram highlights the main thematic directions addressed in Chapter 3, dedicated to the presentation and discussion of original results, with each subchapter corresponding to a specific node in the proposed scheme. This graphical representation is selective with respect to the full content of the thesis. Chapter 4, dedicated to the presentation of future research directions and to demonstrating the individual capacity to coordinate research teams, goes beyond the thematic scope of this diagram and is treated independently; the purpose of this conceptual diagram is to offer an overview for understanding the correlations between the methods used and the results obtained.

Within the present habilitation thesis, scientific results were presented regarding aerosol mixtures, the predominant aerosol type during pollution events, and the role of the PBL structure in aerosol dispersion, using multi-instrumental data and numerical models. For instance, it was observed that in the urban environment the background aerosol loading is dominated by fine continental aerosols of urban-industrial and traffic origin, characterized by high Ångström exponent values and a high fine-mode fraction, particularly in winter, indicating the predominance of local sources. Within the studies conducted in the urban area of Iași, it was shown that during the summer of 2012 the dominant aerosol type was urban/industrial, while the influence of mineral dust and biomass burning manifested in pollution episodes superimposed on this background. In Bucharest, the analysis for the period 2015–2024 shows low mean AOD values and high AE values, with dominant aerosol types of continental and urban-industrial origin, while smoke and mineral dust appear during pollution episodes. In Greece (Athens), local urban sources produce a lower layer (below 1.5 km) with high backscattering coefficient and low depolarization, associated with traffic and industrial pollution, clearly separated from the dust layers in the free troposphere.

Regarding long-range aerosol transport, results were presented from various measurement campaigns. For example, during the AGRO measurement campaign (September 2012), a Saharan dust episode was captured as it successively crossed Greece and Romania, with well-defined dust layers between 2 and 4 km above Athens, Oxyliothos, Timișoara, Bucharest, and Iași, with HYSPLIT confirming the origin in the northern Sahara and trajectories over the Mediterranean and Italy. In Bucharest, a 10-year analysis confirms that Saharan and Middle Eastern dust is detected mainly in spring and summer at altitudes between 2 and 8 km, while smoke layers from North Africa, North America, and Europe appear

predominantly in summer and in the lower troposphere. The Sahara remains the dominant source of mineral dust transported towards Europe and Romania, with transport facilitated by Mediterranean cyclones centered over southern Italy and by South-West to North-East circulation. Nevertheless, significant secondary sources exist: the Middle East (spring–summer), the Kalmyk steppe, and the Turan Plain of Central Asia, identified during the August 2022 event in eastern Romania. Results were also presented regarding planetary boundary layer dynamics (inversions, local atmospheric circulation) that influence the retention of local pollutants (PM, gases) and the manner in which these can form mixtures depending on the predominant aerosol type in the free troposphere.

Based on turbulence theory, in recent studies, laminar channels were interpreted as preferential pathways for vertical transport (of aerosols, water/ice) in the atmospheric column, identified from lidar (ceilometer) measurements at ACTRIS stations: INOE-Bucharest and UGAL-Galați.

Towards the end of Chapter 3, through the use of AOD values (AERONET) and integrated risk, a differentiated rescaling procedure was presented for a risk classification directly applicable to air quality policies and standards. Through the analysis of the PM<sub>2.5</sub>/PM<sub>10</sub> ratio in urban areas of Suceava, Iași, and Botoșani over periods exceeding five years, it was shown that, although annual PM averages comply with the limit values, the PM<sub>2.5</sub>/PM<sub>10</sub> ratio is elevated in winter and decreases in summer, indicating the predominance of anthropogenic combustion sources (residential heating, traffic) in the cold season and coarse/natural sources in the warm season. Thus, through statistical analysis of PM<sub>2.5</sub> and PM<sub>10</sub> concentrations, new indicators were proposed that can be used in compliance scenarios, air quality plans, and the prioritization of measures.

In Chapter 4, new research topics were proposed, aiming at the continuation and deepening of investigations regarding the planetary boundary layer, building upon the infrastructure and expertise accumulated through participation in the ACTRIS-RO network, as reflected in the majority of studies presented in this thesis (as principal author and co-author).

These research directions benefit from a consolidated instrumental framework, including the Raman lidar systems and ceilometers operational at ACTRIS-RO stations, as well as extensive personal experience in the processing and interpretation of both passive and active remote sensing data. The continuous monitoring of PBL height through these systems, with a focus on the automatic identification of nocturnal thermal inversions and their correlation with

episodes of dense fog and PM<sub>2.5</sub>/PM<sub>10</sub> pollution, represents a current scientific challenge with direct implications for air quality and meteorological forecasting. One of the research directions is in continuity with already initiated studies, among which the first PBL climatology for southern Romania stands out, carried out over a 50-year period (1973–2023), through which a significant increasing trend in mixed-layer height after the year 2000 was identified, correlated with the intensification of thermal convection in the context of regional climate change. In the same context, another proposed research direction targets the integration of low-cost sensors (LCS) into a hybrid short-term air quality forecasting system (1–2 hours), through the fusion of LCS data with data from the reference stations of the National Air Quality Monitoring Network (RNMCA) and with PBL physical parameters derived from active remote sensing (lidar, ceilometer). In the case of aerosol mixtures, optical properties differ significantly from those of the "pure" components, both spectrally and in terms of scattering and absorption efficiencies, which complicates their parametrization in models and the direct interpretation of standard measurements. This complexity justifies the need for studies focused on the local "fingerprint" of aerosol mixtures, combining long time series of optical and microphysical observations with information on sources and transport conditions, not only to describe regional particularities but also to more clearly delineate the limits of applicability of classical optical parameters and of the mixing schemes currently in use. Accordingly, a new research direction was proposed in the study of complex urban aerosols and advanced optical parameters. The link between optical properties (AOD, SSA, depolarization) and chemical composition in urban and suburban environments will yield "optical signatures" of different sources such as traffic, residential heating, and mixed episodes with Saharan dust. Furthermore, through the characterizations of hygroscopicity and the increase of optical depth with relative humidity from Raman lidar data, particle physics (i.e., the size-composition-water relationship) is directly linked to smog episodes and to radiative effects in urban areas. Within this direction, new inversion and classification methods will be tested, combining lidar, ceilometer, photometer, and, where available, in situ data, to characterize the optical and microphysical properties of urban and peri-urban aerosols (including BC, smoke, dust, and mixtures).

The author's capacity to coordinate research teams has been demonstrated (principal investigator of two research grants, member of doctoral supervision committees, member of doctoral thesis examination committees) as well as the capacity to manage teaching activities (new laboratory sessions and seminars, contributions to the development of a laboratory manual, teaching mobility activities).