GUIDANCE DOCUMENTS ON MEASUREMENTS & MODELLING OF NOVEL AIR QUALITY POLLUTANTS:

VERTICAL PROFILES OF AEROSOLS

Research Infrastructures Services Reinforcing Air Quality Monitoring Capacities in European Urban & Industrial AreaS (GA n. 101036245)

By

Italian National Research Council

29/11/2024

Authors: Lucia Mona (CNR), Doina Nicolae (INOE), Francesca Barnaba (CNR), Annachiara Bellini (CNR, now ARPA Val d'Aosta) Simone Kotthaus (CNRS), Martial Haeffelin (CNRS)

Reviewers: Xavier Querol (CSIC), Ewan O'Connor (FMI), Adolfo Comerón (UPC), Iwona Stachlewska (UW), Arnoud Apituley (KNMI), Andreas Petzold (Jülich)

Table of Contents

ABBREVIATIONS I

1. ABOUT THIS DOCUMENT 1

2. DEFINITION OF AEROSOL PROFILING QUANTITIES 1

3. MEASUREMENT METHODS AND QUALITY CONTROL OF AEROSOL PROFILING VARIABLES 2

3.1 State of harmonisation 2
3.2 High power lidars requirements and SOPs 4
3.2.1 Quality assurance, quality control and measurement uncertainty 5
3.3 Data management summary 7

4. PAN-EUROPEAN OVERVIEW OF AEROSOL PROFILES ACROSS URBAN ENVIRONMENTS 8

4.1 Introduction 8
4.2 Aerosol optical properties climatology over Europe 2000-2019 8
4.2.1 Methodology 8
4.2.2 Highlights 9
4.3 Aerosol Typing over Europe: long term analysis 12
4.3.1 Methodology 12
4.3.2 Highlights 12

5. RECOMMENDATIONS 15

5.1 Maintenance and quality control 15
5.2 Data management summary 16

6. REFERENCES 17

ABBREVIATIONS

ACTRIS	Aerosols, Clouds and Trace gases Research InfraStructure
ACTRIS ARES	ACTRIS Center for Aerosol remote sensing profiling
ACTRIS CARS	ACTRIS Center for Aerosol Remote Sensing
AERONET	AErosol RObotic NETwork
AHL	Aerosol high power lidars
ALC	Automatic lidar-ceilometers
BC	Black carbon
CALIPSO	Cloud-aerosol lidar and infrared pathfinder satellite observations
CEN	European Committee for Standardization
EARLINET	The European Aerosol Research Lidar Network
EC	Elemental carbon
GAW	Global Atmospheric Watch programme by WMO
MPQC	Multi product quality control
NATALI	Neural network aerosol typing algorithm based on lidar data
PM	Particulate matter
QA/QC	Quality assurance and quality control
RI-URBANS	Research Infrastructures Services Reinforcing Air Quality Monitoring Capacities in European Urban & Industrial AreaS EU-project
SCC	Single calculus chain
SOP	Standard operation procedure
WMO	World Meteorological Organization

1. ABOUT THIS DOCUMENT

This document was prepared as part of the "Research Infrastructures Services Reinforcing Air Quality Monitoring Capacities in European Urban & Industrial AreaS" (RI-URBANS) EU-project that connects the atmospheric observation expertise from the Aerosols, Clouds and Trace gases Research InfraStructure (ACTRIS), as well as the urban air quality observation capacities of the regulatory air quality monitoring networks.

The directive underlines the importance of emerging pollutants to air quality and the well-being of citizens. Particulate matter (PM), and in particular some of its components such as ultrafine particles (UFPs), black carbon (BC) and elemental carbon (EC), need to be monitored in both rural and urban supersites in order to support the scientific understanding of their effects on health and the environment, as recommended by WHO.

Currently, most Air Quality Monitoring Networks (AQMNs) miss information about important processes and quantities in the vertical dimension that are necessary to better understand surface-level pollution data. The vertical dimension is especially relevant when considering potential non-local sources of aerosols (e.g. those arriving via medium-to-long-range transport) and for evaluating vertical dilution of locally emitted pollutants and, in specific conditions, episodes associated with new particle formation and related particle growth processes.

Several techniques are currently available for aerosol profiling, with different levels of product quality and uncertainty and maturity in terms of standardization. Standardized measurement techniques and methods across distributed sites are essential for guaranteeing sufficiently comparable air quality investigation over Europe. The aim of this document is to facilitate the consideration of aerosol profiling within air quality networks. We provide a concise summary of the currently available methodologies, focusing on very precise methods, a synthesis of Pan-European observations, and finally concise recommendations on the profiling measurements in urban environments.

This is a RI-URBANS/ACTRIS guidance for this specific service tool that is part of the RI-URBANS deliverable D46 (D6.1, containing guidance for all service tools provided in the project) with the support for publication from AXA Research Fund to build up the final dissemination D55 (D7.6). Any dissemination of results must indicate that it reflects only the author's view and that the European Commission is not responsible for any use that may be made of the information it contains.

2. DEFINITION OF AEROSOL PROFILING QUANTITIES

Aerosol profiling quantities are typically measured through range-resolved optical remote sensing, so that optical properties are the quantities measured and are typically dependent on the chemical composition and shape of the particles and on their concentration (e.g. aerosol extinction coefficient profiles). Some intensive parameters can then be directly obtained (typically through calibrated ratio of optical properties, e.g. depolarization ratio). Finally, some quantities can be retrieved from the previous quantities, such as the mass concentration.

Following Climate Forecast nomenclature (https://cfconventions.org/Data/cf-standard-names/current/build/cf-standard-name-table.html), the **aerosol particle backscatter coefficient** (shortly known as **aerosol backscatter**) is the fraction of radiative flux, per unit path length and per unit solid angle, scattered by aerosol particles at 180 degrees angle respect to the incident radiation and obtained through ranging techniques like lidar and radar. The backscattering coefficient is related to the same wavelength of the incident radiation, unless otherwise specified.

The **volume aerosol particle extinction coefficient** (shortly known as **aerosol extinction**) is the fractional change of radiative flux per unit path length. Extinction is the sum of absorption and scattering effects, sometimes called "attenuation".

The **atmosphere backscatter linear depolarization ratio** (often called **volume depolarization ratio**) ratio is defined as the ratio of the cross-polarized lidar return signal to the parallel-polarized backscatter signal, due to both particle and molecular parts present in the atmospheric investigated volume. It is a calibrated measurement.

From these quantities, the following intensive properties are obtained:

The **aerosol particle extinction to backscatter ratio** (known as **lidar ratio**) is the ratio of the aerosol particle extinction coefficient to the aerosol particle backscatter coefficient of radiative flux by ranging instrument in air due to ambient aerosol particles. The ratio is related to the same wavelength of incident radiation.

The **aerosol particle extinction Ångström exponent** (often called **Angstrom exponent**) is the Angstrom exponent obtained for the aerosol extinction instead of that for the aerosol optical thickness and is *alpha* in the relating aerosol extinction at the wavelength *lambda* as a function of aerosol extinction at a different wavelength *lambda0*: ext(lambda) = ext(lambda0) * [lambda/lambda0] ** (-1 * alpha).

The **aerosol particle backscatter Ångström exponent** (often called often called **backscatter Angstrom exponent**) is the Angstrom exponent obtained for the aerosol backscattering instead of aerosol optical thickness and is *alpha* in the relating aerosol backscatter at the wavelength *lambda* as a function of aerosol backscattering at a different wavelength *lambda0*: back(lambda) = back(lambda0) * [lambda/lambda0] ** (-1 * alpha).

The **aerosol particle backscatter linear depolarization ratio** (often called **particle linear depolarization ratio**) is defined as the ratio of the perpendicular polarization component to the parallel component of aerosol scattering only (without the molecular contribution). It is obtained combining the volume linear depolarization ratio with the molecular and aerosol backscatter referred to the same investigated atmospheric volume.

The **aerosol type** is a classification of the aerosol made, in the case of lidar measurements, on the basis of the measured intensive properties. The classification can be more or less detailed depending on the measurement capabilities.

3. MEASUREMENT METHODS AND QUALITY CONTROL OF AEROSOL PROFILING VARIABLES

3.1 State of harmonisation

At the present there are no CEN standards for aerosol profiling measurements and ISO standard definition for backscatter lidar is in progress: Anyhow, these quantities are standardized at European level within ACTRIS (Aerosol Clouds and Trace Gases Research Infrastructure). Aerosol profiling variables are part of the ACTRIS aerosol remote sensing observations, made through high power lidars coupled for advanced products with collocated photometers, standard operational procedures (SOPs) (https://www.actris.eu/sites/default/files/inline-files/SOPs-CARS-Nov2023-v01-rev08.pdf) and quality assurance/quality control procedures (QA/QC) that are established within ACTRIS for aerosol profiling (https://www.actris.eu/sites/default/files/inline-files/QAPs-CARS-Jan2024-v01-rev12.pdf and https://www.earlinet.org/index.php?id=293).

In addition, automatic lidar-ceilometers (ALC; also referred to as low power lidars) can be useful for monitoring the presence and type of aerosol particles. In particular, ALC equipped with a polarization sensitive channel (polarization-sensitive ALC, or PLCs) are increasingly being operated in European networks (e.g., E-PROFILE). Such systems are able to provide insights about aerosol type for example detecting the presence of desert dust intrusions. This is routinely done in Italy within the operational automated lidar-ceilometer network ALICEnet (www.alice-net.eu) which provides real-time aerosol observations at several sites across the country. Within RI-URBANS the ALICEnet software (Bellini et al., 2024) have been also tested over the RI-URBANS Paris Pilot site for three specific desert dust events that occurred in 2022-2023 (Bellini et al., 2025). Figure 1 show an example of the results of this 'tool upscaling' in Paris, demonstrating the additional insights gained from using remote sensing to support AQ monitoring.

In particular, Figure 1a shows the temporal (11-18 June 2022) and vertical (0 – 7 km) evolution of the aerosol mass profiles (a) as estimated by ALC measurements (HydroMett CHM15K instrument) performed over Paris (Qualair-SU site). Figure 1b shows the same scene in terms of particle shape (from spherical, blue, to highly irregular, red), demonstrating the ability of a PLC (Vaisala CL61 instrument operated in the Paris-Hotel de Ville site) to identify the presence of irregular particles as those typical of desert dust layers. In Figure 1c we show the temporal evolution of particle matter data at ground level, and namely, the particle mass estimate derived by ALC in the vertical range closest to the surface (light blue dots) and the corresponding in situ PM10 measurements (grey dots) performed in the AQMN site of Paris-Vitry. Overall, Figure 1 shows a typical example of desert dust transport and related impact on AQ. In fact, a marked increase in particle mass can be observed in the upper atmospheric levels (3-5 km) on 16 June (yellow colour in Figure 1a). This aerosol layer is clearly loaded with irregular shaped particles (the red colour in Figure 1b) and progressively intensifies and descends towards the lowermost atmospheric levels on June 17 and 18 (black arrow), when it finally mixes within the planetary boundary layer (Figure 1a, b) (see RI-URBANS ST7 Measurements of boundary level height documentation for more details), leading to increased PM10 values down to the ground (Figure 1c).

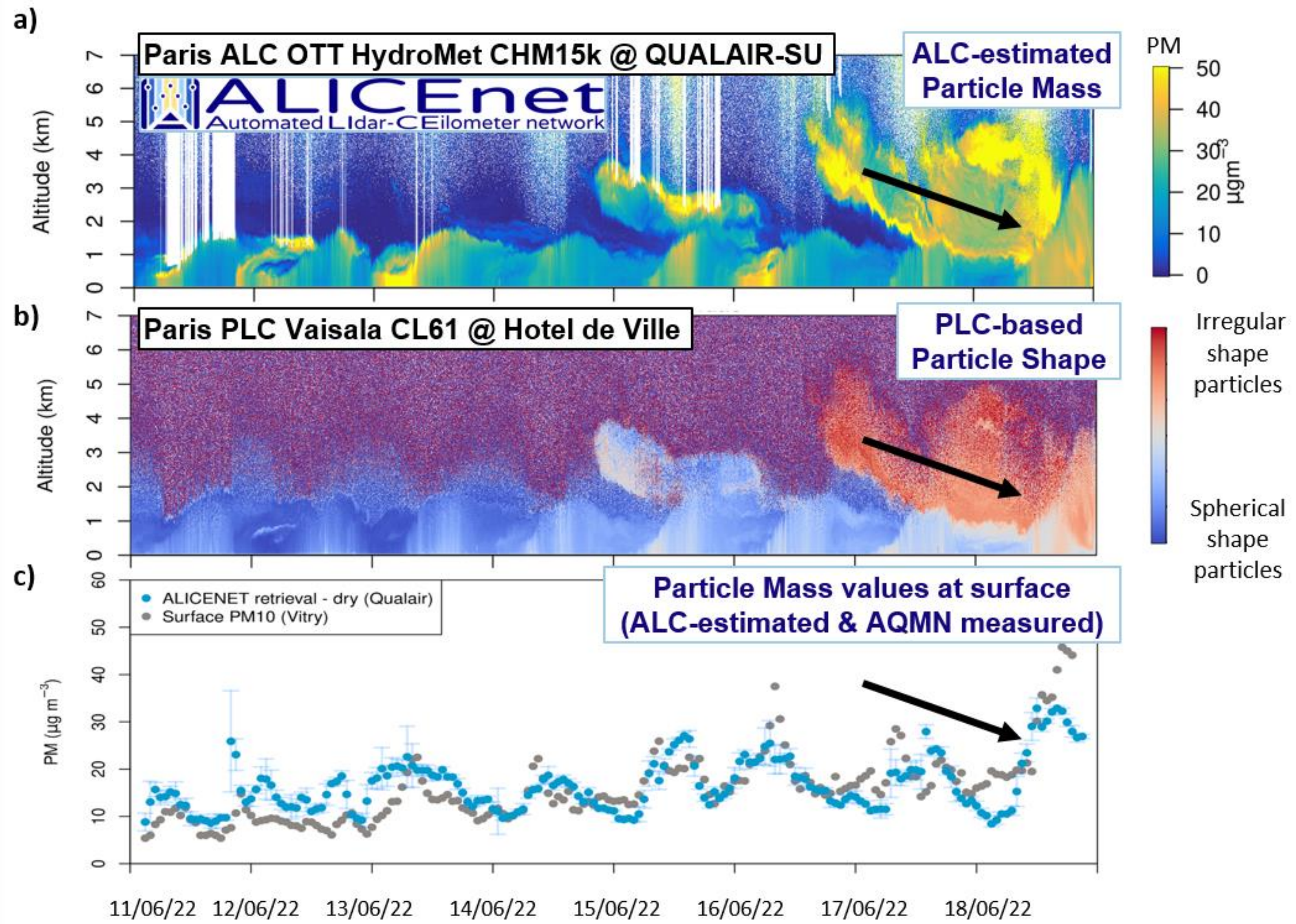

Figure 1. *Example of application of ALICEnet methods upscaling over the RI-URBANS Paris Pilot site during a desert dust event in June 2022. Vertical (0 -7 km, y axis) and temporal (June 11-18, x axis) evolution of (a) particle mass vertical profiles, and (b) particle shape; (c) temporal (June 11-18, x axis) evolution of particle mass values at the ground as derived from in situ PM10 measurements (grey dots) and ALC at surface-level (adapted from Bellini et al., 2025, in preparation).*

More on the potential of ALC for aerosol profiling is reported in Haeffelin et al., (2022), Wagner at al., (2024) and Bellini et al., (2024), this latter including description of data QC/QA procedures and quantification of the expected accuracy of ALICEnet products.

The results obtained thanks to RI-URBANS clearly show how use of ALC (PLC) systems is promising for a low-cost and geographically dense aerosol profiling of interest for AQ issues. The experience within the Italian network ALICEnet also demonstrate these systems could be easily operated by regional/national EPAs within Member States AQMN. From the ACTRIS side, the CARS topical centres already deal with the ALC QA procedures, which are mature for cloud remote sensing applications, but not yet consolidated for the aerosol profiling, so that ALC aerosol data are being provided for the time being only as experimental/research product and mostly as 'case study-related' (e.g. the Paris example in Figure 1). Such data will be provided through ACTRIS ARES DC using the opportunity offered by the ATMO-ACCESS homeless data portal by the end of Ri-URBANS.

More details are provided for standardized and harmonized data obtained with high power lidars (HPL) as this is a service provided routinely by ACTRIS RI.

3.2 High power lidars requirements and SOPs

High-power aerosol lidars are often located and operated at suburban or rural sites and are useful for characterizing background conditions. In several locations lidars are based in the urban environment (e.g. Warsaw and Wrocław ACTRIS facilities in Poland). These instruments are typically high-power, multi-wavelength (backscatter at 1064, 532 and 355 nm, extinction at 532 and 355 nm, depolarization at 355 and 532 nm) systems. When complemented with multi-wavelength photometers, aerosol concentration profiles can be also derived.

<u>Site requirements</u> defined within ACTRIS are:

- A high-power aerosol lidar should be operated together with an automatic sun/sky/lunar photometer. These instruments should be collocated, i.e., situated at the same site to measure the same atmospheric layer. The maximum allowed horizontal distance is 1 km.
- The automatic sun/sky/lunar photometer requires clear field of view with a maximum 10° elevation mask in all the directions is requested. In addition, the field of view of the instrument in the South direction should be not obstructed.
- Reliability of internet (stable) connection and power (uninterruptible) supply are required.
- Compliance with local safety and security rules: high power aerosol lidar may need specific security clearance because their lasers are not eye safe. Regulations applicable for the specific measurement location should be checked (e.g. ANSI and ISO standards for outdoor laser operation).

<u>Technical/operational requirements</u> defined within ACTRIS are:

- Minimum setup: Aerosol remote-sensing station consists of a one-wavelength Raman with polarization discrimination capability and a sun/sky photometer. The photometer must be set up for automatic operation.
- Optimum setup: Aerosol remote-sensing station consists of a three-wavelength Raman or high spectral resolution lidar with polarization discrimination capability and an automatic sun/sky and moon photometer (optionally also polarized) according to ACTRIS/AERONET (Aerosol Robotic NETwork) standards, both operating continuously.
- Measurements of aerosol extinction, backscatter and depolarization-ratio profiles are to be performed, at least, at one wavelength, either 355 or 532 nm.
- Technical system parameters such as laser power, telescope aperture, receiver bandwidth and data acquisition system must be chosen such that profiles can be acquired throughout the troposphere up to the lower stratosphere with the required accuracy and temporal and spatial resolution. A separate near range receiving system is recommended for observations in the lower planetary boundary layer. Configurations may vary to account for climatic circumstances, e.g., the typical height of the boundary layer for the location.

• It is recommended to operate the aerosol high-power lidar and the sun/sky/lunar photometer continuously, weather permitting. If the instrument is not automated, it must at least provide unbiased long-term regular observations following the pre-defined schedule with 5 observations per week, each with a duration of minimum 3 hours (details available in Guidelines available here https://www.actris.eu/topical-centre/cars/announcements-resources/documents)
• The photometers are fully automatic and follow standard measurement protocols.
• Use of centralized ACTRIS ARES (Aerosol Remote Centre) processing (namely the Single Calculus Chain - SCC) is required for standardized fully quality controlled aerosol profiles.

- Overlap below 300m (optical profiles) for allowing low altitude investigation.
- Minimum high altitude > 15 km (range corrected signals) for allowing detection and characterization of the long-range transported aerosols in the troposphere and lower stratosphere.
- Height resolution raw <15 m (recorded signal) for allowing layer and cloud discrimination.
- Time resolution raw < 60 s (recorded signal) for allowing cloud screening.
- Specific Requirement for Aerosol typing and Aerosol concentration profiles
  - Depolarization profile at least at 1 wavelength (aerosol typing and concentration profiles).
  - Co-located photometer being part of AERONET (microphysical properties and concentration profiles).
  - ACTRIS/EARLINET format (optical profiles, aerosol typing).

Further and more detailed requirements for the implementation of high-power aerosol lidars can be found in the guidelines and standard operating procedures of the ACTRIS Centre for Aerosol Remote Sensing (ACTRIS High power lidars: Standard Operation Procedures, 2023).

3.2.1 Quality assurance, quality control and measurement uncertainty

Maintenance, checking and calibration procedures to follow are specified in the ACTRIS CARS (Centre for Aerosol Remote Sensing) documentation.

In particular, several tests are to be executed at the stations, and then checked and qualified by the CARS experts:

The **Telecover test** is used to check the laser-telescope alignment of the aerosol high-power lidars (AHL): Deviations of near range signals from different parts of the telescope and the comparison of such deviations of different lidar channels and with theoretical ray-tracing simulations can reveal the distance of full overlap and possible reasons for the deviations from the ideal case. Telecover test to be done every 6 months or after each instrument upgrade. *Schedule*: Every 6 months and after major upgrades

Polarization calibration: the calculation of the volume linear depolarization ratio profile (VLDR) and particle linear depolarization ratio profile (PLDR) requires calibration of the polarization sensitive lidar channels. In order to be able to perform a polarization calibration, the AHL must be equipped with a polarization calibration module. A reliable solution for calibrating the polarization measurements is represented by the and photo. This calibration implements a 45° rotation of the polarization analyser (polarization beam splitter and photomultiplier tube) with respect to the polarization plane of the laser in order to equalize the light intensity in the cross and parallel channels. *Schedule*: Every 6 months and after major upgrades.

Rayleigh-fit test: the Rayleigh-fit is a normalization of the range corrected lidar signal to the calculated attenuated molecular backscatter coefficient (βm attn, Rayleigh signal) in a range where we assume clean air without aerosols and where the calculated signal fits the lidar signal within the noise limits. The comparison of lidar signals in clean air ranges with the signals calculated from air density and temperature profiles from radiosondes is the only absolute calibration of lidar signals. To be able to calibrate lidar signals with the so-called Rayleigh (molecular) backscatter signals, the optoelectronic detection systems must have a high dynamic range. *Schedule*: Every 6 months and after major upgrades.

Zero-bin test: A trigger-delay between the actual laser pulse emission and the assumed zero range of the signal recording (zero bin) can cause large errors in the near range signal up to about 1 km range. Especially the inversion of the Raman signals can be distorted dramatically, because the signal slope in the near range changes very much when the zero-bin for the range correction is varied. Hence it is worth some effort to verify that the zero-bin is really where we assume it to be. *Schedule*: once and after major upgrades.

Extended Dark Signal measurements: If signal distortions are independent of the lidar signal itself but synchronous with the laser repetition, they can be determined with so-called dark-measurement. The measured dark-signals without atmospheric backscatter from the laser can be subtracted from the regular lidar signals just as the skylight background or the analogue DC-offset, but as a range dependent offset. *Schedule*: Every 6 months (analogue) and after major upgrades (analogue or pc).

All these aspects (calibration, dark signal and so on) are sources of uncertainty that, by being monitored, can be in some way estimated. Other sources of uncertainties are related to the assumptions needed in the optical properties' determination starting from the acquired lidar signals. Estimations of such errors are reported in literature (as example in Mona et al., 2012).

Additionally, statistical errors related to the signal acquisition process lead to an overall statistical error in the final product, which is calculated (typically through a Monte Carlo process) into each one of the final products: each data product reported in the ACTRIS/EARLINET database is accomplished with its own statistical uncertainty.

Quality check procedures are automatically and centrally carried out on each data file. The current quality check procedure automatically working on the data during the submission phase is described in (EARLINET Data Quality Check Procedure - v3.1, 2024). This is version 3.1 of the QC working on ACTRIS/EARLINET database. The history of the quality check versions is reported in (EARLINET Data Quality Check- Action report, 2024).

The following main aspects are checked in ACTRIS/EARLINET data:

- Technical quality controls (BQC) are executed to ensure the product is compliant from the technical point of view according to the defined standard.
- Advanced quality controls (AQC) are executed to assess the quality from a physical point of view of the product.
- [optional] multi-product quality controls (MPQC) are executed to assess the quality from a physical point of view of the product, by comparing different products from the same measurement. This step can only be applied to the products that are being submitted automatically from the SCC (Single Calculus Chain), the official analysis tool of the network)

Specifically, AQCs check that:

- absolute errors are not negative,
- positive-defined quantities are positive within 3 standard deviations,
- no unphysical peak values are recorded (unless the scene is classified as cloud contaminated one),
- intensive properties are in the established validity range interval within 3 standard deviations,
- the atmospheric density profile used in the retrieval is estimated by model forecast,
- the lidar system and its set up are approved by CARS.

The multi-product quality controls check that different backscatter profiles are consistent with each other, and that lidar ratio and Angstrom exponent resulting from different data files are consistent within 3 standard deviations from a physically meaningful range of values.

3.3 Data management summary

The ACTRIS/EARLINET database (https://data.earlinet.org) is one of the major archives for data on atmospheric aerosol profile worldwide hosting data from EARLINET network which is a regional GAW network. In collaboration with the other GALION – GAW regional networks, ACTRIS/EARLINET developed standards, procedures, nomenclature and vocabulary for aerosol lidar data and products.

All ACTRIS/EARLINET data are open and freely available and can be used under CC-BY-4.0 license.

Networks outside Europe work with data, service and digital tools developed by the ACTRIS aerosol remote sensing data centre, demonstrating the interest of a large scientific community.

Standard formats and data products are defined in a Level structure by ACTRIS (ACTRIS Data Management Plan https://github.com/actris/data-management-plan/blob/master/DMP/ACTRIS-DMP.md).

ACTRIS ARES centralized data processing (namely the SCC - Single Calculus Chain) (https://www.earlinet.org/index.php?id=281) is available for all ACTRIS/EARLINET stations and can be offered to externals under agreements and/or as a service.

Aerosol optical profiles are available through the ACTRIS data portal at https://data.actris.eu and through the specialized ACTRIS ARES DC data portal https://data.earlinet.org. All the products are available through REST API working on top of ACTRIS ARES DC (https://data.earlinet.org/api/swagger-ui/#).

Quicklooks are also available providing a quick insight of the aerosol presence and main properties over the stations (https://www.earlinet.org/index.php?id=295)

Additional interesting quantities, as the ones reported in section 4, can be obtained from aerosol optical products reported in the ACTRIS/EARLINET database either as Level 1 or Level 2 data. **Level 1 data** are calibrated and quality assured data with minimum level of quality control, while **Level 2 data** are approved and fully quality controlled ACTRIS data product or geophysical variable. Namely Level 1 optical products passed all technical quality controls, while Level 2 data passed both technical and advanced + if feasible multiproduct quality controls.

Level 3 climatological datasets are obtained as aggregated products from the fully quality controlled (QC) aerosol optical products (i.e. Level 2 products). For RI-URBANS purposes, anew release of the ACTRIS/EARLINET climatology has been released with associated DOI, and related software package has bene made freely available on GitHub (https://github.com/actris-ares/actris-level3-aerosol-profiling-climatology).

A dataset of **aerosol typing profiles** have been elaborated within RI-URBANS based on optical properties fully compliant with ACTRIS/EARLINET quality controls (i.e. Level 2 products) acquired in 2015-2023 period. Such a long-term network-wide dataset is going to be associated to a DOI and provided as an advanced dataset through the ACTRIS ARES DC.

Finally, vertical profiles of the fine and coarse mass concentration are going to be obtained (release expected by Autumn 2024) using the combination of collocated aerosol lidar profiles and AERONET photometer measurements through the application of the GARRLiC algorithm (Lopatin et al., 2013).

Within RI-URBANS, all available data processed using the centralized processing chain are going to be made available through the ACTRIS-ARES DC.

4. PAN-EUROPEAN OVERVIEW OF AEROSOL PROFILES ACROSS URBAN ENVIRONMENTS

4.1 Introduction

ACTRIS/EARLINET database contains currently about 200.000 aerosol optical properties being therefore the largest ground-based database of aerosol profiles. The data covers a period exceeding 20 years and 40 stations even if stations do not provide data for the whole period; being measurements often done on a voluntary basis, some gaps in observations are present. Due to their cost, measurements, especially in the first decade of EARLINET activities, were sparse (3 3-h slot of measurements per week + special event observations). Since 2020 the scheduled measurements have been doubled and a significant number of stations are operating 24h/7d.

This wide database allows for long term investigations, which of course needs high quality measurements, guaranteed by the QA/QC operated by ACTRIS (ACTRIS High Power Lidar SOPs, 2023; EARLINET Data Quality Check Procedure - v3.1, 2024).

All the QA/QC procedures, data services and tools previous described are contributing to feed the ACTRIS/EARLINET database with more and more profiles of aerosol optical properties.

In addition, within RI-URBANS specifically 2 main datasets and related investigations have bene obtained and are here described: climatological analysis of aerosol optical properties collected over Europe in 2000-2019, with a focus on highly polluted regions (like Ispra in the Po Valley) provides insight into the aerosol presence and its vertical distribution in the long term, and long-term analysis of aerosol typing profiling over Europe in 2015-2023 period.

In the following, a short description of the methodology for obtaining such datasets and results of these data investigations are reported.

4.2 Aerosol optical properties climatology over Europe 2000-2019

4.2.1 Methodology

This study is based on EARLINET long term observations performed in 2000-2019, namely the Level 3 climatological products (EARLINET Level 3 Data Product Catalogue, 2022). More details are being provided in a paper in preparation by Mona et al., 2024. The Level 3 standard products contain climatological datasets obtained as aggregated products from the fully quality controlled (QC) aerosol optical products (i.e. Level 2 products) (EARLINET Data Quality Check v3.0, 2017). Methods for guaranteeing the high quality of the data and to avoid biases due to the not continuous measurements performed at ACTRIS/EARLINET stations are applied and described in devoted documentation (EARLINET Level 3 Algorithm Theoretical Basis Document, 2019).

For each station, three types of data are released: profile values, integrated quantities, layer statistics. Thirty-three stations have been considered for the Level 3 products and are reported in Table 1. For each type of data, four different temporal aggregations are provided: seasonal, annual, normal seasonal, normal monthly. *Normal* means that the statistical products are computed for a uniform and relatively long period, following the WMO definition (WMO, 2017). They are of big interest, firstly because they form a benchmark or reference against which conditions (especially current or recent conditions) can be assessed, and secondly because they are widely used (implicitly or explicitly) as an indicator of the conditions likely to be experienced in a given location and in a given time period.

More details about the Level 3 climatological products are reported in the Level 3 documentation available at the EARLINET website (EARLINET Level 3 Data Product Catalogue, 2022) and data are published as a collection and single station datasets (ACTRIS/EARLINET Level 3 2000-2019 climatological dataset, 2024).

4.2.2 Highlights

Level 3 climatological dataset for the 2000-2019 period contains data from 33 stations over Europe and beyond (Dushanbe) as reported in Figure 2.

In the following, examples for stations identified as of interest for urban conditions considering their population in the area are reported, and compared to stations in clean conditions to be considered as a reference/background. As representative of urban conditions, the following stations are reported Madrid (ES), Barcelona (ES), Ispra (IT), Naples (IT), Athens (GR), Thessaloniki (GR), Warsaw (PL), Bucharest (RO). Paris and Rome, even if present in the climatological datasets were not included because of the few data available.

Leipzig and Lecce stations are also reported as representative for clean conditions of a continental "standard" site and a marine station.

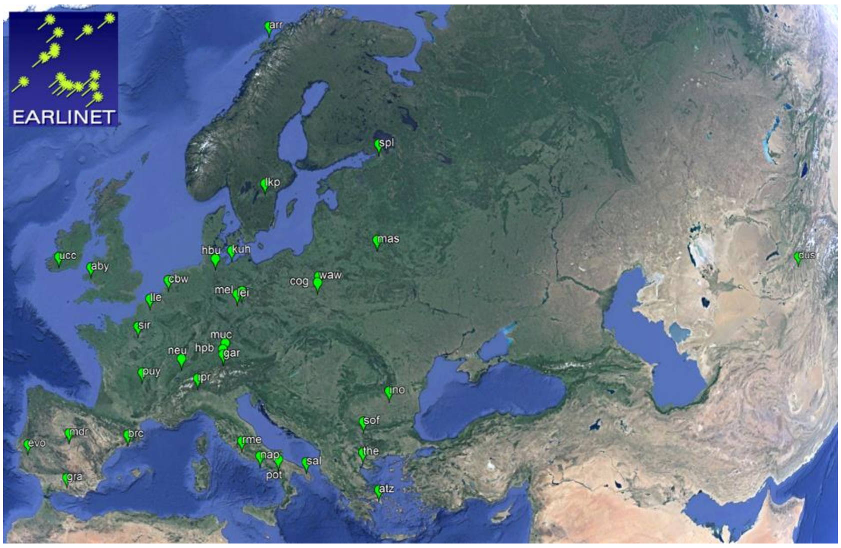

Figure 2. *ACTRIS/EARLINET stations included for which Level 3 climatology is available.*

Figure 3 reports normal months average profiles at 532 nm for each station, showing the seasonal dependence as observe during the last 20 year and relative differences between the stations. As reference of clean conditions, Granada, Leipzig and Lecce station behaviors are also reported as representative of continental “standard” site and marine station.

The false colour images of backscatter coefficient (Figure 3 and 4) provide a direct insight of the presence in the atmospheric column of the aerosol particles: warm colours indicate the presence of high amount of aerosol, while cold ones lower aerosol content until reaching the blue where the aerosol presence can be considered negligible.

Apart from signatures of aerosol presence above 6 km, due to special events of long range transported aerosol particles, most of the aerosol content is confined up to 4-5 km of altitude. The presence of aerosol up to higher altitude during warm period is common to all the reported sites. Anyhow, it is observed a tendency of more pronounced trapping of aerosol closer to the surface at big cities locations (first two columns). This is clearly evident in the case of Naples, a metropolitan area located close to sea. Comparing the false colour maps in Figure 3 with the corresponding Lecce’s one (smaller city on the sea) it is clear the presence of intense red area close to ground for Naples while the yellow dominates with different nuances for Lecce, even if all the 3 show the same general pattern with higher altitude affected by aerosol in June-August period.

The same behaviour is observed for Iberian region (Barcelona and Madrid compared to Granada) and for central-East Europe (Bucharest and Warsaw vs Leipzig). To be noted that the high aerosol content observed over Granada is probably related to the considerable presence of dust being in a very arid region and strongly affected by desert dust.

Particular attention has to be paid to Ispra observations: even if located in Italy this site being close to the Alps shows a seasonal behaviour closer to the Central-Europe one which 2 peaks, but, more important, the aerosol backscatter values are significantly higher at all altitudes of the other sites (to be noted that the scale is uniform across the plots) confirming the extreme polluted conditions of the area. It is worth mentioning here that the presence over such site of a relevant aerosol content for almost the whole year up to 3 km, makes it a potential source of pollution for close by areas: air masses can easily pick air at such altitudes and bring far away polluted air from such hot spot.

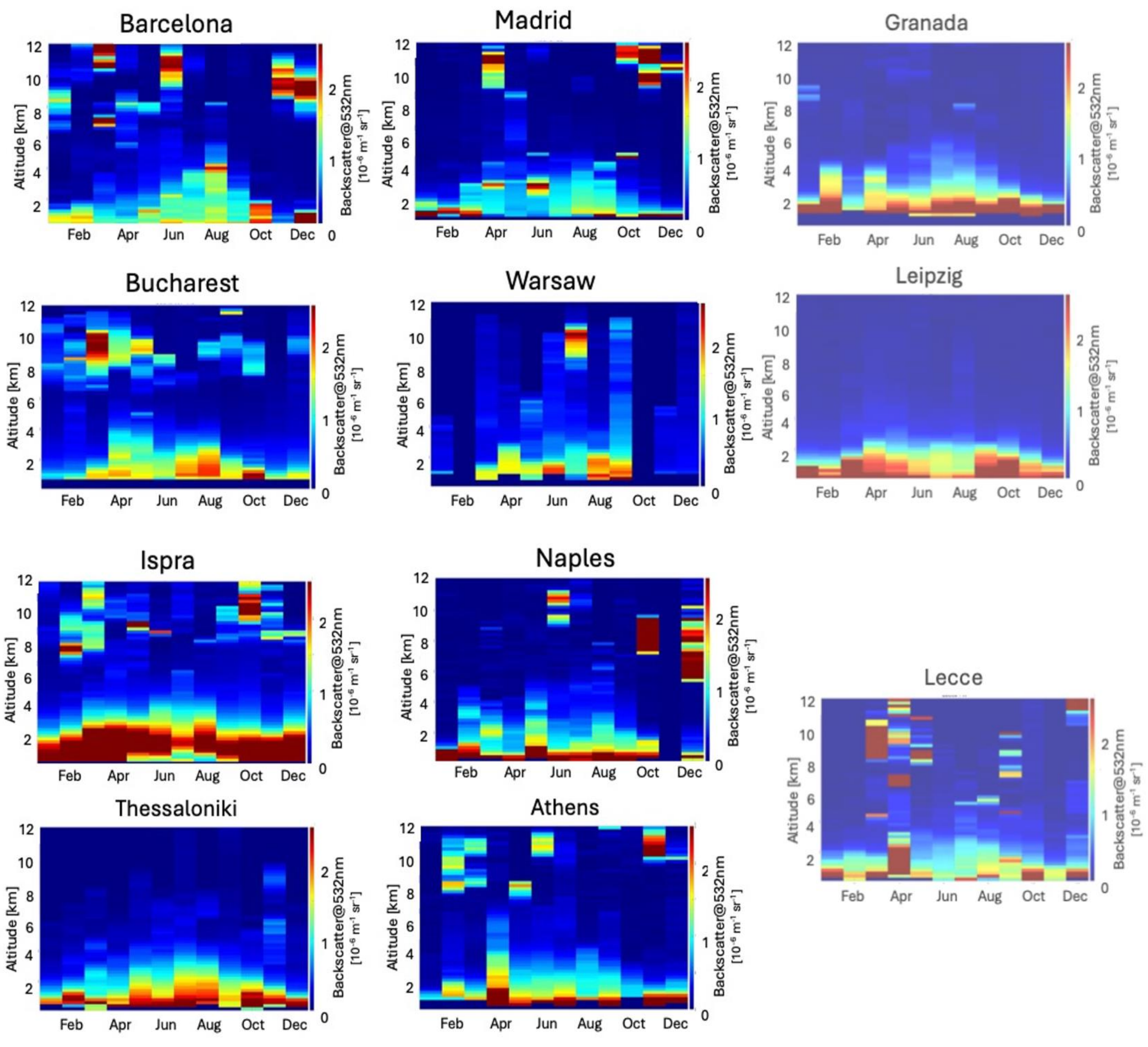

Figure 3: *Climatological monthly averages of the aerosol backscatter profile at 532 nm in suburban locations close by 8 big cities: Barcelona, Madrid, Bucharest, Warsaw, Ispra, Naples, Thessaloniki and Athens. In the third column the same quantity is reported, as partially transparent, for 3 sites far from big cities located in the same macro-area of the previous reported 8 ones.*

Figure 4 reports the annual averages of aerosol backscatter profile at 532 nm as mean for investigating potential trends. Apart from Ispra which shows different behavior even in this case, all the sites (both the 8 big cities and the reference ones) observed a slightly decreasing trend in the altitude of the transient between aerosol presence and aerosol free zone in the atmospheric column and a shift of the colors from warm to mid intensity colors

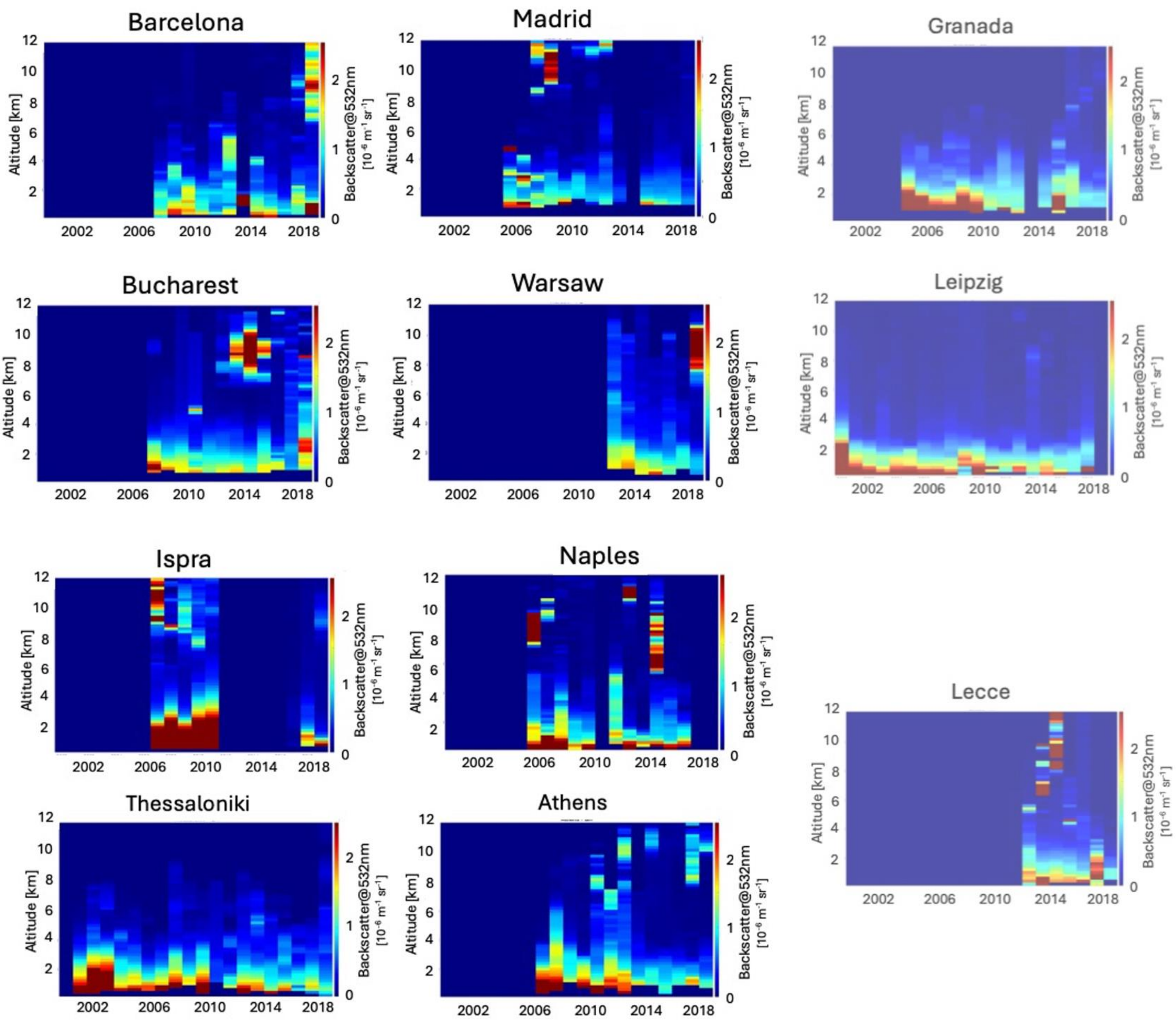

Figure 4. *Annual averages in the period 2000-2019 of the aerosol backscatter profile at 532 nm in suburban locations close by 8 big cities: Barcelona, Madrid, Bucharest, Warsaw, Ispra, Naples, Thessaloniki and Athens. In the third column the same quantity is reported, as partially transparent, for 3 sites far from big cities located in the same macro-area of the previous reported 8 ones.*

4.3 Aerosol Typing over Europe: long term analysis

4.3.1 Methodology

A dataset of aerosol typing over Europe for the 2015-2023 period is provided within Ri-URBANS. The aerosol type profile is defined as a vertical sequence of aerosol classes (like continental, continental polluted, smoke, dust, marine) which are predominant between tops and bottoms of aerosol layers in the troposphere. Aerosol type profiles are of interest for AQ because the long-range transport of aerosols may affect the air quality at the ground, even if they are not emitted locally (e.g., Janicka et al., 2023).

The NATALI (Neural Network Aerosol Typing Algorithm based on LIdar data) method (Nicolae et al., 2018) I was used. NATALI is a method to retrieve aerosol type profiles using a neural network algorithm applied to Lidar data. High power lidars with multiwavelength Raman and depolarization capabilities can provide a solid aerosol typing classification. If depolarization is missing the classification is still possible even if with less fine aerosol classification.

NATALI comprises first a module for the identification of aerosol layers and then it identifies the aerosol type on the base of aerosol intensive parameters measured by the aerosol lidar. In order to identify the aerosol type the measured intensive parameters and the whole set of them should satisfy some criteria about statistical errors, amount of aerosol in the identified layer and consistency of the intensive parameters respect to literature, climatological and modelled values used for training the NATALI algorithm.

4.3.2 Highlights

Lidar datasets collected between 2015 and 2023 by 26 ACTRIS/EARLINET stations were used as input. Methodology and results are the subject of a paper in preparation by Nicolae et al., 2024. In the analysis, only fully quality controlled, namely level 2, ACTRIS/EARLINET data products (EARLINET Data Quality Check v3.0, 2017) are used.

We analysed the results for the lower and upper troposphere separately to highlight local influences and long-range transport. In the following, “low troposphere” (LT) means the first identified layer closest to the ground, it is the lowest aerosol layer the lidar can see and is including the boundary layer region.

“Higher troposphere” refers to all the other layers, higher than the first one, which are decoupled from the ground and potentially carry particles transported from large distances.

To improve the statistical significance, the stations were grouped according to their geographical location and specific regions were defined: a) a region in the belt of latitudes between 52 and 63 degrees north: including maritime climate (Northwest Europe) and central Europe climate (Northeast Europe); b) three regions in the belt of latitudes between 43 and 52 degrees North: West Europe (including maritime climate), Central Europe (including central Europe climate), East Europe (including continental climate); c) three regions in the belt of latitudes between 35 and 43 degrees North: West Mediterranean (including oceanic climate), Central Mediterranean, and East Mediterranean (including continental climate).

The location of the contributing stations is represented in pins and contained in colour coded frames in Figure 5, according to the cluster region to which the station belongs to. The size of the markers in Figure 5 represents the number of aerosol layers typed with high confidence and used in the following analysis.

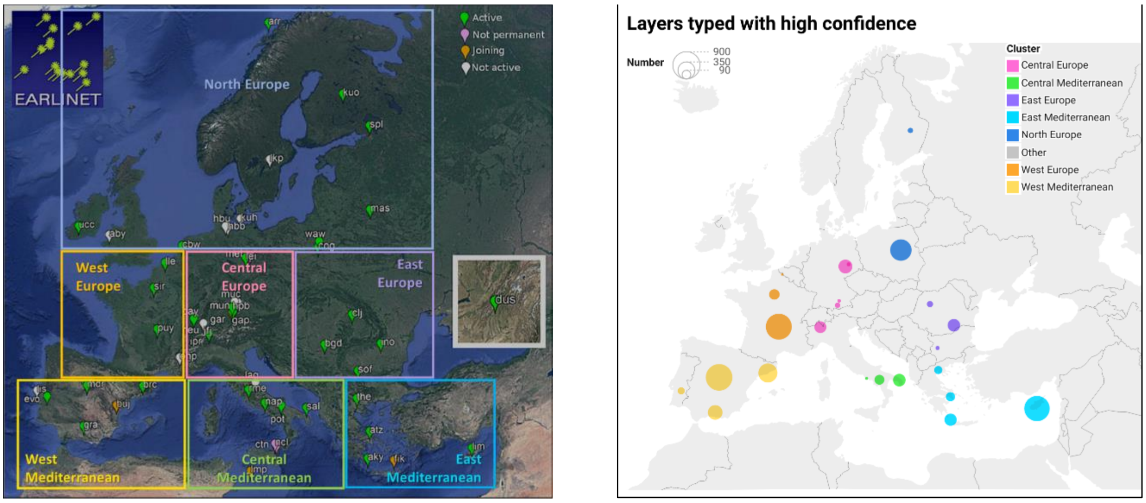

Figure 5. *Distribution of stations across cluster regions: EARLINET map (left panel); number of layers typed with high confidence, for each participating station (right panel)*

The NATALI software was used to identify the predominant aerosol type in the mixture of each layer. Because the calibrated depolarisation was not generally available in the datasets (only in the recent years the quality assurance of the particle linear depolarisation was established), it was not possible to use NATALI in high resolution mode. It means that only the type predominant in the mixture could be identified. In low resolution mode, NATALI accounts for the following basic aerosol types: continental, continental polluted, dust, marine/clean continental, smoke, and mineral mixtures of various kinds.

Out of the 19989 aerosol layers identified in the available dataset, 7717 presented all spectral parameters that are required for NATALI (about 39% of the total number of layers), and only 1922 layers fulfilled all quality criteria, meaning about 25% of the total number of layers typed by NATALI.

Figure 6 shows the statistics of the identified predominant aerosol types for each cluster region, separately for the low and high troposphere.

Based on these results, in the lowest part of the atmosphere, local aerosol sources are the main contributor. Continental aerosol is predominant in Central Europe, West and Central Mediterranean. Smoke is predominant in East and West Europe, and East Mediterranean, while continental polluted is predominant in Central Mediterranean and North Europe. Contribution of dust particles is present in the West and East Mediterranean but also in East and North Europe. Note that not using in this study the depolarization capability desert dust occurrence is probably underestimated because under certain circumstances it can assume lidar ratio and colour ratio similar to smoke and/or continental pollution. This is surely in the case of Central Mediterranean where many papers showed a relevant present of desert dust here not identified (e.g. Mona et al., 2014).

In the high troposphere, transport of particles from remote areas becomes important. Dust particles are measured in all regions, smoke is identified everywhere except Central Mediterranean (where it is maybe misclassified as continental polluted), while marine/clean continental particles seem to be frequent in some regions.

Seasonal variation of the predominant aerosol types in the low and high troposphere are shown in Figure 7.

In the low troposphere, as expected, smoke and continental polluted are more important during summer and autumn due to vegetation fires, while dust is almost constant, with slightly large percentages in spring. The large relative occurrence in winter is probably due to the fact that lidar measurements are inhibited in bad weather conditions, so a lower number of measurements are usually collected in winter and typically during high pressure conditions when dust intrusions from Northen Africa are possible.

In the high troposphere the seasonal variation follows similar patterns with much less continental polluted and more marine/clean continental cases. A particularity of the high troposphere is that continental polluted contributes more in spring and summer, and not so much in autumn as is the case of low troposphere. This might be related to the higher planetary boundary layer height during warm seasons, favouring the uplift of the pollution to upper levels in the atmosphere. Long range transport of mineral dust from North Africa adds up during spring and summer, while smoke particles transported through various routes (Canada, Russia and Ukraine, Greece) are frequent in the high troposphere during summer and autumn (Hu et al., 2019).

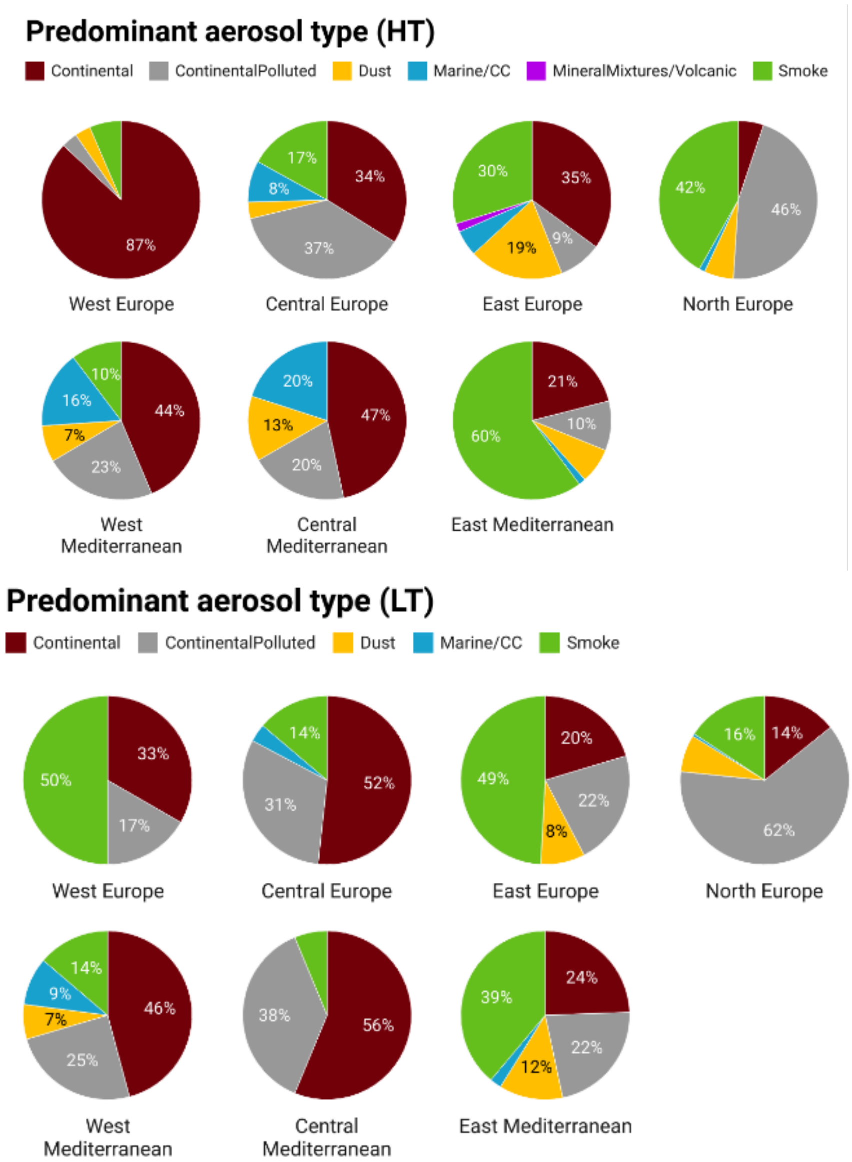

Figure 6. *Predominant aerosol types for each cluster region: in the low (bottom panel) and in the high troposphere (top panel).*

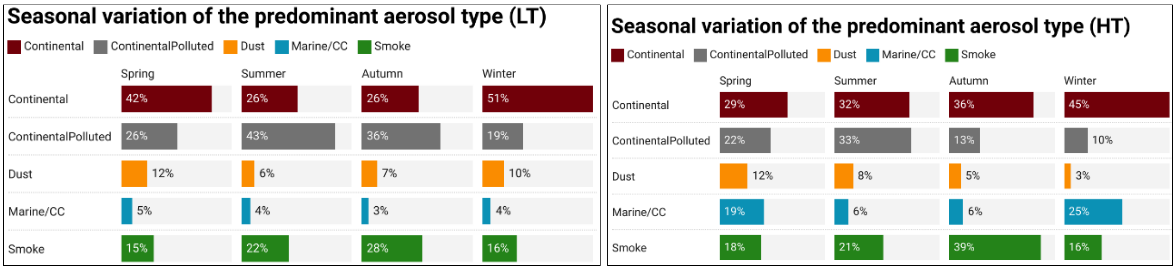

Figure 7. *Seasonal variation ranges of the predominant aerosol types: in the low (left panel) and in the high troposphere (right panel).*

5. RECOMMENDATIONS

Aerosol profiling data collected over Europe demonstrates the relevance of getting information on the vertical distribution of the aerosol and their typing. ACTRIS high power aerosol lidar systems are powerful in this sense providing quality assured/quality control profiles of aerosol optical properties with no critical assumptions. Ceilometer and ALC data are very promising for filling in the gaps related to the sparse geographical coverage and not continuous lidar measurements at ACTRIS aerosol remote sensing. Anyhow such measurements are affected by different issues, as stability of the instruments, low signal-to-noise ratio, lacking of QA/QC procedures for aerosol profiling, no quality assured procedure for the particle depolarization ratio, and needs of many assumptions for aerosol properties retrieval, so that ceilometer data vary valuable for the description of aerosol vertical distribution, should be used with careful attention when aerosol properties are retrieved from such observations.

High power lidars instead are typically not operable in densely populated areas like urban locations, but their observations can be used as reference in local pollution investigation and climatological studies of interest for AQMN. In particular, high power lidar observations are highly valuable for tracking intrusions of long range transported aerosol particles. So that typically of interest of the AQMNs are more the data products, and post processing products of the high power lidars, even if the possibility to run less complex and commercialized aerosol high power lidar is feasible for AQMN. Basic and essential elements for an aerosol high power lidar (as reported in ACTRIS SOPs) are the presence of one Raman and one depolarization channel at the same wavelength, allowing backscatter extinction and particle depolarization data profiles the same wavelength. For air quality issues, low overlap is highly desirable.

About retrievable products, the vertical profiles of fine and coarse concentration profiles can be retrieved only if in addition a photometer is available, while an accurate aerosol typing can be obtained only if the lidar is a multiwavelength Raman system.

5.1 Maintenance and quality control

Maintenance and operation are key issues because of the wide difference technically existing among the different high power lidars. ACTRIS provides all the tools and documentation for setting, running and checking an aerosol high power lidars: guidelines and recommendation for setting up such kind of system and standard operating procedures are provided by CARS and ARES (ACTRIS High power lidars, 2023). Quality control procedures performed on data for guaranteeing the quality of data products are implemented, fully documented (EARLINET Data Quality Check- Action report, 2024) and are running automatically on ACTRIS aerosol remote sensing data.

Calibration and quality assurance tests are handled by ACTRIS CARS Topical Centre, while ACTRIS ARES DC is responsible for aerosol remote sensing data curation, access and provision.

CARS is also the Topical Centre in charge of QA for the ceilometer in the characterization of aerosol profiles. Still work about this topic has to be done and activities are in progress. Similarly, aerosol products eventually provided by ALC in ACTRIS will be provided by ACTRIS ARES DC.

5.2 Data management summary

ACTRIS Aerosol profiles are handled by ACTRIS ARES DC, as described in the ACTRIS Data Management Plan (https://github.com/actris/data-management-plan/blob/master/DMP/ACTRIS-DMP.md).

Data are freely available as open data to all the users. Aerosol optical properties profiles (aerosol extinction, backscatter, depolarization ratio and lidar ratio) are provided through the ACTRIS data portal at https://data.actris.eu and through the specialized ACTRIS ARES DC data portal https://data.earlinet.org. All the products are also available through REST API working on top of ACTRIS ARES DC (https://data.earlinet.org/api/swagger-ui/#): this service tool offers an easier way to download massive data of files.

At today almost 200.000 profiles of aerosol optical products are offered by ACTRIS. This number is continuously improving because about 30 stations across Europe and beyond are providing data, in some cases also in NRT.

Centralized processing chain is available at ACTRIS ARES for the analysis of lidar and photometer data for providing aerosol profiles, which are then classified as Level 2 data if approved and fully quality controlled ACTRIS data product or geophysical variable, and Level 1 data otherwise.

The API system working at ARES offers the possibility to access machine-to-machine also the data products which are not optical products, which are at the present, pre-processed signals at low and high resolution, and cloud masking.

The API will provide the access also to vertical profiles of the fine and coarse mass concentration. These are going to be obtained (release expected by Autumn 2024 at ACTRIS ARES DC) using the combination of collocated aerosol lidar profiles and AERONET photometer measurements, applying GARRLiC algorithm (Lopatin et al., 2013).

Climatological datasets (Level 3) are centrally obtained as aggregated products from the fully quality controlled (QC) aerosol optical products (i.e. Level 2 products) and provided by the ACTRIS ARES DC. The dataset is released with its DOI and it is a collection of the single station DOIs. Data and DOIs info are available at (https://www.earlinet.org/index.php?id=319). The code used for obtaining these from QC optical properties is freely available on GitHub (https://github.com/actris-ares/actris-level3-aerosol-profiling-climatology).

Specifically obtained for RI-URBANS, ceilometer aerosol profiles obtained within RI-URBANS are considered research products and will be provided thanks to the Homeless Data portal service offered by ATMO-ACCESS project through ACTRIS ARES DC by the end of the project as Level 1 data.

Aerosol typing profiles on Europe for 2015-2023 period are provided within RI-URBANS based on optical properties fully compliant with ACTRIS/EARLINET quality controls (i.e. Level 2 products). Such long-term network-wide dataset is going to be associated to a DOI and provided as an advanced dataset through the ACTRIS ARES DC, as result of a request of service to ACTRIS ARES in the ATMO-ACCESS project.

All the processing and curation tools offered by ACTRIS ARES can be applied to aerosol high power lidar also outside ACTRIS (as already done for EARLINET no-ACTRIS stations). The ACTRIS ARES climatological production software can be applied to no ACTRIS data. In both cases, the format of input files should follow the ACTRIS ARES data format.

6. REFERENCES

ACTRIS High power lidars SOPs, 2023. High power lidars: Standard Operation Procedures for NF operation Aerosol Remote Sensing Measurements; https://www.actris.eu/sites/default/files/inline-files/SOPs-CARS-Nov2023-v01-rev08.pdf.

ACTRIS/EARLINET Level 3 2000-2019 climatological dataset, 2024. Collection ACTRIS ARES Data Centre, Lucia Mona, Giuseppe D'Amico, Sergio Ciamprone, Ermann Ripepi, Francesco Amato, Claudio Dema, Pilar Gumà-Claramunt, Version 1 of Collection published 2024 via National Research Council - Institute of Methodologies for Environmental Analysis; https://doi.org/10.57837/cnr-imaa/ares/actris-earlinet/level3/climatological/2000_2019/all.

Atkinson, K. E., 1989. An introduction to Numerical Analysis (2nd ed.), New York: John Wiley & Sons, 720p, ISBN 978-0-471-62489-9.

Beck, H.E., McVicar, T.R., Vergopolan, N., Berg, A., Lutsko, N.J., Dufour, A., Zeng, Z., Jiang, X., van Dijk, A.I.J.M., Miralles, D.G., 2023. High-resolution (1 km) Köppen-Geiger maps for 1901–2099 based on constrained CMIP6 projections, Sci. Data, 10, 724; https://doi.org/10.1038/s41597-023-02549-6.

Bellini, A., Diémoz, H., Di Liberto, L., Gobbi, G. P., Bracci, A., Pasqualini, F., and Barnaba, F., 2024. ALICENET – an Italian network of automated lidar ceilometers for four-dimensional aerosol monitoring: infrastructure, data processing, and applications, Atmos. Meas. Tech., 17, 6119–6144, https://doi.org/10.5194/amt-17-6119-2024

Bellini, A., Barnaba, F. et al., Operational aerosol profiling in support of urban air quality monitoring: examples of applications in key European RI-URBANS pilot cities, in preparation, 2025

EARLINET Data Quality Check Procedure - v3.1, 2024. https://www.earlinet.org/index.php?eID=tx_securedownloads&p=293&u=0&g=0&t=1719480753&hash=f1a89f5fdaf608c9c0bacb784c2fc903dcf4aa45&file=fileadmin/user_upload/EARLINET_QC_v3_1_20240325.pdf.

EARLINET Data Quality Check v3.0, 2017. https://www.earlinet.org/index.php?eID=tx_securedownloads&p=310&u=0&g=0&t=1719484030&hash=22c5ba383ff73deca2519e4c33fd56477351e268&file=fileadmin/user_upload/EARLINET_QC_v3_0_20210325.pdf.

EARLINET Data Quality Check- Action report, 2024. https://www.earlinet.org/index.php?eID=tx_securedownloads&p=293&u=0&g=0&t=1719906255&hash=30fa64fdb09c14b3ec7e56f3693e5d988d5e695f&file=fileadmin/user_upload/EARLINET_Data_Quality_Check_ActionReport_20240325.pdf.

EARLINET Level3 Data Product Catalogue, 2022. https://www.earlinet.org/index.php?eID=tx_securedownloads&p=319&u=0&g=0&t=1719480877&hash=a1e42bc63de479358cf91b7408e70781d0032871&file=fileadmin/user_upload/EARLINET_Database/EARLINET_Level3_v2.0_relNov2022.pdf.

Haeffelin, M., Kotthaus, S., Mona, L., Barnaba, F., Bellini, A.C., Nicolae, D., Petzold, A., Mahnke, C., Apituley, A., 2022. D1.6: Observational methodologies for horizontal & vertical profiling for AQ purposes, RI-URBANS; https://riurbans.eu/wp-content/uploads/2022/10/RI-URBANS_D6_D1_6.pdf.

Hu, Q., Goloub, P., Veselovskii, I., Bravo-Aranda, J.-A., Popovici, I. E., Podvin, T., Haeffelin, M., Lopatin, A., Dubovik, O., Pietras, C., Huang, X., Torres, B., Chen, C., 2019. Long-range-transported Canadian smoke plumes in the lower stratosphere over northern France, Atmos. Chem. Phys., 19, 1173–1193; https://doi.org/10.5194/acp-19-1173-2019.

Janicka, L., L. Davuliene, S. Bycenkiene, I. S. Stachlewska, Long term observations of biomass burning aerosol over Warsaw by means of multiwavelength lidar, Opt. Express 31, 33150-33174 (2023)

Lange, K., 1999. Numerical Analysis for Statisticians (2nd ed.), Springer, 622p, ISBN 978-1441959447.

Lopatin, A., Dubovik, O., Chaikovsky, A., Goloub, P., Lapyonok, T., Tanré, D., and Litvinov, P., 2013. Enhancement of aerosol characterization using synergy of lidar and sun-photometer coincident observations: the GARRLiC algorithm, Atmos. Meas. Tech., 6, 2065–2088; https://doi.org/10.5194/amt-6-2065-2013.

Mona, L., Liu, Z., Müller, D., Omar, A., Papayannis, A., Pappalardo, G., Sugimoto, N., Vaughan, M., 2012. Lidar measurements for desert dust characterization: an Overview, Adv. in Met., 7, 356265, 36; http://dx.doi.org/10.1155/2012/356265.

Mona, L., Papagiannopoulos, N., Basart, S., Baldasano, J., Binietoglou, I., Cornacchia, C., Pappalardo, G., 2014. EARLINET dust observations vs. BSC-DREAM8b modeled profiles: 12-year-long systematic comparison at Potenza, Italy, Atmos. Chem. Phys., 14, 8781–8793; https://doi.org/10.5194/acp-14-8781-2014.

Nicolae, D., Vasilescu, J., Talianu, C., Binietoglou, I., Nicolae, V., Andrei, S., and Antonescu, B., 2018. A neural network aerosol-typing algorithm based on lidar data, Atmos. Chem. Phys., 18, 14511–14537; https://doi.org/10.5194/acp-18-14511-2018.

Pappalardo, G., Amodeo, A., Apituley, A., Comeron, A., Freudenthaler, V., Linné, H., Ansmann, A., Bösenberg, J., D'Amico, G., Mattis, I., Mona, L., Wandinger, U., Amiridis, V., Alados-Arboledas, L., … & Wiegner, M., 2014. EARLINET: towards an advanced sustainable European aerosol lidar network, Atmos. Meas. Tech., 7, 2389–2409; https://doi.org/10.5194/amt-7-2389-2014, 2014.

Philipp, A., Beck, C., Huth, R. and Jacobeit, J., 2016. Development and comparison of circulation type classifications using the COST 733 dataset and software, Int. J. Climatol., 36: 2673-2691; https://doi.org/10.1002/joc.3920.

Wagner, F., Geiss, A., Barnaba, F., Belegante, L., Bellini, A., Buxmann, J., Diémoz, H., Fenner, D., Kotthaus, S., Osborne, M., Ruefenacht, R., Ruiz de Morales Céspedes, J., & Van Hove, M., 2024. JND European networks observing the atmospheric boundary layer: Overview, access and impacts - Chapter 2a: Automatic low-power Lidar and Ceilometer (ALC), Zenodo, https://doi.org/10.5281/zenodo.11211873.

WMO, 2017. WMO Guidelines on the Calculation of Climate Normals; library.wmo.int.