# 1 ALICENET - An Italian network of Automated Lidar-Ceilometers

for 4D aerosol monitoring: infrastructure, data processing, and
 applications

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11 Abstract. Vertically-resolved information on aerosol particles represents a key aspect in many atmospheric studies, 12 including aerosol-climate interactions and aerosol impacts on air guality and human health. This information is primarily 13 derived by lidar active remote sensing, in particular with extensive networks currently in operation worldwide. In Italy, the Institute of Atmospheric Sciences and Climate (ISAC) of the National Research Council (CNR) established a network of 14 Automated Lidar-Ceilometers (ALCs), ALICENET, in 2015. Since then, ALICENET grew up as a cooperative effort of 15 16 Italian institutions dealing with atmospheric science and monitoring, and currently includes instruments run by regional 17 Environmental Protection Agencies, Universities, Research Centres and private companies. In the current configuration, the network makes use of both single-channel ALCs and dual channel, polarisation-sensitive systems ALCs (referred to as 18 19 PLCs). The systems operate in very different environments (urban, coastal, mountainous and volcanic areas) from Northern 20 to Southern Italy, thus allowing the continuous monitoring of the aerosol vertical distribution across the country. ALICENET 21 also contributes to the EUMETNET program E-PROFILE, filling an Italian observational gap compared to other EU 22 Member States, these generally running extended ALCs networks through National Meteo Services. In this work, we present 23 the ALICENET infrastructure and the specifically-developed data processing centralised at CNR-ISAC, converting raw 24 instrumental data into quantitative, quality controlled information on aerosol properties, ranging from attenuated backscatter 25 to aerosol mass and vertical stratifications. This setup allows to get insights into the 4D aerosol field over Italy with 26 applications from near real-time monitoring to long-term analyses, examples of which are reported in this work. Specific comparisons of the ALICENET products to independent measurements obtained with different techniques, such as 27 28 particulate matter (PM) concentrations from in-situ samplers and aerosol optical depth (AOD) from sun photometers, are 29 also included here, revealing the good performances of the ALICENET algorithms. Overall, ALICENET represents a

30 valuable resource to extend the current aerosol observational capabilities in Italy and in the Mediterranean area, and

31 contributes to bridge a gap between atmospheric science and its application to specific sectors, among which air quality,

32 solar energy, aviation safety.

## 33 1. Introduction

34 Aerosols influence the Earth system and human life in several ways. They affect the planetary radiation budget directly by extinction of solar radiation and indirectly by modification of cloud properties and lifetime, thus also influencing the 35 36 hydrological cycle (IPCC, 2022). Deteriorating Air Quality (AQ), atmospheric particles of both anthropogenic and natural 37 origin are also a main concern for human health (WHO, 2021). Furthermore, high aerosol loads reduce visibility and, during 38 major events such as desert dust storms, volcanic eruptions, and wide forest fires, can damage aircraft engines, thus 39 representing a threat to the aviation sector (e.g. Flentje et al., 2010; Papagiannopoulos et al., 2020, Brenot et al., 2021; Monteiro et al., 2022, Ryder et al., 2024). The vertical aerosol distribution is a key aspect to correctly quantify aerosol 40 effects on climate and human activities, this being related to radiative transfer and atmospheric heating rates (e.g., Fasano et 41 42 al., 2021; Fountoulakis et al. 2022), aerosol-cloud-precipitation interactions (e.g., Napoli et al., 2022), particle dispersion and 43 transformation processes (e.g., Curci et al., 2015; Gobbi et al., 2019; Diémoz et al., 2019a,b), the state of high-altitude, pristine environments (e.g., Balestrini et al., 2024). 44

45 Active remote sensing through lidar sensors is a very efficient tool to provide range-resolved, accurate profiles of aerosol 46 properties (e.g., Gobbi et al., 2001; Tesche et al. 2009; Ansmann et al., 2011). In the last decades, both ground-based and 47 space-based lidar systems have been developed and widely used for scientific research purposes, and they are expected to play an increasingly important role in climate and public health studies (Remer et al., 2024). From space, the recently 48 49 dismissed NASA-CNES CALIOP sensor onboard CALIPSO (Winker et al., 2010) provided one of the most valuable. 50 vertically-resolved, global aerosol datasets (2006-2023), that is expected to be extended by the just launched ESA-JAXA 51 mission EarthCARE (Cloud, Aerosol and Radiation Explorer, Illingworth et al., 2015), From the ground, lidar remote 52 sensing is often performed in the framework of globally distributed research networks. In Europe, a wide Aerosol Research 53 Lidar Network (EARLINET, Pappalardo et al., 2010) has been developed in the last decade, which is currently an important 54 component of the European Strategy Forum on Research Infrastructures - Aerosol, Clouds, and Trace Gases Research 55 Infrastructure (ESFRI - ACTRIS). Such a research-oriented network runs high power, multi-wavelength Raman lidar 56 systems, which were not designed for monitoring purposes. In fact, EARLINET lidar measurements are generally not 57 performed continuously, and the spatial density of the measuring sites is still insufficient to capture the high spatio-temporal 58 variability characterising aerosols.

In the last two decades, the use of automatic, low-energy, affordable and robust single-channel elastic lidars, referred to as Automated Lidar-Ceilometers (ALCs), spread out. These systems emit single-wavelength laser pulses, mostly in the infrared range, and measure the time- (thus range-) dependent radiation that is elastically backscattered by atmospheric components (molecules, aerosols, cloud droplets/ice crystals). ALCs were originally conceived to only monitor the 'cloud ceiling', but

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63 recent technological improvements enabled ALCs to provide continuous information on aerosol profiles within the 64 troposphere, including the boundary layer region, albeit with a lower Signal-to-Noise Ratio (SNR) compared to high-power research lidars. This favoured the development of extended networks of such systems worldwide, among which the NASA 65 Micro-Pulse Lidar Network (MPLnet; Welton et al., 2018), the US Environmental Protection Agency (EPA) network for 66 Photochemical Assessment Monitoring Stations (PAMS: Caicedo et al., 2020), or the Asian Dust and aerosol lidar 67 68 observation network (ADnet; Shimizu et al., 2016). In Europe, several Member States currently run dense ALC networks for 69 monitoring purposes, mostly managed by national meteorological services, such as the DWD in Germany (Flentie et al., 70 2021) and the MetOffice in the UK (Osborne et al., 2022). Recently, ACTRIS started considering automatic low-power 71 lidars as useful tools within its Aerosol Remote Sensing (ARS) component, although these systems are not vet included in 72 the relevant 'minimum' or 'optimal' setups recommended bv ACTRIS-ARS 73 (https://www.actris.eu/topical-centre/cars/announcements-resources/documents, last access: 25-07-2024). Most ALC 74 observations at EU level are currently collected and further exploited in the framework of the E-PROFILE program run by 75 the European Meteorological Services Network EUMETNET 76 (http://www.eumetnet.eu/activities/observations-programme/current-activities/e-profile/, last access: 25-07-2024). The 77 development of such an extended ALC observational capacity was further accelerated after the eruption of the Icelandic 78 volcano Eyjafjallajökull in 2010, which disrupted air transport due to the lack of readily accessible information on the 79 horizontal and vertical displacement of the aerosol plume (Flentie et al., 2010, Mortier et al., 2013), Moreover, ALCs have 80 been proven to be extremely useful in support of AQ evaluations, providing information on the vertical dilution of pollutants, 81 transboundary transport of particles from medium-to-long-range distances (e.g., Rizza et al., 2017; Bucci et al., 2018; 82 Diémoz et al., 2019a,b), secondary aerosol formation (e.g., Curci et al., 2015), or even particles reaching the boundary layer through evaporating rain (virgas, e.g., Karle et al., 2023). However, with few exceptions, standard Air Quality Monitoring 83 84 Networks (AOMNs) in the EU currently miss such profiling capability. The feasibility of filling this gap is currently 85 explored in the framework of the EC-H2020 Project RI-URBANS (https://riurbans.eu, last access: 25-07-2024), aiming at 86 the development of service tools in support to AQ monitoring in European urban areas and pollution hotspots. In fact, the 87 current ALC technology has been proven to be mature enough to allow a robust retrieval of the planetary boundary layer height (Kotthaus et al., 2023), a key parameter in AO, and evaluations are currently ongoing at the EU level to assess 88 89 readiness of ALC-based retrievals for quantitative Particulate Matter (PM) monitoring (e.g., Shang et al., 2021; Osborne et 90 al., 2024). The recently completed EC Action PROBE (PROfiling the atmospheric Boundary layer at European scale; Cimini 91 et al., 2020; Kotthaus and Bravo Aranda, 2024) supported by the European Cooperation in Science and Technology (COST) 92 was key to promote and coordinate such activities, which are now further explored within the E-PROFILE and ACTRIS 93 communities.

In Italy, an effort to coordinate ALC activities at national level and contribute to E-PROFILE has been done by the National
Research Council - Institute of Atmospheric Sciences and Climate (CNR-ISAC), which set up the ALICENET network in
2015 (<u>https://www.alice-net.eu/</u>, last access: 25-07-2024), filling an observational gap over Italy.

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97 The ALICENET measurements are particularly relevant in for Mediterranean area, this being a climatic hotspot (IPCC, 98 2022) affected by a complex mixture of atmospheric circulations (e.g., Lelieveld et al., 2002) and aerosol types (e.g., 99 Barnaba and Gobbi, 2004; Di Iorio et al., 2009; Andres Hernandez et al., 2022). ALICENET is conceived as an open 100 consortium with increasing contributions from several collaborating Partners, among which regional Environmental 101 Protection Agencies, Universities, Research Institutions, and private companies.

The present work aims at presenting the ALICENET infrastructure and its data processing chain, designed to derive quantitative and quality checked vertically-resolved information on aerosol properties and layering. The ALICENET data processing, centralised at CNR-ISAC, allows the homogeneous retrieval of aerosol properties from North to South Italy. It is based on specifically developed algorithms, taking benefit from past and ongoing collaborations with the EU ALCcommunity, particularly in the framework of the EC COST Actions TOPROF (2013-2016) and PROBE (2019-2024), the H2020 Project RI-URBANS (2021-2015), and the E-PROFILE initiative (2019-2023, 2024-2028).

108 The work is organised as follows. Section 2 describes the ALICENET infrastructure. Section 3 introduces the main data

109 processing steps and includes different examples of the relevant ALICENET products and accuracy. To facilitate the

110 reading, the detailed technical aspects of each processing step were included in separated supplement sections (S1-S6), these

111 being thus targeted to readers interested in a deep understanding of the processing chain, and possibly in reproducing it. Sect.

112 4 shows three examples of the near-real time ALICENET monitoring capability, while Sect. 5 summarises the ALICENET

113 achievements and some foreseen future developments within the network.

#### 114 2. ALICENET sites and instruments

115 The ALICENET stations are geographically distributed from the North to the South of the Italian peninsula as shown in Fig. 1. The network configuration allows the monitoring of aerosol vertical profiles over a wide range of environmental and 116 117 atmospheric conditions (e.g. urban, coastal and mountain) within the Mediterranean area. In fact, some stations are located in 118 highly anthropised areas, such as those in the Po Valley and main urban/industrial sites in Italy (Milan, Genova, Turin, 119 Florence, Rome, Taranto), some operate in coastal sites (e.g. Genova, Taranto, Lamezia Terme, Messina, Capo Granitola, 120 Catania), and other at high-altitude (> 550 m asl) stations (Aosta, Mt Cimone, Potenza, Mt. Etna). Most sites are frequently 121 impacted by desert dust advections, particularly relevant in Central and Southern Italy (e.g. Barnaba et al., 2017; Gobbi et 122 al., 2019; Barnaba et al., 2022), and by both short- and long-range transport of biomass burning plumes (e.g. Barnaba et al., 123 2011). Volcanic plumes are also registered in the ALICENET southernmost sites, mainly in the 5 stations located at the 124 foothills of the Etna volcano, and in the Messina and Lamezia Terme stations, due to their proximity to the other active 125 sicilian volcano of Stromboli.



Figure 1: Location and naming of the ALICENET stations (panel a, bullets). The yellow rectangle over Sicily in panel a) is zoomed in
panel b) to show location of the 5 stations in the Etna volcano area, from the northern to the southern foothills, down to the city of Catania.
Background Map credits: a) EUMETSAT, and b) Google Maps.

131 For homogeneity of operations, since the beginning of the ALICENET activities (set as 1<sup>st</sup> January 2016), it was agreed to 132 operate standardised systems across the network choosing the ones that allow to probe at least up to the middle troposphere. 133 also for calibration purposes (e.g., Wiegner et al., 2014; see also Sect. 3.2). The single-channel, bistatic CHM15k 134 instruments manufactured by Lufft (formerly Jenoptik ESW and now Ott Hydromet) were selected for this purpose. These 135 are bi-static ALCs with a Nd:YAG solid-state laser emitting linearly polarised light at 1064 nm, with a 5-7 kHz repetition 136 rate, a maximum vertical resolution of 5 m and a maximum range of 15 km. The only exception in this instrumental setup 137 was a modified-CHM15K prototype with polarisation-sensitive capabilities designed and developed in 2013 by Jenoptik 138 ESW in collaboration with CNR-ISAC in the framework of the EC Life+ DIAPASON project (Gobbi et al., 2019). This first 139 ever polarisation-sensitive ALC (hereafter PLC) was conceived to explore the possibility of producing an affordable, robust 140 system to be widely used in the identification and profiling of non-spherical (e.g. mineral dust) aerosol layers. The prototype 141 PLC was tested in Rome (Italy), where it has been operating successfully since then (e.g., Gobbi et al., 2019; Andres 142 Hernandez et al., 2022), but was never marketed by Lufft. More recently, PLC systems have been made available on the 143 market by Vaisala (CL61 systems, operating at 910 nm) and, due to the important capability of such instruments to 144 discriminate particle sphericity/non sphericity, these are being progressively included in ALICENET.

For both CHM15k ALCs and CL61 PLCs, the signal is characterised by high temporal and vertical resolution, with some variability depending on the system type and configuration (e.g., in ALICENET the CHM15k standard configuration implies a vertical and temporal resolutions of 15 m and 15 s, respectively). A summary table with details on the ALICENET sites and instrumentation operating therein is provided in Table 1. It includes indication of the beginning of operations in each site of the ALICENET network (joining date), or the operating period for those systems no longer active. Some systems joined the network very recently and are thus indicated as 'ready to go' as instrumental set up and data transfer to the ALICENET database is currently in progress.

Name	Lat	Lon	Altitude (m a.s.l.)	System Type	Status	Joining Date or Operating period	Reference Institution (Collaborating Institution)
Aosta 1	45° 44' 32"N	07° 21' 24"E	555	ALC (Lufft CHM15k)	active	02/05/2015	Arpa Valle d'Aosta (CNR-ISAC)
Aosta 2	45° 44' 32"N	07° 21' 24"E	555	PLC (Vaisala CL61)	active	28/07/2023	Arpa Valle d'Aosta (CNR-ISAC)
Milano Bicocca	45° 30' 38"N	09° 12' 42"E	135	ALC (Lufft CHM15k)	active	01/01/2016	CNR-ISAC (Univ. Milano Bicocca)
Milano Rubattino	45° 28' 38"N	09° 15' 41"E	110	PLC (Vaisala CL61)	active	31/05/2023	RSE (CNR-ISAC)
Torino	45° 03' 28"N	07° 39' 24''E	250	PLC (Vaisala CL61)	active	20/06/2023	Politecnico Torino (CNR-ISAC)
San Pietro Capofiume	44° 39' 12"N	11° 37' 24''E	135	ALC (Lufft CHM15k)	ended	13/12/2011 – 17/01/2015	CNR-ISAC
Genova	44° 24' 41''N	08* 53' 30"E	10	PLC (Vaisala CL61)	active	04/12/2022	Arpa Liguria (CNR-ISAC)
Monte Cimone	44° 11' 35"N	10" 42' 05"E	2165	ALC (Lufft CHM15k)	active	13/06/2022	CNR-ISAC
Firenze	43° 49' 08''N	11° 12' 06"E	60	PLC (Vaisala CL61)	ready to go		CNR-IBE (CNR-ISAC)
Roma Down Town	41° 54' 34"N	12° 29' 48"E	58	PLC (Lufft Prototype)	active	13/05/2015	CNR-ISAC (Arpa Lazio)
Castel di Guido	41° 53' 22"N	12° 15' 59''E	135	ALC (Lufft CHM15k)	ended	10/09/2013 - 18/12/2014	CNR-ISAC
Roma Tor Vergata	41° 50' 32"N	12° 38' 50"E	100	ALC (Lufft CHM15k)	active	01/01/2016	CNR-ISAC
Potenza	40° 36' 50''N	15° 43' 26''E	760	ALC (Lufft CHM15k)	active	21/03/2024	CNR-IMAA (CNR-ISAC)
Taranto	40° 29' 37"N	17° 13' 01"E	17	ALC (Lufft CHM15k)	active	01/01/2014	Arpa Puglia (CNR-ISAC)
Lamezia Terme	38° 52' 35"N	16° 13' 56"E	5	ALC (Lufft CHM15k)	active	29/11/2023	CNR-ISAC
Messina	38° 11' 41"N	15* 34' 22"E	5	ALC (Lufft CHM15k)	active	22/06/2016	CNR-ISAC (CNR-IRBIM)
Etna Acireale	37° 38' 26"N	15° 10' 55"E	12	ALC (Lufft CHM15k)	ready to go		Etna High Tech (INGV, CNR-ISAC)
Etna Piedimonte Etneo	37° 47' 31"N	15* 08' 18"E	720	ALC (Lufft CHM15k)	ready to go		INGV (Etna High Tech, CNR-ISAC)
Etna Nicolosi	37° 36' 49"N	15° 01' 11"E	730	PLC (Vaisala CL61)	active	15/03/2023	INGV (Etna High Tech CNR-ISAC)
Etna San Giovanni La Punta	37° 34' 44"N	15° 06' 11"E	350	ALC (Lufft CHM15k)	active	08/06/2022	Etna High Tech (INGV, CNR-ISAC)
Capo Granitola	37° 34' 16"N	12° 39' 35"E	5	ALC (Lufft CHM15k)	active	19/05/2021	CNR-ISAC
Catania Airport Fontanarossa	37° 27' 59"N	15° 04' 57"E	10	ALC (Lufft CHM15k)	ready to go		SAC (Etna High Tech, INGV, CNR-ISAC)

**Table 1:** ALICENET sites from northern to southern Italy, and relevant details.

#### 157 3. ALICENET data processing and relevant products

- The ALICENET data processing chain is summarised in Fig. 2, with indication of main inputs and outputs. It starts with generation of standardised and harmonised data files from instrumental raw data (using the raw2l1 tool, <u>https://gitlab.in2p3.fr/ipsl/sirta/raw2l1</u>, last access: 25-07-2024), and then proceeds with pre-processing and calibration procedures, the inversion of the ALC signal into aerosol properties, and the detection of aerosol layers. It is convenient to
- 162 first introduce the main equations and variables used in the description of the different steps.
- 163 As in any elastic backscatter lidar, the raw signal P(r,t) recorded by the ALC is a function of the distance from the emitter
- 164 (range, r) and of the observation time t, and can be described through the lidar equation:

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$$P(r,t) = r^{-2} Ovl C_L(\beta_p(r,t) + \beta_m(r,t)) e^{-2 \int_0^{1} (\alpha_p(r',t) + \alpha_m(r',t)) dr'}$$
 (1)

166 Equation 1 includes the particle (p) and molecule (m) backscatter ( $\beta$ ) and extinction ( $\alpha$ ) coefficients at the laser wavelength, 167 and some instrumental factors, embedded into the instrument-specific calibration coefficient C<sub>L</sub>. Furthermore, particularly 168 for bistatic systems (i.e., the CHM15k), measurements in the near range (generally < 500-700 m) are affected by signal 169 losses due to the incomplete superposition (overlap) of the laser beam and the receiver field of view. The term Ovl in Eq. 1 170 therefore indicates the instrument-specific overlap function used to correct the signal loss in the near range. Equation 1 171 allows to simply derive the total (i.e., aerosol + molecules) attenuated backscatter,  $\beta_{att}$ , as follows:

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$$\beta_{att}(r,t) = \frac{P(r,t)r^2}{Ovl(r,t)C_L(t)} = \left(\beta_p(r,t) + \beta_m(r,t)\right)e^{-2\int_0^t (\alpha_p(r',t) + \alpha_m(r',t))dr'}$$
(2)

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174 The complete ALICENET data processing chain (Fig. 2) includes pre-processing procedures (namely cloud screening, 175 denoising, and overlap correction; Sect. 3.1), the absolute calibration (to determine C<sub>1</sub> and, in turn,  $\beta_{att}$ ; Sect. 3.2), the 176 quantitative retrieval of aerosol optical ( $\beta_p$  and  $\alpha_p$ ) and physical (surface area,  $S_p$ , volume,  $V_p$ , and mass concentrations,  $M_p$  or 177 PM) properties (Sect. 3.3) using an ALICENET-original approach, and the detection of aerosol layers (Mixed, Continuous, 178 and Elevated Aerosol Lavers, MAL, CAL, and EALs, respectively) through the ALICENET automatic Aerosol LAver 179 DetectIoN algorithm (ALADIN; Sect. 3.4). The full processing chain is currently applied to CHM15k systems since, as 180 mentioned above, these were the ones firstly implemented in the network. A similar scheme is under development for CL61 181 systems, for which data processing is currently limited to the cloud screening and denoising, the absolute calibration to 182 monitor the stability of the instrument, and the detection of aerosol layers.



**Figure 2:** Scheme of the ALICENET processing chain from the raw (L0) data to aerosol products (L1-L3). The different colours in the processing box are used to indicate inversion steps valid for CHM15k (light green), CL61 (cyan), or both (dark green) systems. This same colour code (bounding box) is used for relevant output data products, which are further coloured from light to dark orange indicating processing level, from the more basic L1 quantities (Range-Corrected Signal, RCS, and depolarisation,  $\delta_{\nu}$ , profiles), through the L2 total attenuated backscatter ( $\beta_{an}$ ) to the L3 aerosol optical (particle backscatter,  $\beta_{p}$ , and extinction,  $\alpha_{p}$ ) and physical (particle surface area, S<sub>p</sub>, volume, V<sub>p</sub>, and mass concentrations, M<sub>p</sub> or PM) properties and layers (Mixed, Continuous, and Elevated Aerosol Layers, MAL, CAL, and EALs, respectively).

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The ALICENET processing chain is completely automatic and allows continuous monitoring of the aerosol field over Italy, with L1/L2 data visualisation accessible in near-real time through a dedicated website (https://www.alice-net.eu/, last access: 25-07-2024). Selected examples of this monitoring capability are provided in Sect. 4. The more advanced, quantitative retrieval of aerosol properties and layering (L3 products) is currently performed in post-processing and is planned to be released in the future through the ALICENET website.

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## 200 3.1 Pre-processing

201 After the input data format harmonisation, the first pre-processing steps are aimed at avoiding cloud, precipitation, and noise

202 contamination in aerosol retrievals (Sect. 3.1.1). Then data need to be corrected for overlap artefacts (Sect. 3.1.2) before

203 proceeding with the determination of the instrument-specific calibration coefficient (Sect. 3.2). The way these preliminary

204 steps are performed within ALICENET is described hereafter.

## 205 3.1.1 Cloud-screening and denoising

At the ALC laser wavelengths clouds generally produce complete extinction of the laser beam above the cloud base. Only in case of optically thin clouds the laser beam is partially transmitted above the cloud base, although in most cases the return signal has a too low SNR to be employed for aerosol retrievals. The cloud-screening applied to the ALICENET data exploits the cloud base height identified by the ALC firmware, with additional requirements to avoid the presence of cloud droplets

210 frequently observed below the cloud base. Technical details of this procedure are reported in supplement S1.

Cloud-screened profiles are then downscaled and denoised to improve accuracy of the aerosol retrievals. Indeed, as mentioned above, the ALC signal is generally collected with high temporal and vertical resolution and features a decrease of the SNR along the profile. Denoising is performed by computing signal mean and standard deviation over specific time and range windows, and filtering those data where the SNR (defined as the ratio between the mean and the standard deviation) is below a given threshold. A minimum SNR of 20% is generally set for aerosol retrievals within ALICENET. The temporal resolution of the downscaled data is tuned depending on the time scales of the processes to be investigated. It may range from 1 min for the investigation of boundary layer dynamics up to 3 hours for the identification of aerosol loaded/aerosol

218 free regions in the upper troposphere, such as within the absolute calibration procedure.

## 219 **3.1.2 Overlap correction**

220 For bistatic systems such as CHM15k, an overlap correction of the signal in the near range is required (see Eq. 1). This is 221 particularly important when ALC data are used for surface AQ applications, and especially in those conditions in which 222 particulate matter is confined in the lowermost atmospheric levels. An instrument-specific overlap function accounting for 223 signal losses is generally provided by the manufacturer ( $Ovl_{man}(r)$ ). However, it has been demonstrated that changes in the 224 instrument sensitivity rather require the use of an instrument-specific, temperature-dependent overlap correction. Within 225 ALICENET, the derivation of such an overlap correction is largely based on the procedure developed by Hervo et al. (2016). Full details on its implementation in ALICENET including additional quality control and quality assurance criteria 226 (QC/QA.OVL) added to the Hervo et al. (2016) procedure are described in supplement S2. The result is an instrument-. 227 228 range- and temperature-dependent 'overlap model'  $Oyl_{model}(r,T)$  to be used in Eq. 1.

Figure 3 shows an example of application of the overlap model on ALC data collected in Rome-Tor Vergata on 12 August 2019. This date was selected because of the high diurnal variation (15 K) of the instrument internal temperature. In Fig. 3, the continuous (24h)  $\beta_{att}$  profiles derived using both the manufacturer overlap function (panel a) and the ALICENET overlap model (panel b) are shown. It is evident that the temperature-dependent overlap model is effective in correcting the falsegradient and the aerosol overestimation in the lowermost 500 m coming from the manufacturer function.

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Figure 3: Overlap-corrected ALC profiles using: (a) the manufacturer overlap function, and (b) the ALICENET overlap correction. Data
 refer to the ALICENET Rome-Tor Vergata site on 12/08/2019.

239 A further effort to evaluate the ability of this overlap correction procedure to provide reliable results was conducted in the 240 ALICENET mountain site of Aosta, by exploiting the clean, nearly-molecular conditions often registered at this alpine 241 station. In fact, due to its location, Aosta is frequently characterised by relatively low aerosol concentrations in the 242 lowermost levels, in particular during Föhn events (e.g., Mira-Salama et al., 2008). This makes it possible to compare the 243 overlap-corrected  $\beta_{att}$  profiles with a theoretical molecular profile at very low altitudes. To perform this exercise, Föhn-244 related, aerosol-free conditions of 3-to-6 hours were identified by exploiting multi-sensor aerosol datasets (namely, surface 245  $PM_{10}$  concentrations measured by an Optical Particle Counter, OPC, and sun photometer-derived Aerosol Optical Depth, 246 AOD) and meteorological parameters (wind, pressure, Relative Humidity) from the AQMN of ARPA Valle d'Aosta 247 (Diémoz et al., 2021). For each of these selected cases, the mean  $\beta_{\text{att}}$  profiles retrieved using both the manufacturer and the 248 ALICENET overlap correction were compared with a theoretical molecular profile. Figure 4 shows results for two cases 249 (referring to 25 May 2021 and 6 October 2021) characterised by different values of the instrument internal temperature (308 250 K and 292 K, respectively) and very low aerosol loads both at the surface ( $PM_{10} < 6$  and 5 µg m<sup>-3</sup>, respectively) and along the 251 atmospheric column (AOD at 1020 nm < 0.04 and 0.03, respectively).



**Figure 4:**  $\beta_{att}$  profiles at 1064 nm derived using the manufacturer overlap function (black line) and the ALICENET overlap correction (blue line) in two nearly-molecular conditions registered in Aosta on: (a) 25 May 2021 (5-8 UTC), and (b) 6 October 2021 (9-12 UTC). The shaded areas represent the  $\beta_{att}$  standard deviations within the selected time windows. A reference, molecular-only  $\beta_{att}$  profile is also reported (green line).

Overall, the results show that, while the manufacturer overlap function is unable to properly account for signal losses and leads to unphysical values lower than the molecular profile in the firsts 750 m, the  $\beta_{att}$  profiles retrieved using the ALICENET overlap correction reasonably approach the nearly-homogeneous, nearly-molecular theoretical profiles expected in the selected episodes down to the ground.

## 263 3.2 Absolute calibration

264	Aim of the absolute calibration is the derivation of the calibration coefficient $C_L$ (see Eq. 1), which is required to convert the
265	ALC signal into quantitative aerosol information. The ALICENET calibration procedure is based on the comparison of the
266	pre-processed ALC signal with a theoretical molecular profile in aerosol-free atmospheric regions (Rayleigh calibration;
267	Klett, 1985), typically in the middle troposphere. The procedure, which is fully automatic, is made in two steps: a) search
268	for the best-suitable molecular window, and b) computation of the calibration coefficient. It was built on the E-PROFILE
269	algorithm, although some specificities and quality controls (QC.CAL) were introduced in both steps. Full description of the
270	technical implementation of these steps is given in the supplement S3.
271	Hereafter, we show two examples of successful calibrations (Fig. 5a, b) and the multi-annual record of $C_L$ (Fig. 5c) derived
272	from three ALICENET systems in northern, central, and southern Italy (Aosta, Roma, and Messina, respectively). Figures 5a
273	and 5b refer to the ALICENET calibrations of the CHM15k in Aosta on 21 May and 25 October 2017, selected as these
274	spring and autumn nighttime calibrations correspond to $C_L$ close to the maximum and minimum values over the year 2017
275	(see Fig. 5c). Figure 5c gives a more general overview of the long-term results of the calibration procedure, further revealing
276	that the three C <sub>L</sub> time series feature a similar seasonal cycle, as also observed in other European ALC networks (e.g.,
277	Buxmann, 2024). The reasons for such a yearly cycle are currently under investigation within the European ALC
278	community, also taking advantage of recent activities conducted within the EC COST Action PROBE (e.g., Van Hove and
279	Diémoz, 2024).
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Figure 5: (a, b) Examples of application of the ALICENET calibration procedure, referring to the nighttime range-corrected signals from the Aosta CHM15k on 21 May and 25 October 2017, with indication of the selected molecular windows and derived calibration coefficients ( $C_L$ ). (c) Multi-annual (2016-2022) time series of  $C_L$  derived for the CHM15k systems operating in Aosta, Rome, and Messina, and associated Loess fits (lines) used to derive the  $C_L$  values used in the operational, all-year-round data inversions.

- 294
- 295 The C<sub>L</sub> values used within ALICENET inversions are currently obtained by filtering out the seasonal cycle and keeping only
- 296 the long-term trends related to slow instrument changes (details are given in supplement S3). Once the main driver of the C<sub>L</sub>
- 297 seasonality will be better identified, it will be taken into account in the calibration procedure. For now, we prefer to use the
- 298 described approach and estimate the uncertainty associated with this C<sub>L</sub> variability (see Sect. 3.3.3).

## 299 **3.3 Retrieval of aerosol properties**

This section describes the ALICENET inversion of the aerosol optical (Sect. 3.3.1) and physical (Sect. 3.3.2) properties. Specific examples of the aerosol products at different ALICENET sites are also given and compared to a series of independent datasets in order to evaluate the relevant retrieval procedure performances.

## 303 3.3.1 Aerosol optical properties

- The aerosol backscatter and extinction profiles are calculated from the total attenuated backscatter ( $\beta_{att}$ ) profile based on the
- 305 forward Klett inversion (Wiegner and Gei $\beta$ , 2012; 2014) of Eq. 1. Since both  $\beta_p$  and  $\alpha_p$  are unknown in Eq. 1, an assumption
- 306 on the relationship linking the two variables is necessary to solve the Klett inversion. Within ALICENET, we do not fix an
- 307 a-priori, vertically-constant extinction-to-backscatter ratio (also referred to as Lidar Ratio, LR), as often done in elastic lidar
- 308 retrievals. Instead, the aerosol extinction is linked to backscatter through a specific functional relationship ( $\alpha_p = \alpha_p(\beta_p)$ ) already
- 309 presented and discussed in Dionisi et al. (2018). This was obtained at the CHM15k operating wavelength (1064 nm) based
- 310 on a large set of simulated optical properties from a continental-type aerosol model. Details on the implementation of the
- 311 functional relationship within the forward Klett inversion are given in supplement S4.1.
- 312 It is important to note that, with this procedure, no ancillary data (e.g. co-located sunphotometer-AOD) and no a-priori
- 313 assumption (e.g. selection of the LR constant value to be used) is needed in the retrieval. Therefore, a-posteriori comparison
- 314 to co-located sunphotometer-AOD provides a way to check the performance of the ALICENET optical properties retrievals.
- 315 These comparisons were performed using both short- and long-term datasets thanks to some co-located or closeby
- 316 AERONET (https://aeronet.gsfc.nasa.gov/, last access: 25-07-2024) or SKYNET (https://www.skynet-isdc.org/, last access:
- 317 25-07-2024) sun-photometers. Specific examples are shown in Figs. 6 and 7, respectively.
- Figure 6a shows the aerosol extinction profiles derived from the Rome-Tor Vergata ALC during the EMERGE-EU field campaign in July 2017 (Andrés Hernandez et al., 2022), while in Fig. 6b the corresponding ALC-derived AOD (blue) is compared with the one measured by the co-located AERONET sun photometer (grey).
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Figure 6: (a) Aerosol extinction profiles in Rome-Tor Vergata retrieved by the ALICENET inversion during the EMERGE campaign in
 July 2017, and (b) comparison between the ALICENET-derived AOD and the co-located AERONET L2 data. Both ALICENET and
 AERONET AODs are hourly averaged (error bars are the AOD standard deviations within the averaging interval).

Figure 6 shows that the time series of the two independent datasets, both averaged at an hourly resolution, agree within the expected AERONET (Giles et al., 2019) and ALICENET (Sect. 3.3.3) uncertainties. Exceptions are found during days strongly impacted by transport of Saharan dust (e.g., 9 July 2017). This is expected because, as mentioned, the functional relationship employed in the inversion was optimised for a continental-type aerosol and does not properly describe the different backscatter-to-extinction relation in presence of non-spherical particles (e.g., Barnaba and Gobbi, 2001). Also note that, despite using L2 AERONET data, the maximum sunphotometer AOD value on July 9 corresponds to a cloud-screened

time window in the ALC record. The extension of the ALICENET retrieval approach to other aerosol types and relevant testing is however planned for the future, also taking advantage of the depolarisation measurements capabilities of PLCs operating within the network.

337 Figure 7 shows a multi-annual (2016-2022), multi-site (Aosta, Roma, Messina) comparison between ALC and 338 sunphotometer AOD. AERONET L2 data were used in Rome-Tor Vergata and Messina, while SKYNET AOD data in Aosta 339 were derived taking into account the temperature correction of the POM-02 photometer as described in Uchiyama et al. 340 (2018). The AOD data were matched in time (measurements within 5 min one from the other) and averaged in time (15 min 341 average). The overall number of pairs considered in each site is reported in Fig. 7. This comparison shows that the 342 ALICENET retrieval is able to quantify the actual aerosol load in a variety of conditions. Infact, the number of data pairs lying within  $\pm 0.01 \pm 0.15$ \*AODsunphotometer from the 1:1 line is 84% in Aosta, 73% in Rome, and 70% in Messina. Some 343 344 ALC overestimations are mainly due to instrumental noise at higher altitudes, while underestimations are mainly related to 345 the presence of non-continental aerosol types, such as dust and marine particles in Messina, or shallow aerosol layers in the 346 blind overlap region (i.e., below 225 m a.g.l.), as is the case of Aosta during winter (see Fig. 9). The effects of non 347 continental aerosol types is better illustrated in the supplement S4.1 (Fig. S4.1.2), where the same data are shown together 348 with their associated Ångstrøm Exponents.



Figure 7: Long-term (2016-2022) comparison between the AOD derived by ALICENET (at 1064 nm) and AERONET/SKYNET sun photometers (at 1020 nm) in (a) Aosta, (b) Rome Tor Vergata, and (c) Messina. Colours refer to the data density. The black line is the linear fit. Fit slope and Pearson's correlation coefficients are reported in each panel together with the total number of data pairs (samples). Gray dashed lines delimit deviations of ± 0.01 ± 0.15\*AODsunphotometer from the 1:1 line.

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## 354 3.3.2 Aerosol physical properties

Aerosol physical properties such as particle surface area and volume ( $S_p$  and  $V_p$ ) are also derived based on functional 355 356 relationships linking these to aerosol backscatter and provided in Dionisi et al. (2018). Being of particular interest for AO 357 applications, aerosol mass concentrations ( $M_p$ ) can then be derived from the estimated aerosol volume as  $M_p = \rho_p V_p$ , using 358 an a-priori aerosol density  $\rho_{p}$ . It is worth highlighting that remote sensing aerosol retrievals provide aerosol properties in 359 'unperturbed' atmospheric conditions, i.e., including hygroscopic effects. Conversely, most in-situ instrumentation (as those 360 operating in AOMN in compliance to the EU AO Directive) generally provide dry particulate matter mass values. Therefore, 361 a RH 'adjustment' is necessary when comparing the ALC-based aerosol properties (including mass) to dry in-situ data (e.g. 362 Barnaba et al., 2010). Details on the hygroscopic correction used within ALICENET are reported in the supplement S4.2. In 363 the following, we show both a short- (Fig. 8) and long- (Fig. 9) term comparison between the  $M_p$  retrieved by ALICENET 364 using ALC data collected in Aosta and in-situ reference measurements. 365 In Fig. 8, the  $M_p$  values at 3500 m a.s.l. extracted from ALC aerosol profiles are compared with the aerosol mass 366 concentrations measured by an OPC at the Testa Grigia - Plateau Rosa observatory (western Alps, 35 km-East of Aosta, see 367 Fig. S4.2.1 in supplement S4 for details on site relative locations) in June 2022. This period was selected because in summer 368 secondary hygroscopic particles from the Po Basin are regularly transported to the western Alps, reaching altitudes > 4 km 369 a.g.l. (Diémoz et al., 2019 a.b). In fact, June 2022 registered both medium-range transport of Po Valley pollution and long-370 range transport of desert dust to Plateau Rosa. Figure 8 shows the 30-days temporal evolution of the ALC-based M<sub>2</sub> (bullets) 371 in the ALC vertical bin  $3500 \pm 200$  m a.s.l. over Aosta and the corresponding values from OPC (grey line). The aerosol 372 density used to derive both ALC and OPC aerosol mass concentrations was 1.2 (1.6) g cm<sup>-3</sup> in the presence of non-dust 373 (dust-dominated) aerosol mixtures (Diémoz et al., 2019b). Moreover, assuming desert dust as mainly hydrophobic, the 374 hygroscopic correction as described in supplement S4.2 was only applied to ALC data in non-dust conditions. This 375 discrimination was done using the linear volume depolarisation ratio ( $\delta_v$ ) profiles of a co-located PLC and assuming that 376 aerosol mixtures associated with  $\delta_v < (>)$  15% are dominated by secondary (dust) particles. Overall, Fig. 8 shows that the 377 two mass concentration series exhibit similar time evolution, with good agreement both in low aerosol conditions (e.g. 6-15 378 June 2022), and during transport events increasing the local aerosol load. In the considered period, main transport events 379 were associated with desert-dust intrusions (e.g., 3-5, 18-22, and 27-28 June 2022) and Po Valley pollution advections (e.g., 380 13-14, and 25-26 June 2022). This result is very promising considering that the horizontal distance between the ALC/PLC-381 probed column and the Plateau Rosa station is > 30 km and that the in-situ OPC measurements may also be influenced by 382 local dynamics and surface emissions. 383

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**Figure 8:** Aerosol mass concentrations derived in the month of June 2022 from the Aosta ALC (bullets) and from in situ instrumentation (grey line). In particular, ALC values refer to the vertical layer  $3500 \pm 200$  m a.s.l. and colour code indicates depolarisation values from the co-located PLC ( $\delta_v$ ). In-situ PM<sub>10</sub> concentrations were derived from an OPC at the mountain (3500 m a.s.l.) observatory 'Testa Grigia' (Plateau Rosa, 35-km from Aosta, data courtesy of Stefania Gilardoni, CNR-ISP).

- 394 A longer comparison for the ALICENET aerosol mass product is reported in Fig. 9. It shows the 1-year (2021) record of 395 ALC-derived  $M_p$  at ground level and the corresponding in situ, surface PM<sub>10</sub> concentrations derived by OPC measurements 396 in Aosta downtown, 4 km away from the Aosta ALC (Diémoz et al., 2021). Data are shown in terms of daily median values 397 and corresponding 25-75 percentiles. To convert volume into mass, the aerosol density was set to 1.5 g cm<sup>-3</sup>, while to convert 398 the ALC-derived wet aerosol mass (blue) into dry aerosol mass (purple), the hygroscopic correction (see Eqs. S4.1, S4.2) 399 was applied using surface-level RH measurements and a constant  $\gamma$  exponent of 0.2. Both  $\rho_p$  and  $\gamma$  values are representative 400 for a mean continental aerosol type, i.e., the one expected to dominate in Aosta. As can be observed, the ALICENET 401 retrieved  $M_p$  is able to reproduce the variability of the in-situ measured  $PM_{10}$ , with some underestimations in the winter 402 months. We investigated these underestimations further and found these are mainly attributable to: a) the shallow (i.e., few 403 tens of metres), frequent temperature inversions occurring during winter in the Alpine valleys and capping aerosols in the 404 lowermost levels (e.g., Giovannini et al., 2020), and b) the higher wintertime local emissions in the urban site of Aosta 405 downtown with respect to the semi-rural site where the ALC is operating (Diémoz et al., 2019b).
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Figure 9: One-year (2021) dataset of surface aerosol mass concentrations as derived by the ALICENET ALC inversion and by OPC
measurements in Aosta. Data refer to daily median values (points) and relevant 25-75 percentiles (vertical bars). ALC-based data are both
those derived from the ALICENET retrieval (wet) and corresponding ones further corrected to dry values (see the supplement S4).

#### 412 **3.3.3 Estimated uncertainty of aerosol properties retrievals**

- 413 The previous sections describe the ALICENET efforts to exploit the great potential of ALC in providing quantitative 414 aerosol-related geophysical parameters, and demonstrate the good performances of the current algorithms. Nonetheless, due to several factors also discussed above, the expected uncertainties associated with the output products range from 20% for 415 416 the attenuated backscatter (product L2 in Fig.1) to 50% for the aerosol mass (L3 in Fig. 1). The main factors are listed 417 hereafter. 418 1) the instrumental noise of the signal. This factor depends on the instrument status and mainly impacts the retrievals in the 419 middle-upper troposphere. 420 2) the overlap correction applied to the signal. As discussed, this factor is critical in the lowermost levels and accurate 421 instrument-specific, overlap-correction models are necessary to derive quantitative information in the first 800 m. Accuracy 422 of the retrievals in this vertical region depends on the statistical and physical representativeness of the ensemble of overlap 423 functions from which the overlap model is derived (supplement S2). 424 3) the variability of the instrument calibration coefficient. This third factor (see Sect. 3.2), directly impacts the accuracy 425 of  $\beta_{att}$ . For example, it is found by error propagation that changes of 30% in the instrument calibration coefficient (which are
- 426 quite usual in some ALICENET and E-PROFILE stations) translates into a variability in  $\beta_{att}$  up to 20%.

427 4) the accuracy of the functional relationships used in ALICENET to link the aerosol backscatter to the o	her aerosol
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428 properties, impacting the estimation of  $\alpha_p$ ,  $S_p$ ,  $V_p$ ,  $M_p$  and, to a lesser extent,  $\beta_p$ . This factor strongly depends on the actual

429 aerosol conditions: the functional relationships can give a good estimate of the aerosol properties in presence of continental

430 aerosols, while in presence of non-continental particles they are less accurate (a relative error of 30-40% was derived by

431 Dionisi et al., 2018). As mentioned, extension of the ALICENET approach to include other aerosol types is foreseen for the

432 next future. In particular, exploitation of the PLC depolarisation profiles for aerosol-typing will drive the selection of

- 433 aerosol-type specific functional relationships (e.g. Gobbi et al., 2002).
- 434 Concerning the retrieval of aerosol mass concentrations, the assumed particle densities are a major source of uncertainty, and
- 435 the accuracy of the retrieval depends on the possibility to better constrain the aerosol density profiles, e.g., through ancillary
- 436 data, including depolarisation information.
- 437 Overall, the above factors result in instrument-, time- and range-dependent uncertainties of the ALC-based aerosol optical
- 438 and physical properties. The expected uncertainty with an optimal SNR up to at least 7 km a.g.l., an overlap error < 10% in
- 439 the lowermost levels, and in presence of continental aerosol types is of 20% for  $\beta_{att}$ , 30-40% for AOD, reaching 50% for
- 440 aerosol mass.
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## 442 **3.4 The ALICENET automatic Aerosol LAyer DetectIoN algorithm (ALADIN)**

443 As already mentioned, a main advantage of ALCs is their ability to operate continuously, which allows detecting and

444 tracking the variability of the aerosol vertical stratifications at multiple timescales using aerosol as passive tracers. This

445 information can be beneficial for several sectors, among which AQ and meteorology (e.g., Moreira et al., 2019; Ravnik et

446 al., 2024; Körmöndi et al., 2024), aviation (e.g., Osborne et al., 2019; Salgueiro et al., 2023), atmospheric research (e.g.,

- 447 Jozef et al., 2024).
- 448 Commonly identified atmospheric stratifications based on ALC data analysis include the Atmospheric Boundary Layer and

449 the Mixed Layer (ABL and ML, respectively, e.g., Poltera et al., 2017; Kotthaus et al., 2020; Caicedo et al., 2020), and

450 lofted aerosol layers in the free troposphere (e.g., Adam et al., 2020). The ABL is a thermodynamic layer connected to the

451 Earth's surface and capped by a temperature inversion, while the ML is an ABL sublayer mixed by turbulent fluxes (Stull,

452 **1988; Kotthaus et al., 2023).** 

However, it should be noticed that aerosols are 'delayed' tracers of atmospheric dispersion processes and may not always consistently represent the thermodynamic state of the atmosphere (Haeffelin et al., 2012). The tracking of thermodynamic layers through aerosol lidars can be complicated by superimposing phenomena such as large-to-medium scale advections, natural and anthropogenic emissions, particle physico-chemical transformations. These processes may remove or transport particles in specific atmospheric ranges (e.g., Collaud Coen et al., 2018; Diémoz et al., 2019a), modulate the daily cycle of aerosol profiles (e.g., Diémoz et al., 2021), form aerosol layers within and above the ABL (e.g., Curci et al., 2015; Sandrini et al., 2015), thus decoupling the aerosol-related and thermodynamic stratifications. This decoupling is expected to be further

- 460 enhanced over complex terrain (e.g., Serafin et al., 2018) and/or over regions affected by multiple natural and anthropogenic
  461 sources, as is the case of the Italian territory.
  - 462 For all these reasons, the choice for aerosol layers detection and naming in ALICENET was to keep a clear link to the
  - 463 aerosol field allowing its identification, avoiding a terminology traditionally based on thermodynamics. In particular, we
  - 464 develop a novel Aerosol LAyer DetectIoN (ALADIN) tool to automatically derive aerosol layering information from
  - 465 ALCs/PLCs across the network, this targeting the following aerosol layers:
  - 466 1. the Continuous Aerosol Layer (CAL): it is the layer extending from the ground level and characterised by the 467 continuous presence of aerosols;
  - the Mixed Aerosol Layer (MAL): it is a CAL sublayer within which aerosols are mixed by surface-driven turbulent
     fluxes;
  - 470 3. Elevated Aerosol Layers (EALs): they are lofted aerosol layers which lie above the MAL, and either within or 471 above the CAL.
  - 472 Within ALADIN, each layer type (CAL, MAL, and EALs) is detected from ALC/PLC L2 data using a specific methodology.
  - 473 The CAL is determined by comparing the aerosol and the molecular  $\beta_{att}$  profiles. The identification of the MAL is based on
  - 474 Dynamic Time Warping (DTW, Giorgino et al., 2009) and variance analyses of the ALC profiles. The detection of EALs is
- 475 performed with Continuous Wavelet Transform (CWT, Du et al., 2006) and iterative techniques. Full details on the
- 476 ALADIN procedures, as well as a schematic description of the ALADIN processing flow are reported in supplement S5.
- 477 Figure 10 shows the 'layering mask' corresponding to the same ALC data shown in Fig. 6. It includes the ALADIN output
- discriminating the CAL, MAL, and EALs, plus the aerosol-free (i.e., molecular, MOL), and cloud-screened (CLOUD)
  regions as inferred from the overall ALICENET processing. In this episode, the EALs above 3 km a.g.l. are mostly due to
- 480 minor (July 7-8 and 10-11) and major (July 9) Saharan dust intrusions, while the ones between 1-3 km a.g.l. to fire plumes
- (e.g., July 11; Andrés Hernandez et al., 2022) and/or to aerosol formation and growth within the residual layer during
  nighttime (e.g., July 5-6).
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Figure 10: Aerosol layering mask derived from the ALADIN processing on the CHM15k operating in Rome - Tor Vergata in the same period presented in Fig. 6. The mask discriminates the following layers: the continuous aerosol layer (CAL), the mixed aerosol layer (MAL) and elevated aerosol layers (EALs). Aerosol-free (i.e., molecular, MOL) and cloud-screened (CLOUD) regions as identified in the overall ALICENET processing are also shown.

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500 Further discrimination of aerosol layers in terms of aerosol type could be derived exploiting PLC  $\delta_{\rm v}$  profiles. In fact, the 501 inclusion of the PLC depolarisation information within the ALICENET processing is in progress, this representing a first 502 step to automate the aerosol typing capacity within the network (thus complementing the aerosol layer typing capacity from 503 more complex lidar systems, e.g., Nicolae et al., 2018; Córdoba-Jabonero et al., 2018). 504 Routine application of the automated ALADIN tool on a daily basis also allows to get statistics of vertical aerosol stratifications in the atmosphere. An example of this long-term application is presented in Figure 11, which shows the 505 506 monthly- and daily-resolved cycle of MAL and CAL heights over Rome-Tor Vergata derived from the 2016-2022 ALC 507 dataset (continuous lines are median values while shaded areas represent 25th-75th percentiles). Figure 11 clearly shows the 508 marked yearly cycle of the CAL height (minimum in winter and maximum in summer), due to the increased convection and 509 photochemistry in the warmest months (e.g. Barnaba et al., 2010). As expected, all over the year the MAL shows a marked 510 daily cycle, with maximum heights in summer (about 2 km thick in July-August) doubling those in winter (about 1 km in 511 December-January). A similar statistics of turbulent kinetic energy (TKE) from a co-located ultrasonic anemometer

- 512 (magenta lines) is also reported as a proxy for convection, which is the main driving factor of the MAL temporal evolution.
- 513 Note that in this Figure the time axis is reported as Central European Time (CET) to better highlight the diurnal variability of
- 514 the addressed quantities.
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Figure 11: Monthly and daily resolved statistics (median, and 25-75 percentiles as shaded dashed areas) of the MAL and CAL heights (left y-axis) derived from the ALADIN tool application over the multi-annual (2016-2022) dataset of the CHM15k in Rome Tor Vergata. Similar statistics of the turbulent kinetic energy (TKE) derived from a co-located ultrasonic anemometer (violet) are also plotted (right yaxis) as a proxy of convection intensity and timing.

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523 A follow up work presenting a more detailed multi-annual analysis of the ALC-based aerosol properties and layering in 524 selected ALICENET sites from North to South Italy, in synergy with in-situ aerosol measurements and model (ERA5, 525 CAMS) products, is currently in progress (Bellini et al., 2024, in preparation).

## 526 4. Potential of 4D near-real time aerosol monitoring

527 A main advantage of lidar-ceilometers networks is their continuous, near real-time monitoring capability. In fact, 528 ALICENET has been already exploited in past events to follow the evolution and characterise specific aerosol transport 529 features and/or to quantify the impact of aerosol dynamics on local aerosol concentrations, mostly in synergy with other 530 tools and measuring techniques as in-situ aerosol observations, ground-based passive remote sensors, satellites or models 531 (Gobbi et al., 2019; Diémoz et al., 2019a,b; Di Bernardino et al., 2021; Rizza et al., 2017, 2022; Tositti et al., 2022; Andres 532 Hernandez et al., 2022). This section describes, through some recently recorded showcases, the potential of this near real 533 time 4-dimensional ALICENET monitoring at the national scale, particularly useful for nowcasting, warnings and alerts in 534 case of noteworthy events.

## 535 4.1 Po Valley local dust front (14 April 2020)

In a previous study (Diemoz et al., 2019a,b), the operational use of ALICENET provided observation-based evidence of the export of pollutants from the Northern Italy Po Valley to surrounding areas. The phenomenon, previously observed by lidar profiling performed at the EC-JRC in Ispra (about 60 km northwest of Milan, Barnaba et al., 2010), was further analysed

and quantified thanks to the ALICENET combination of sites (Milan and Aosta, i.e., within and at the border of the Po Valley). That study demonstrated that such pollution-rich advections markedly affect PM-related AQ even in the 'pristine' mountain environments mainly transporting hygroscopic particles of secondary origin. However, transport of particles of primary origin (particularly from soil-related sources) across the Po Valley has been also observed, particularly during dry periods. Figure 12 shows an example of such events (14 April 2020), largely impacting regional AQ and visibility.

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**Figure 12:** (a) Total attenuated backscatter profiles at Aosta and Milan-Bicocca sites on 14/04/2020; (b) central Milan webcam (Source: Arzaga meteorological observatory, https://www.osservatorioarzaga.it/) showing the rapid decrease of visibility on 14/04/2020 (from top to bottom: 16:08, 16:15, 16:20, 16:25 UTC), (c) Po Valley satellite true colour image (14/04/2020 18:10 UTC; Credits: EUMETSAT) with indication of the regional dust front (orange arrow), and (d) 10 m wind speed and direction simulated by WRF over North Italy (14/04/2020 17:00 UTC, data courtesy of Stefano Federico CNR-ISAC) illustrating the extension of the gust and wind fronts. The arrival of the dust front in Milan at 16:20 UTC and in Aosta at 20:40 UTC is clearly visible from ALC profiles in panel a.

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This episode was due to an extended (about 100 km) gust front originating from the cold and intense Bora winds from East, as well as to anomalous dry conditions affecting Europe in April 2020. Resuspended, soil-originated particles from the

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cultivated fields were transported across the whole Po Valley as also visible from space (Fig. 12c). ALC profiles (Fig. 12a) well captures the timing of the plume's arrival in Milan (as also seen from central Milan webcams, Fig. 12b) and show the vertical extent of the particle-rich layer associated with the episode. As also revealed by satellite measurements (Fig. 12c) and model simulations (Fig. 12d), after impacting the Milan area, the plume continued to travel westward and was detected by the ALC in Aosta 4 hours later, indicating a wind speed > 12 m/s.

#### 561 **4.2** Advection of Saharan dust and Canadian fire plumes over Italy (19-28 June 2023)

562 The Mediterranean area is frequently affected by the transport of desert dust from North Africa and the Middle East (e.g., 563 Barnaba and Gobbi, 2004; Querol et al., 2009; Basart et al., 2012a; Greilinger et al., 2019; Gama et al., 2020). In Italy, these 564 events are estimated to reach the ground on 10% (Northern regions) to over 30% (Southern regions) of the days in a year, and to impact on surface daily-mean PM<sub>10</sub> concentrations with 10-15 µg/m<sup>3</sup> (Barnaba et al, 2022). Transport of fire plumes 565 566 from global-to-medium distances is also an important contributor to aerosol loads in Europe. A significant contribution is given by forest fires regularly developing during boreal summers in Canada (e.g., Ceamanos et al., 2023; Shang et al., 2024), 567 568 and a major contribution from agricultural fires in Eastern Europe and Russia has also been detected over the continent, 569 particularly in spring and summer (Barnaba et al., 2011). Summer 2023 was particularly impacted by multiple episodes of 570 severe wildfires in central Canada. Almost 480 megatonnes of carbon were emitted, resulting in a major impact on AO 571 across Canada and the Northern US. The plumes have also been observed to be regularly transported towards Europe 572 (https://atmosphere.copernicus.eu/copernicus-canada-produced-23-global-wildfire-carbon-emissions-2023, last access: 6-3-573 2024). Figure 13 shows a composite of measurements collected at multiple ALICENET sites across the country during a 10-574 days period (19-28 June 2023) affected by both desert-dust (time-altitude windows identified by orange boxes) and forest-575 fire plumes (time-altitude windows identified by magenta boxes). More specifically, this period was characterised by the 576 intrusion of Saharan dust to Southern to Northern Italy (19-24 June 2023), followed by the transport of Canadian fire plumes over Central and Northern Italy (27-28 June 2023). The ALC profiles ( $\beta_{att}$  and  $\delta_{v}$ ) at the 7 selected ALICENET sites (central 577 578 panel in Fig. 13) allow to follow the spatio-temporal evolution of the different aerosol layers and identify the relevant aerosol 579 type. The Saharan dust layers were firstly observed over South-West Italy (Capo Granitola, June 19 in the morning), then 580 moving westward to Messina and Catania (June 19, afternoon), and northward to Turin, Aosta, Milano, Mt. Cimone, where 581 the dust plume is detected in the evening. All over Italy, the dust plume affects atmospheric layers up to 7 km altitude, 582 reaching down to the surface on June 20. In fact, the PLC systems clearly indicate the presence of irregularly-shaped mineral particles aloft (depolarisation values  $\delta_v > 30\%$ ) and mixing with local (mainly spherical) particles, with  $\delta_v \sim 10-20\%$  when 583 584 reaching the lowermost levels.

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589 **Figure 13:** Vertical profiles of total attenuated backscatter  $\beta_{\text{att}}$  (for both ALCs & PLCs) and volume depolarisation  $\delta_v$  (for PLCs) as 590 recorded at selected, North-to-South ALICENET sites in the period 19-28 June 2023, affected by Saharan dust and Canadian fire plumes 591 (orange and magenta boxes, respectively).

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593 The Canadian fire plumes were firstly observed by ALICENET systems operating in North-Western Italy (Aosta, Turin) on 594 27 June 2023 in the range 2-7 km a.s.l. Then travelled through the whole Po Valley, being clearly observed in Milan and Mt. 595 Cimone. Being mainly composed of processed particles, these long-range transported fire plumes do not show increased 596 depolarisation, and appear as thinner aerosol layers with respect to the ones typically associated with dust layers. These 597 vertically resolved measurements well complement the information that can be gathered from satellites. For instance, a 598 comparison between ALICENET data and MSG and Metop retrievals was conducted with respect to the dust event (e.g., 599 https://vuser.eumetsat.int/resources/case-studies/dust-transport-from-the-sahara-to-the-mediterranean, last access: 6-3-2024). 600 At the same time, vertical aerosol profiling also provides an observational verification of the picture that can be obtained by 601 modelling tools. In this respect, Fig. 14 shows the CAMS EU forecast maps (Ensemble model) for two dates within the 602 temporal window addressed, i.e.: 22/06/2023 (dust intrusion, left panels) and 27/06/2023 (Canadian fires, bottom panels), at

two altitude levels (100 and 3000 m a.g.l., top and bottom panels respectively). The horizontal evolution of the aerosol advections qualitatively agrees with the ALICENET observations. It is more difficult to correctly model the aerosol vertical distribution, due to both their coarse vertical resolution and simplified parameterizations of the aerosol-related atmospheric processes (e.g., Koffi et al., 2016). Indeed, remote sensing observations by ALC/PLC represent an added value for both AQ monitoring and modelling. In fact, specific efforts are currently ongoing in the assimilation of ceilometer information into the IFS (Integrated Forecasting System)/CAMS (e.g., the H2020 CAMs AERosol Advancement (CAMAERA) Project, https://camaera-project.eu/, last access: 25-07-2024).



Figure 14: CAMS EU forecast of the total PM10 and PM10-dust component concentrations during the desert dust (22/06/2023 00:00 UTC
- left panels) and the Canadian fires (27/06/2023 21:00 UTC - right panels) events of Figure 13, top (bottom) panels referring to 100 m
(3000 m) altitude.

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## 615 **4.3 Aerosol particles from the Mt. Etna eruption (13-14 August 2023)**

A recent showcase from the Etna volcano eruption is reported in Figure 15 to highlight the important information that ALC/PLC observations can provide in volcanic areas to complement in situ, satellite-based and modelling data (e.g.,

618 Corradini et al., 2018, Scollo et al., 2019, Bedoya-Velásquez et al., 2022). During the night between 13 and 14 August 2023, 619 this Europe's most active volcano erupted, its Southeast Crater emitting a volcanic cloud that the PLC in Nicolosi detected to 620 reach up to 5 km at 21 UTC (Fig 15a). On August 13, at 20:41 UTC, a Volcano Observatory Notice for Aviation (VONA) 621 was issued by INGV (https://www.ct.ingv.it/Dati/informative/vona/VONA Etna 202308132041Z 2023005708E01.pdf, last access: 06-03-2024) with a 'red alert' for aviation. VONA are short, plain-English messages aimed at dispatchers, pilots, and 622 623 air-traffic controllers to inform them of volcanic unrest and eruptive activity that could produce ash-cloud hazards. In fact, 624 flights serving Catania were halted. The most intense phase of the eruption occurred between 01:40-02:30 UTC, when PLC 625 depolarisation reached values > 40% indicating a predominance of irregular ash particles. The ash plume was then observed 626 to rapidly reach the ground, while moving southward in the Mediterranean Sea (Fig 15b). In fact, less than 5 hours after the 627 beginning of the eruption the plume was detectable east of Malta. In agreement with the ALC record, the VONA issued by INGV at 05:54 UTC indicates that no ash plumes were produced and that the volcanic ash was confined in the summit areas 628 629 of the volcano, this corresponding to an orange Aviation colour code 630 (https://www.ct.ingv.it/Dati/informative/vona/VONA Etna 202308140554Z 2023005808F01.pdf, last access: 06-03-2024).

59 60



Figure 15: (a) Total attenuated backscatter,  $\beta_{att}$ , plus volume depolarisation,  $\delta_v$ , profiles observed at the ALICENET Etna Nicolosi site on 13-14/08/2023; (b) METEOSAT Natural Colour Enhanced RGB (SEVIRI) image referring to 14/08/2023, 05:15 UTC (Credits: EUMETSAT).

## 637 **5 Conclusions and and future perspectives**

In this work we present ALICENET, the Italian network of automated lidar-ceilometers (ALCs) operating from North to South across the peninsula. It is a cooperative network set up by CNR-ISAC in 2015, and currently running with active contributions from several regional EPAs, Universities, Research Centres and private companies. The network contributes to fill an Italian observational gap at the EU level, where most Member States generally run extended ALC networks managed by national meteorological agencies (e.g. the German weather service, DWD, running over 100 instruments, https://www.dwd.de/EN/research/observing\_atmosphere/composition\_atmosphere/aerosol/cont\_nav/aerosolprofiles.html,

644 last access 25-07-2024). Since its set up, the ALICENET network kept expanding (Table 1), and currently covers very 645 different environments (urban, coastal, mountainous and volcanic areas), thus providing information in a large spectrum of 646 atmospheric conditions and aerosol regimes. ALICENET promoted a standardisation of instruments and an homogeneous 647 data processing specifically developed within the network. It mainly runs single-channel ALCs (CHM15k systems by Ott 648 Hydromet) but is progressively introducing polarisation-sensitive systems (PLCs) recently commercialised by Vaisala 649 (CL61) to further exploit the ability of these systems to discriminate among aerosol types. Since the beginning of the 650 ALICENET activities, particular care has been devoted to data retrievals and exploitation, this also taking advantage of technical/scientific exchanges within European initiatives, such as the EC Cost Actions TOPROF (2013-2016) and PROBE 651 652 (2019-2024), the ongoing EUMETNET program E-PROFILE (2020-2028) and the EC H2020 Project RI-URBANS (2021-653 2025). In this context, ALICENET developed a specific, centralised and automated data processing chain with associated 654 data quality control (OC) procedures, as presented in detail in this work. The data processing steps were either refined from 655 previously published work (e.g. Hervo et al., 2016, Dionisi et al., 2018), or are completely new, as the automatic aerosol 656 layers detection algorithm (ALADIN). Overall, the processing chain includes signal correction and calibration procedures 657 (Sects. 3.1, 3.2), the aerosol properties inversion (Sect. 3.3), and the identification of vertical stratifications (Mixed, 658 Continuous and Elevated Aerosol Lavers, MAL, CAL and EALs, respectively, Sect. 3.4). Output products with different 659 levels of complexity and associated uncertainties are thus provided (Fig. 2). These range from more basic L1 quantities (as 660 the Range-Corrected Signal, RCS, and, where applicable, depolarisation,  $\delta_{\rm v}$ ), through the L2 total attenuated backscatter  $\beta_{\rm att}$ 661 to the L3 aerosol optical ( $\beta_p$ ,  $\alpha_p$  and thus AOD) and physical ( $S_p$ ,  $V_p$ , and  $M_p$ ) properties plus vertical layering.

Level 1 and Level2 products are provided in near real time on a dedicated website (https://www.alice-net.eu/, last access: 25-07-2024), while L3 products are obtained offline and are currently only available upon request. Examples of product types are reported in Sect. 3 and 4. For L3 products, this work also includes direct comparisons with relevant, independent data (in-situ or remote sensing, depending on the variable addressed), showing that the ALICENET data processing is able to provide robust and quantitative aerosol information, within the discussed limits of the data accuracy (Sect. 3.3.3). In fact, long-term comparisons of aerosol mass retrievals with surface  $PM_{10}$  data show mean discrepancies of 35%, while AOD comparisons to thousands of relevant data points from co-located sun photometers show correlation coefficients > 0.8 and fit

669 slopes ranging between 0.8-1.0, depending on the site location.

670 Efforts to evaluate the ALICENET retrieval performances are constantly performed as well as comparisons to different 671 inversion approaches and tools. For example, a preliminary algorithm intercomparison exercise was recently performed 672 within PROBE to evaluate differences in the outcomes produced by different national networks in the EU (namely:

- 673 ALICENET Italy, MetOffice UK, V-PROFILE Norway, DWD Germany; Osborne et al., 2024). An additional analysis
- 674 of the ALICENET L3 products is currently in progress based on multi-annual datasets of selected ALICENET systems
- 675 located across Italy and relevant comparisons to independent data and models (Bellini et al., 2024, in preparation).
- 676 Next steps foreseen within the network are: a) a better characterisation of the instruments artefacts and calibration, b) the
- 677 extension of the ALICENET ALC retrieval methodology to different aerosol types, c) the development of a full retrieval for
- 63 64

- PLCs (CL61), further exploiting the depolarisation information to identify aerosol types. Since the CL61 operates at a different wavelength with respect to CHM15k, the evaluation of water vapour absorption corrections (e.g., Wiegner and Gasteiger, 2015), and the definition of new, wavelength specific functional relationships (e.g. Dionisi et al., 2018) to be used within the data inversion process are also required and will be explored. The feasibility of a regular dissemination of
- 682 ALICENET L3 products via the network website in addition to the near-real time L1 and L2 ones is also under evaluation.
- 683 Overall, ALICENET represents a valuable resource to complement the aerosol observational capabilities in Italy with the

684 unique capacity of continuous 4D monitoring. The maturity of both instrumental technologies and data processing tools as

- 685 the ones described here suggest that ALC/PLCs could fruitfully contribute to aerosol measurements within European
- 686 Research Infrastructures (e.g. ACTRIS) and/or air quality monitoring networks (AQMNs).
- At the national level, ALICENET also intends to bridge a gap between the research-oriented and the operational use of
  active aerosol remote sensing in several sectors, among which: a) air quality (AQ), b) radiative budget/solar energy, c)
  aviation safety, thus representing a good example of earth observation science applications for society. Its outputs were
- 690 already proven to be also useful in validation of models and satellite products.
- 691 Of particular interest for the AO sector are the abilities of the ALC/PLC-based ALICENET data to: i) automatically identify 692 medium-to-long range aerosol advections and estimate the relevant contribution to surface PM10 concentrations, and ii) provide continuous information on particulate matter layering, including the Mixing Aerosol Layer (MAL), i.e. on the 693 atmospheric volume in which locally emitted particles are diluted (e.g., Kotthaus et al., 2023), and the Elevated Aerosol 694 695 Layers (EALs) reaching the surface. The effectiveness of using these ALC/PLC abilities in support of standard AOMNs is 696 being currently explored within the ongoing EC H2020 Project RI-URBANS, aimed at developing an air quality monitoring 697 system that complements those currently available. In this framework, tests of upscaling the ALICENET tools to other urban 698 sites in the EU are in progress (e.g., Barnaba et al., 2024). Concerning the other applications mentioned above, the 699 continuous ALC-based information on the aerosol properties vertical distribution and layering is useful to better estimate the 700 relevant radiative effects (beneficial for example within an operational short-term solar forecasting system based on a 701 multisensor approach, e.g. Papachristopoulou et al., 2024), for validation of/assimilation in models (e.g. Chan et al., 2018; 702 Valmassoi et al., 2023), or for the provision of near-real time alerts for aviation safety during specific extreme events such as 703 desert dust storms and volcanic eruptions (e.g., Papagiannopoulos et al., 2020). Continuous aerosol monitoring capabilities 704 of ALC/PLC systems and availability of relevant long-term records is also expected to be particularly important in the 705 verification of satellite aerosol products including vertical lavering (e.g., Janicke et al., 2023), considering that aerosol 706 vertical profiles and planetary boundary layer are recognised as priority targeted observable for space-based Earth 707 observation programs (e.g. NASEM, 2018) and that the joint ESA-JAXA mission EarthCare with a lidar instrument onboard was recently successfully launched (e.g., van Zadelhoff et al., 2023). 708
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- 710

711	List of acronyms
712	ABL: Atmospheric Boundary Layer
713	ACTRIS: Aerosol, Clouds, and Trace Gases Research Infrastructure
714	AERONET: Aerosol Robotic Network
715	ALADIN: Aerosol LAyer DetectIoN algorithm
716	ALC: Automated Lidar-Ceilometer
717	ALICENET: Automated LIdar-CEilometer NETwork
718	AQ: Air Quality
719	AQMN: Air Quality Monitoring Network
720	AOD: Aerosol Optical Depth
721	ARS: Aerosol Remote Sensing
722	BG test: Breusch-Godfrey test
723	CAL: Continuous Aerosol Layer
724	CAMAERA: CAMs AERosol Advancement
725	CAMS: Copernicus Atmosphere Monitoring Service
726	CHM15k: Lufft automated lidar-ceilometer instrument
727	CL61: Vaisala polarisation sensitive lidar-ceilometer instrument
728	CNR-ISAC: National Research Council - Institute of Atmospheric Sciences and Climate
729	CWT: Continuous Wavelet Transform
730	DTW: Dynamic Time Warping
731	DWD: German Weather Service
732	EAL: Elevated Aerosol Layer
733	EARLINET: Aerosol Research Lidar Network
734	EarthCARE: Cloud, Aerosol and Radiation Explorer
735	EC: European Community
736	ECMWF: European Centre for Medium-Range Weather Forecasts
737	EPA: Environmental Protection Agency

- 738 ESA: European Space Agency
- 739 ESFRI: European Strategy Forum on Research Infrastructures
- 740 E-PROFILE: EUMETNET program coordinating the measurements of wind, aerosol and cloud profiles from radars and
- 741 <mark>lidars</mark>
- 742 ERA5: fifth generation ECMWF reanalysis for the global climate and weather
- 743 ESFRI: European Strategy Forum on Research Infrastructures

744	EUMETNET: European Meteorological Services Network
745	IFS: Integrated Forecasting System)
746	INGV: Istituto Nazionale di Geofisica e Vulcanologia
747	JAXA: Japan Aerospace Exploration Agency
748	LR: Lidar Ratio
749	MAL: Mixed Aerosol Layer
750	ML: Mixed Layer
751	MPLnet: Micro-Pulse Lidar Network
752	NASA: National Aeronautics and Space Administration
753	NASA-CALIPSO: NASA-CNES CALIOP sensor onboard CALIPSO
754	OPC: Optical Particle Counter
755	PLC: Polarisation-sensitive automated Lidar-Ceilometer
756	PM: Particulate Matter
757	PROBE: PROfiling the atmospheric Boundary layer at European scale
758	QA: Quality Assurance
759	QC: Quality Control
760	QC.CAL: Quality Control applied within the absolute calibration procedure
761	QC.EAL: Quality Control applied within the ALADIN detection of elevated aerosol layers
762	QC.OVL: Quality Control applied within the overlap correction procedure
763	RI-URBANS: EC H2020 project aimed at developing advanced service tools for air quality monitoring networks
764	RH: Relative Humidity
765	SNR: signal-to-noise ratio
766	SKYNET: ground-based radiation observation network dedicated to aerosol-cloud-solar radiation interaction researches
767	TOPROF: Towards Operational ground based PROFiling with ceilometers, doppler lidars and microwave radiometers
768	VONA: Volcano Observatory Notice for Aviation
769	WHO: World Health Organization
770	<b>Data availability:</b> The presented detects will be made freely accessible and linked to a dei, should the revision present

- Data availability: The presented datasets will be made freely accessible and linked to a doi, should the revision process lead
   to a positive outcome.
- 772 Author Contribution: Conceptualization, Data curation, Investigation: AnB, FB, HD; Formal analysis and Software: AnB;
- 773 Visualization: AnB, FB, HD; ALC instruments and database management: LDL, AlB, FP, HD, GPG; Funding acquisition
- and Supervision: FB; Writing original draft preparation: AnB, FB, HD; Writing review & editing: AnB, FB, HD, AlB,
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