HARMONIA
International network for harmonization of atmospheric aerosol retrievals from ground based photometers

HARMONIA CA21119

Deliverable 2.1

Report on synergistic approaches towards better quality products

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1. Introduction

According to the sixth Assessment Report (AR6) of Intergovernmental Panel on Climate Change (IPCC, 2023), the observed human- caused earth system warming is dominated by the increasing atmospheric concentration of greenhouse gases, such as carbon dioxide (CO2) and methane (CH4), but it's partly masked by the aerosol cooling effect, whose quantification still represents one of the major challenges faced by the scientific community. Additionally, it has been proven that fine aerosol and particulate matter (PM) has serious implications on human health (Arfin et al., 2023) and can be assumed as a proxy indicator for air pollution. It is therefore essential to provide an accurate aerosol characterization to the scientific community, mostly focusing on the optical properties affecting absorption and scattering of the sunlight (Li et al., 2022).

Aerosol Optical Depth (AOD), Single Scattering Albedo (SSA), refractive index and size distributions are the most important columnar optical properties typically used to describe the aerosol absorption and scattering capabilities and the size properties. They can be retrieved from sky radiation measurements performed by international photometers networks (AERONET, Holben et al., 1998; SKYNET, Nakajima et al., 2020; GAW-PFR, Kazadzis et al.,2018a) and radiative transfer modelling (Dubovik and King, 2000, Nakajima et al., 2020). The main target of HARMONIA is to work on their homogenization and harmonization.

To study the possible improvements of aerosol measurements using solar, lunar and star photometry, a census of existing field campaigns or experiments, was carried out by the Working Groups 2 and 1 (WG 2-1). They collected information about day-time and night-time measurements and aerosol retrievals, mostly performed in Europe, including established networks, low-cost sensors and other independent instruments databases. Information about field campaigns and long-term measurement was gathered separately to track possible differences in instrument's performances and measurements quality. In the former case, the personnel are constantly present ensuring the care of the equipment, while in the latter, instruments are typically remotely controlled and mostly left to their own. WG2 focused its activity on the establishment of a cooperative networking towards a better quality of products and measurements and to understand the possible synergistic approaches. List of permanent stations having a symphony of diurnal and nocturnal instruments suitable for performing harmonic intercomparison studies was also provided.

Results on these preliminary overviews are summarized in the following sections.

2. Current status of aerosol observations

The current status of aerosol observations was analyzed by collecting information about the campaigns and long-term measurements where WG members attended or have contact for data access. The campaigns must have involved at least one photometer (sun, lunar or star) or a Skycamera co-located. The availability of ancillary measurements, as laboratory analysis or models application, was checked. A recognition of permanent stations having diurnal and nocturnal data, with at least one photometer, was also performed.

2.1 Campaigns and long-term measurements

To draw a picture of the current status, the following information was asked:

Name
Campaign/Long-term
Purpose
Start and end period
Location
Instruments involved
Range of wavelengths
Day/Night/Continuous
How are instruments calibrated?
Where raw data are available?
Are data processed/analyzed?
Where processed data are available?
List of processed products
List of products to be processed
Are data cloud-screened? (if necessary)
Are results published? Where?
Ancillary measurements (use of laboratory and/or models analysis)
Contacts

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24 field campaigns were recorded in the census, ranging from 2014 up to September 2023, but many others are supposed to be collected during the next years of HARMONIA. Most of the campaigns have been performed in the European continent, 1 in South America and 3 in Africa (Figure 2.1). The duration (Figure 2.2) varies from 9 to 35 days, but longer campaigns as QUATRAM 2 and 3 (87 and 152 days, respectively), Montevideo 2 (67 days), Biosure (503 days) and Cycare (521 days) were listed. An increase in the numbers of field campaigns can be observed from 2021. The instruments involved (shown in Figure 2.3) can be grouped in: i) photometers or similar (PREDE, PFR, CIMEL, MIDDLETON, MFRSR, STELLAR, Sky CAMERAS, MICROTOPS, SP1) providing AOD and other aerosol properties as SSA, refractive index and size distribution; ii) lidars or similar (LIDAR, CELIOMETER, CLIDAR) providing aerosol vertical profiles; iii) wind, moisture and temperature profilers (WIND LIDAR, MWR, Radiosounding); iv) on board measurements (unmanned aerial vehicle/UAV, AIRCRAFTS); v) in situ measurements (aerosol samplers, and meteo stations); vi) spectrometers (BREWERS, other spectrometers as Pandora, and PSR) proving gas concentrations and AOD, vii) radiation meters (PYRANOMETERS, PYRHELIOMETERS). The most commonly deployed photometer is the CIMEL, as expected since they are part of the AERONET network. In many campaigns lidars are co-located and, in a smaller amount, also microwave radiometers. 13 long-term measurements sites were recorded, some of them became long-term by taking advantage of the instruments previously involved in field campaigns. The start date for each site is shown in Figure 2.4. In comparison to the campaigns list, the involved equipment (Figure 2.5) includes more observations from Sky cameras, Microptops, AOD from Brewer (which is the longest series), in situ aerosol sampling and meteorological observations.

Figure 2.1. Deployment of the campaigns (yellow markers) and long-term measurements (red markers). The yellow line in Figure a) refers to a shipboard campaign from Vigo (Spain) to Abu Dhabi (UAE)

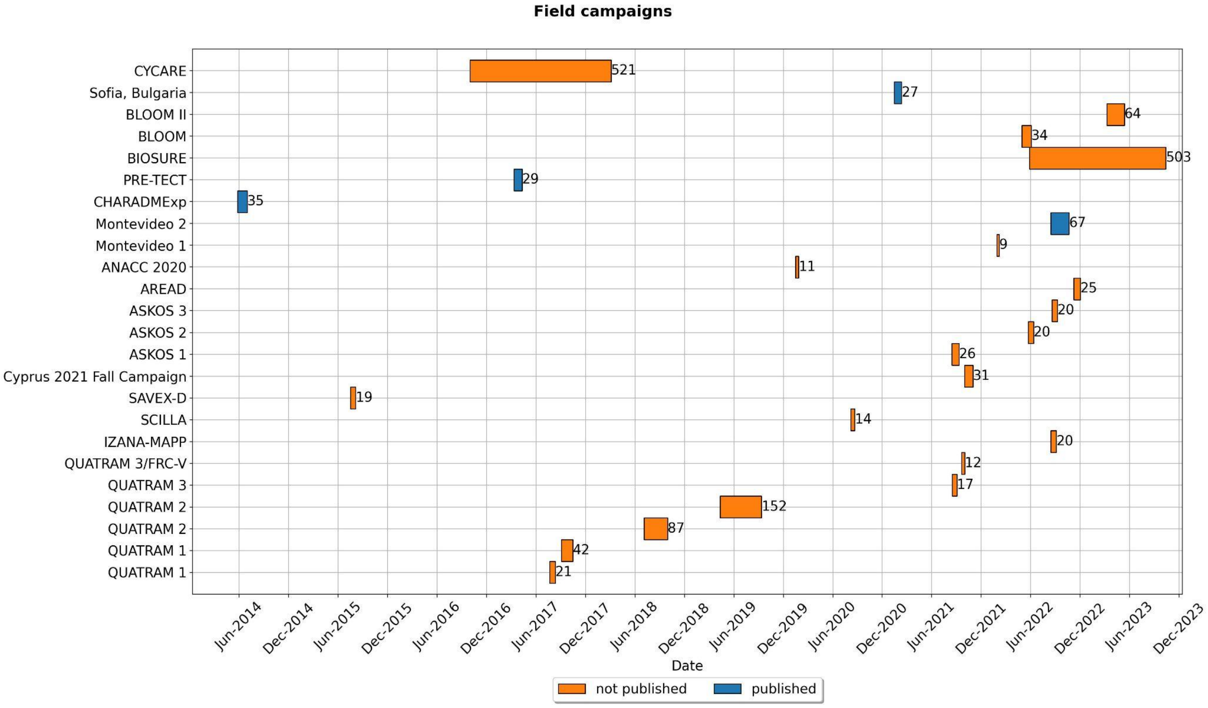

Figure 2.2: Temporal duration of the campaigns. The numbers indicate their duration (days), the colors refer to published and unpublished results, as detailed in the legend.

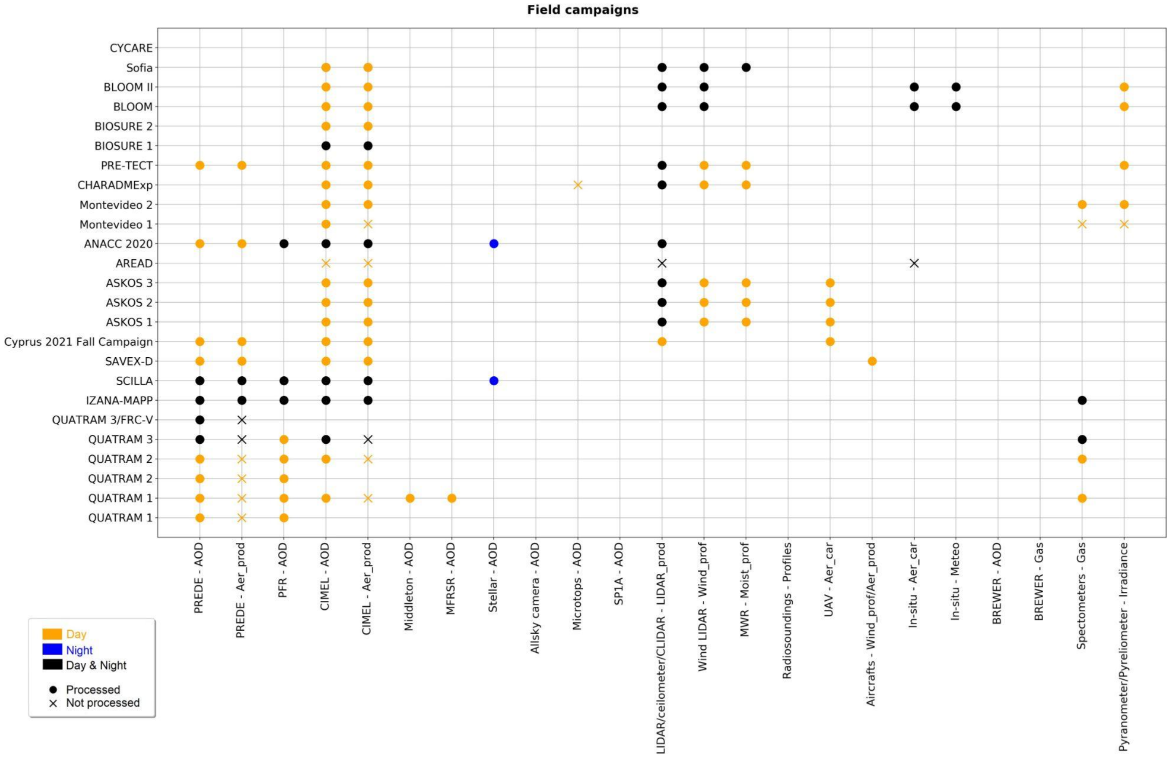

Figure 2.3: Instruments involved in each field campaign.

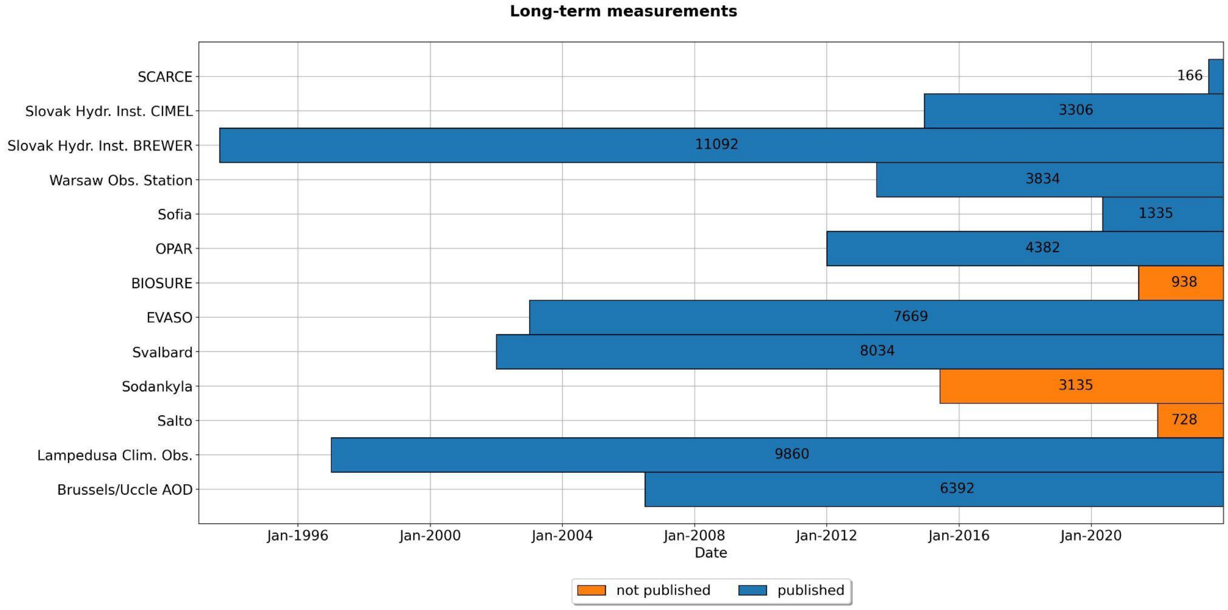

Figure 2.4: Temporal duration of the long-term measurements. The numbers indicate their duration (days), the colors refer to published and unpublished results, as described in the legend.

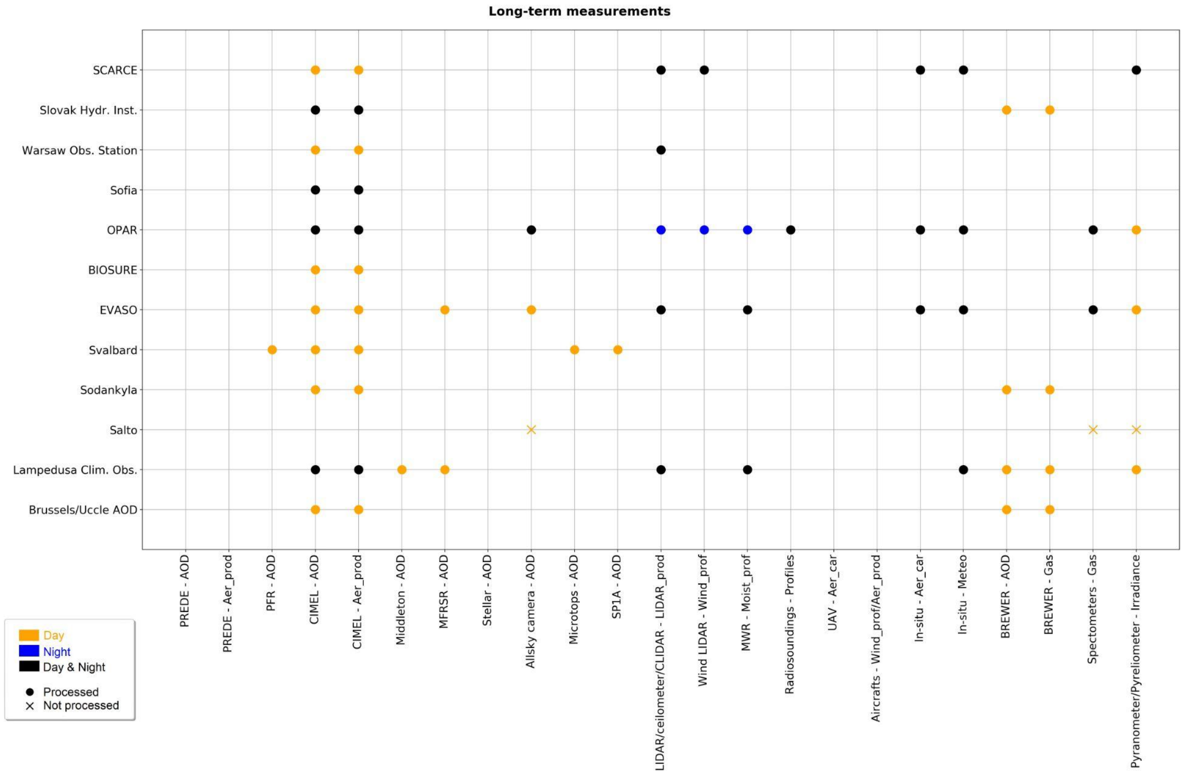

Figure 2.5: Instrument involved in the long-term measurements.

2.2 Permanent stations

Permanent observatories are important for several reasons. In fact, they allow to perform good quality long-term measurements thanks to the presence of the personnel which continuously check the status of the equipment. Moreover, they build large datasets of different instruments taking simultaneous measurements. These observations are important for climate models validation and models assimilation (Rubin et al., 2017; Randles et al., 2017; Benedetti et al., 2018; Gueymard and Yang, 2020; Mortier et al., 2020 ;Torres et al., 2021), satellite validation (Omar et al., 2013; Gupta et al., 2018; Sogacheva et al., 2020; Levy et al., 2018), climatological and trend studies (Holben et al., 2001; Che et al., 2018; Raptis et all., 2020; Barreto et al., 2022; García-Sunyer et al., 2023), and environmental effects (Kazadzis et al., 2016; Amiridis et al., 2009). Figure 2.6 shows the location of the surveyed permanent observatories, while the instruments operating are depicted in Figure 2.7 . Observatories are quite homogeneously distributed in Europe, but also in this case it is expected to add more sites in the next years of HARMONIA. Low cost instruments, PREDE sun and lunar photometers and stellar photometers are the equipment less deployed in the listed laboratories, whereas lidars/ceilometers and spectrometers are the most common.

Figure 2.6: Deployment of the permanent observatories.

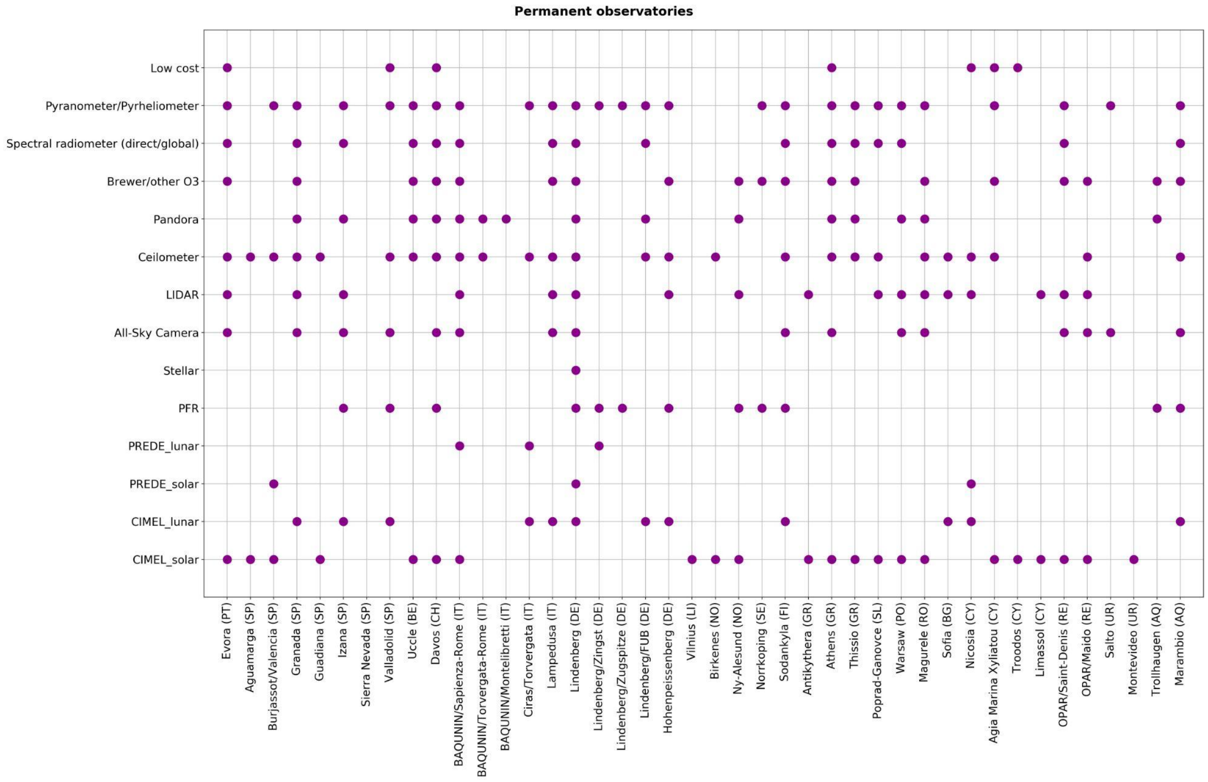

Figure 2.7: Instrument deployed in the permanent observatories.

3. Synergistic approaches for measurements

Sections 2.1 and 2.2 show the availability of a good amount of measurements taken both during night and day, and the accessibility to databases of different instruments. The simultaneity of aerosol vertical profiles from lidars, gases concentrations from spectrometers, is situ particles sampling and measures of meteorological parameters at ground level and along the atmospheric column, co-located in many campaigns and observatories, make possible accurate surveys and characterization of events that happen into the atmosphere, such as the passage of volcanic or fires plumes, but also pollution studies and radiative effects related to the aerosol optical properties. Below we report some synergist studies already performed. They can be considered a starting point from which to understand deficiencies and possible improvements.

In a report from a Virtual Mobility (VM) of the HARMONIA cost, J. L. Guerrero Rascado analyzed the horizontal extent and persistence of volcanic emissions from the Hunga Tonga eruption at the end of 2021, through synergy of ground-based Sun-photometry and satellite data. He performed a synergic analysis of the ash/SO2 plume horizontal extent from satellite imagery and observed aerosol background levels and precipitable water content from ground-based Sun-photometers and radiosoundings. He also found that the quality of water vapor retrieval from the sun-photometers needs to be improved.

Campanelli et al. (2021) conducted “A widE-Ranging investigation of the COVID-19 LOCkdown effects on the atmospheric composition in various Italian Urban Sites” (AER–LOCUS), with the aim of integrating observations from different platforms, such as in-situ and remote sensing, from ground-based and space-borne instruments. Particle and gas concentrations from in situ sampling, column aerosol and gas properties from photometers and spectrometers belonging to different observation networks, as well as TROPOMI NO2 determinations, were analyzed. This synergistic network of measurements, together with the examination of differences in meteorological conditions occurring in 2020, allowed to identify the medium- and long-range transport cases, and isolate the variations of the main atmospheric pollutants due to the restrictions. In fact, the measured concentration changes are not always due to variations in local emissions, as non-local particles and gases can be carried from distant places and the atmospheric structure and circulation can contribute to reduce or enhance the pollutant accumulation. The method for identifying the transport cases could be automatized and used in larger areas.

Andrés Hernández et al., (2022) provided an overview of the highlights from the data analysis of the EMeRGe (Effect of Megacities on the transport and transformation of pollutants on the Regional to Global scales) international project. EMeRGe focuses on atmospheric chemistry, dynamics, and transport of local and regional pollution originating in Megacities and other major population centers. Airborne measurements are a central part of the project. The synergistic use and consistent interpretation of observational data sets of different spatial and temporal resolution (e.g. from ground-based networks, airborne campaigns, and satellite measurements) supported by modelling within EMeRGe provide unique insight to test the current understanding of pollution outflows. The use of UAVs could be implemented in these kinds of synergic observations.

In Campanelli et al., (2012) results from a day-time intensive field campaign (URBan Sustainability Related to Observed and Monitored Aerosol – URBS ROMA) held in Rome (Italy) in June and July 2011 are presented. Chemical analysis of the aerosol particles was performed on particulate collected by PM10 collectors. Columnar aerosol optical and physical properties in clear sky were retrieved by using a PREDE sun-sky radiometer, part of SKYNET network. Vertical profiles of aerosol were obtained by a Lidar and incoming total solar radiation was measured by a Black and White Pyranometer. A Brewer spectrophotometer, a Sodar, and a MFRSR provided columnar ozone, wind profiles, diffuse and direct solar radiation at several selected wavelengths, respectively. The Rstar 6b radiative transfer code (Nakajima and Tanaka 1988) was used to calculate the vertical profiles of downward direct and total flux of solar radiation and of aerosol optical depth. The code was adapted to the needs of this work by changing: 1) "urban" standard vertical profile of aerosol according to the profiles of backscatter ratio measured by Lidar; 2) vertical ozone concentration according to the columnar ozone amount, measured by Brewer; 3) type and relative quantities of three chemical components of urban standard model according to the chemical analysis. The analysis of the direct effect of aerosol on surface incident solar radiation, by using Rstar code, was presented as a function of the changes in the measured mixtures of aerosol. From that time many improvements can be made to this kind of studies, including nighttime measurements and newly developed radiative transfer models.

Finally, an important issue to assess is whether, despite the marked technical differences between sun-photometers of different networks, their long-term AOD data are comparable and consistent. To this aim many studies were already done (Metrology for Aerosol Properties, MAPP 19ENV04, 2020), and campaigns performed (Cuevas et al., 2019; Kazadzis et al., 2018a) mostly involving instruments that are part of established international networks. A significant implementation is the inclusion in these studies of low cost and new developed equipment as resulted from the census performed in HARMONIA.

4. Synergistic approaches for algorithms

The use of different platforms measuring aerosol properties simultaneously, requires the development of synergistic approaches among the various algorithms that typically manage a multi-instrument database. The approaches to improve the quality and synergy of the algorithms mainly include: 1) development of algorithms; 2) intercomparison and validation of inversion algorithm products; 3) combined use of algorithms. Several lines of collaborative work were proposed in the different meetings during the first year of the HARMONIA cost action. Also, different short term scientific mobility (STSM) and virtual mobility (VM) grants took place that contributed to this general objective. Below we describe the different work lines and associated activities granted by the action.

4.1 Development and validation of in-situ improved calibration techniques

Sun-sky photometers and sun-photometers are instruments performing measurements of solar direct irradiance and diffuse sky radiance in the wavelength regions where gases’ absorption is low or negligible. Algorithms are therefore needed for both calibrations and data processing.

The solar calibration constant, which is the instrument counts for a direct normal solar flux extrapolated to the top of the atmosphere, is obtained using centralized calibrations on high mountain with the Standard Langley method, or on-site Langley calibration procedures (Campanelli et., 2004). These latter methods, performed as frequently as possible (daily, when possible, to get an average monthly calibration) in order to monitor the change of the machine condition, allow operators to track and evaluate the calibration status on a continuous basis, considerably reducing the data gaps incurred by the periodical shipments for performing centralized calibrations. By reducing the periodical shipments to calibration centers, research groups also reduce the instrument maintenance cost and the probability of calibration drift, damage, even of losing the instrument.

Because of the aforementioned advantages, the on-site calibration procedure has been long used in the frame of the SKYNET network. Previous validation studies have been performed with good results leading to uncertainties of 1 - 2% in the calibration constants of field instruments (Campanelli et al., 2004). However, current specifications on the AOD traceability demand reducing the associated AOD uncertainties further. Therefore, improvement of the on-site calibration procedures and associated QC criteria to make more robust determinations, is envisaged within HARMONIA.

In order to improve these algorithms, different approaches have been suggested: A) test of different new models for the inversion, that constitutes the core of the method; B) test of new QC criteria; C) assessment of site dependences and sensitivities; D) validations against calibrations provided by the Standard Langley method performed at high sites, or by traceable lamp calibrations performed at national metrology institutions.

A STSM granted to PhD student G. Kumar from University of Valencia (Implementation of a method for retrieving lunar AOD in different European Skynet Radiometers (ESR) network sites, hosted by the CNR at Rome, during 2 - 6 July 2023; https://harmonia-cost.eu/stsm-on-lunar-aod-from-prede-sunphotometer/) was partially devoted to the on-site procedure analysis within this objective. Harmonization of specific scripts, learning of particular procedures, and preliminary validation were performed. In relation to items A and D, two different inversion models SKYRAD version 4.2 and 5 (Nakajima et al., 2020) and GRASP (Dubovik et al., 2021) were tested against the Standard Langley method performed at a high-altitude station (Izaña observatory). The data chosen for the analysis is that from a campaign held in Rome in September 2021, included in the census list in section 2. The results showed some important sensitivity of the calibration constant on the algorithm used, that must still be deeply analyzed.

Therefore, future work will involve the detailed analysis of the data from the September 2021 campaign at Rome to propose using new QC criteria in the selection and averaging of monthly calibrations, following the items B and C. The proposed on-site procedure would be then applied to data from other campaigns in the census, in order to check the site specificity and limitations of the new method. The long run objective of this synergistic algorithm analysis is to develop a new proposal of a site independent on-site calibration method that includes an estimation of the associated uncertainties, that should comply with or better the current WMO limits.

4.2 Adaptation of GRASP inversion method to standard instruments from international networks

The GRASP (Generalized Retrieval of Atmosphere and Surface Properties) algorithm has become a reference for the retrieval of aerosol properties from different measurement platforms such as satellite, lidar, insitu or ground sun-photometry (Dubovik et al., 2011; Dubovik et al., 2014; Torres et al., 2017; Benavent-Oltra et al., 2019; Dubovik et al., 2021) and in fact it is already used with the Cimel CE318 measurements for the retrieval of the columnar aerosol properties and the associated uncertainty, using as input the AOD and sky radiance measurements.

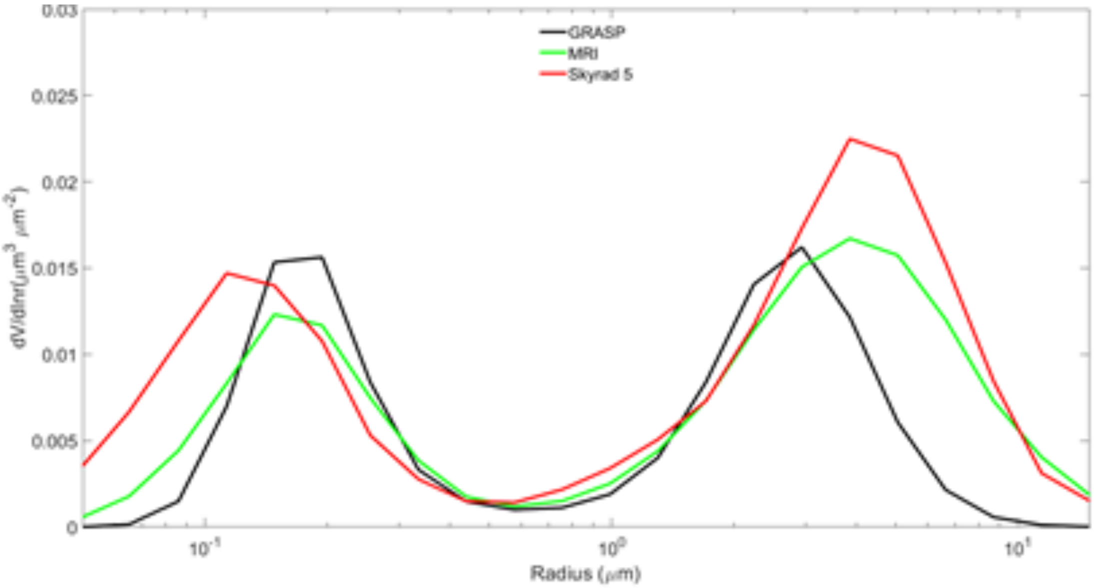

Figure 4.2: Preliminary comparison of volume size distributions obtained by SKYRAD versions 5 and MRI, and GRASP, for an example case on 1st September 2021 at Rome campaign.

During the first year of HARMONIA, adaptation to Prede POM radiometers has been started, not only within the on-site calibration procedure (section 4.1) but also for the inversion of aerosol properties, in a collaboration mainly between CNR at Rome, University of Valencia and GRASP team.

Preliminary results showed good correspondence with the inversion codes traditionally employed with the Prede radiometers, although more insight and adaptation are needed to get optimum results. In Figure 4.2., a preliminary comparison of the volume size distributions obtained with the SKYRAD algorithm versions 5 and MRI, and the GRASP retrieval, is shown. The general correspondence between the three distributions is very promising, mainly in the interval where the uncertainties are lower (0.2 – 2.0 μm). The particular differences between the three codes are still being investigated.

Future improvements of the quality of the aerosol retrievals of the Prede radiometers include: 1) the adaptation of the GRASP algorithm to Prede POM radiometers data; 2) assessment of differences between the different algorithms available using the Rome campaign held in September 2021 (but also other campaigns and permanent sites from the HARMONIA census, in order to check location dependencies of the results); 3) use of a synergistic approach to estimate the aerosol properties uncertainties from combined analysis of SKYRAD and GRASP codes.

The GRASP algorithm was also the objective of a STSM granted to A. Karanikolas from PMOD/WRC, which took place between 25 May and 2 June 2023, hosted by the GRASP team (“Retrieval of aerosol properties from solar irradiance data with high temporal and spectral resolution using GRASP”; https://harmonia-cost.eu/stsm-karanikolas-2023/). In this case, new algorithms for the adaptation of PFR and PSR radiometers data to GRASP were implemented, in order to retrieve effective radius and volume concentration separately for fine mode and coarse mode, aside from the total effective radius and total volume concentration. During the STSM, the work involved sensitivity analysis and comparisons with AERONET inversions. Data from the GAW-PFR station Hohenpeisenberg (Germany) and Izaña observatory (Spain) were used. From the comparison of the PFR retrievals for different settings it was deduced that the vast majority of retrievals were consistent. However, in future work the team needs to investigate whether this is true for different locations and years, by using data from other stations from the GAW-PFR network, such as Davos in eastern Switzerland. Also, it is important to know under which conditions the settings result in very large differences and how to potentially resolve the issue. The final goal is to add these parameters to GAW-PFR network products using GRASP and increase data quality and availability.

4.3 Validation of sunphotometer algorithms by use of simultaneous in-situ observations

Ground-based sunphotometers provide invaluable long-term information on atmospheric aerosols and are increasingly relied upon, providing columnar aerosol properties such as AOD, particle size-distribution (VSD), SSA, or refractive index. But significant discrepancies between inversion algorithms have been found over the time. No sufficient explanation is provided always for these differences, although it is crucial to understand the responsible processes and address them so as to provide harmonised aerosol products. Detailed analysis of these differences provide the means to better understand all the links between measurements to products.

Two field campaigns have been selected so far in the census compiled (section 2): the Sun photometer Airborne Validation Experiment in Dust (SAVEX-D 2015) campaign (Estellés et al., 2018) and the CyI UAV campaigns (Marenco et al., 2023). In both cases, aerosol size distributions were simultaneously measured with ground based sun photometers and in-situ instrumentation mounted on aircrafts or aerial platforms. This is an very valuable opportunity to address the homogeneity of AOD and VSD products obtained from different algorithms such as those from AERONET, SKYNET and GRASP, together with a validation with in-situ measurements in the vertical profile.

For example, the SAVEX-D campaign (Marenco et al., 2018; Ryder et al., 2018) was an experiment held in Cape Verde archipelago during summer 2015. Cape Verde islands are frequently and heavily influenced by the Sahara outflow of dust during summer months. Two different sun/sky radiometers were deployed at ground level (Cimel CE318 and Prede POM) in the island of Praia and performed continuous measurements in the period 1-25 August 2015. The algorithms used by-default for the retrieval of the VSD were AERONET and SKYRAD, in different versions. Simultaneously, two flights from the BAe-146 FAAM (NERC, Met Office, NCAS from UK) were performed in two different mornings of the campaign (16 and 25th August 2015) with clear skies and dominated by dust outflows from the Sahara, as shown in Figure 4.3.1. The average AOD during both flights were 0.4 - 0.6.

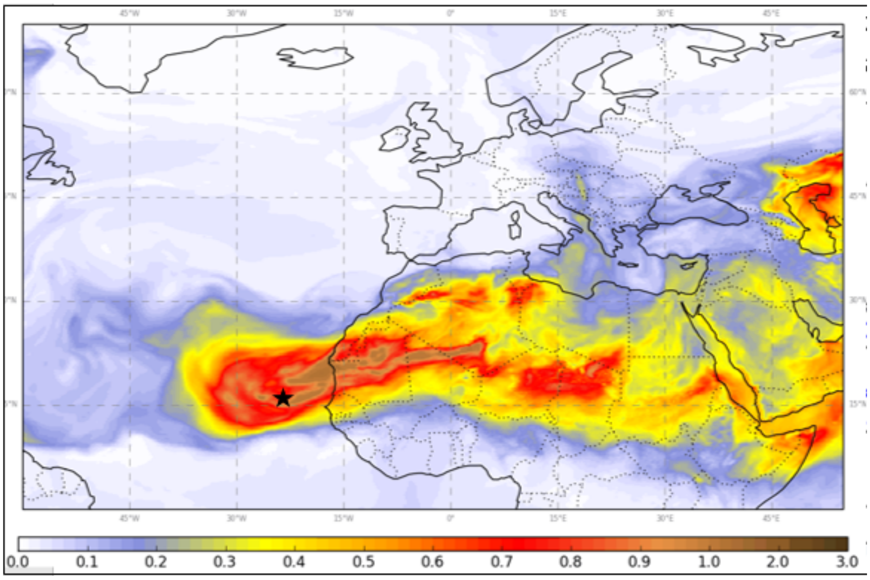

Figure 4.3.1: the Met Office operational global model dust forecast for 16 August 2015. Cape Verde archipelago is represented as a black star in the map.

Simultaneously, the BAe-146 aircraft overflown the area, carrying different in-situ instruments and a nadir-looking lidar. Insitu instruments (PCASP, CDP, CAPS, GRIMM, SMPS, CIP-15) measured the size distributions at several size intervals , and scattering and absorption coefficients were provided by a nephelometer and PSAP. The vertical integration of the insitu size distributions has been performed based on the information from the lidar and nephelometer profiles. In Figure 4.3.2, preliminary comparison with different algorithms for sunphotometer data inversion is also shown: AERONET algorithm version 2 applied on Cimel CE318 data; Skyrad versions 4.2, 5 and MRI applied on Prede POM radiometer data.

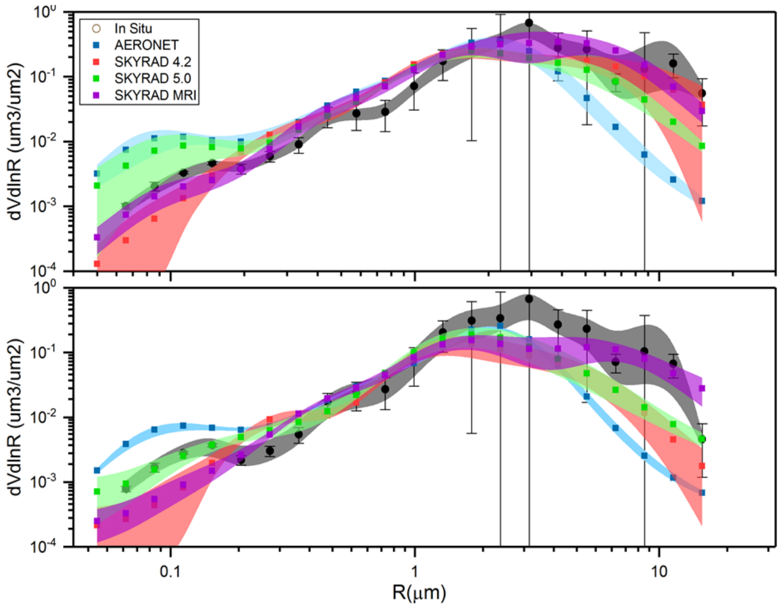

Figure 4.3.2: Comparison of size distributions for days 16th (up) and 25th (down). Grey: airborne distributions; Blue: AERONET; Red, green, purple: SKYRAD. Shadow bands indicate standard deviations during flight sample plus sensitivity analysis to the vertical integration method.

From the visual analysis of the Figure 4.3.2, it can be seen that in general, the different algorithms approach the reference in-situ size distribution very well, for both the flights, mainly in the interval 0.2 - 2 μm, where the inversion methods are known to perform better. From all the algorithms used, it looks apparent that SKYRAD version MRI approaches the reference better, for these two cases.

However, the analysis of the SAVEXD campaign is not complete. Newer developments of the AERONET and SKYRAD algorithms have been provided in recent years; therefore, it is important to use this same dataset for further refinement and validation of the new codes. Same time, it would be of ultimate importance to include in the validation the GRASP retrievals that will be performed on both Cimel CE318 and Prede POM datasets, once the GRASP algorithm is adapted to Prede radiometers data, within HARMONIA section 4.2.

Another interesting set of campaigns identified thanks to the census created in HARMONIA are the 2021 Fall campaign from Cyprus Institute. Collaboration between WG2 members including mobility grants for students are intended in order to use these results for the objective of section 4.3. This set of campaigns include the simultaneous use of ground based columnar measurements and unmanned aerial vehicles (UAV) (Kezoudi et al., 2021) for vertical profiling of the atmosphere with in-situ instrumentation, including light-weight particle sizers but also aethalometers, up to an altitude of 6000 m. Similarly to SAVEXD campaign, these campaigns are oriented at the study of dust, and constitute a very valuable dataset for the validation of ground based columnar retrievals with integrated profiles of in-situ data. Several AERONET sites (Cimel CE318) and a Prede POM radiometer are available in Cyprus during the 2021 Fall campaign (see Figure 2.3).

4.4 Validation of nighttime AOD obtained by different types of lunar and stellar instruments.

Sun/sky radiometers have been long used to retrieve the AOD during the day time, by applying the well-known Beer-Bougher law and using the sun as a constant and powerful radiation source. However, the AOD could not be retrieved during night time: the radiation reflected by the moon is much weaker and very dependent on the moon phase; and the radiation arriving on the earth from the far emitting stars is even weaker, pushing the technology to its limits and leading to high uncertainties in the estimated AOD, when measured. This limitation creates systematic gaps in the data series for periods of hours every 24 hours at mid or low latitudes, and during periods of months in polar regions (polar nights). It then puts a limitation on our understanding of daily and seasonal variation of aerosols, and its dynamics and transport mechanisms at night time. Improvement of AOD measurements needs more sensitive hardware and adapted algorithms. For example, Barreto et al (2013) proposed new methods to perform calibrations, and to retrieve the aerosol optical properties, using the new lunar-pointing versions of the CIMEL CE318T sun/sky photometer. Recently, Uchiyama et al. (2019) also proposed alternative methods to calibrate and retrieve aerosol optical properties using lunar versions of the PREDE POM radiometers. Other approaches to determine the AOD at night consist of star-photometers that measure the extinction of light coming from one or two known stars (Doppler et al., 2015) but in general these instruments are more complex to operate, leading to scarcity of global data. Other types of compact stellar instruments are being developed in order to provide night AOD at harsh remote environments, but they are still not fully validated, and then are subject to improvement. Therefore, new hardware and algorithm development and improvement are being investigated within HARMONIA.

Currently, the most spread and known lunar AOD data is available from AERONET lunar mode. The standard hardware is the lunar Cimel CE318T sunphotometer, and the algorithm is based on Barreto et al. (2013). The moon is not a uniform source of light, because it has an uneven surface and the sun-moon-earth angular configuration (lunar phase) changes everyday. In order to get the equivalent disk reflectance of the lunar surface, the ROLO model (Kieffer and Stone, 2005) was adapted.

For example, a preliminary analysis of the lunar AOD from AERONET has been performed within the first year of HARMONIA, using solar and lunar Cimel CE318T data obtained at Burjassot site during years 2015 - 2022, level 2.0 (Burjassot station included both in AERONET and SKYNET, as seen in the census described in section 2). A good correlation between solar and lunar AOD has been found, although more insight is needed, because temporal variations happening during both the day and night time lead to an underestimation of the correspondence between both AOD methods. In order to check the goodness of the lunar AOD in comparison to solar AOD, a combined representation has been done in Figure 4.4.1 for a short period of summer 2022 (10 - 20 August 2022) coincident with a saharan dust episode. It is clear that the continuity of AOD data has improved very much thanks to the lunar products, pointing at the need for the synergistic use of the two types of algorithms. This is particularly important when rapid variations exist, such as at night 16 to 17th August. In order to improve the AOD validation, we plan to compare the lunar and solar AOD at the end of the day/beginning of the night, when some criteria apply, such as a maximum temporal mismatch, maximum variability, and analysis of colocated ceilometer or lidar data to ensure the continuity of the aerosol conditions during the intermediate period. The validation analysis will be extended to other permanent sites in the HARMONIA census, in order to understand causes for occasional differences between both selected datasets. Both VM or STSM grants type will be very valuable to exchange knowledge between our network members in order to do further research on this issue.

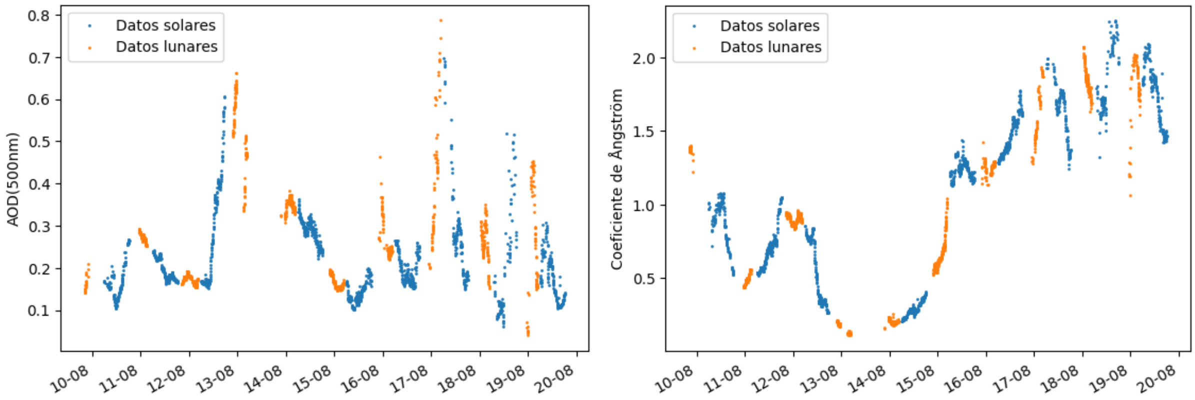

Figure 4.4.1: Evolution of (a) AOD at 500 nm and (b) Angström exponent during the period 10-20th August 2022, using both solar and lunar AOD datasets from AERONET website. The period 12 - 15th August corresponds to a saharan dust intrusion.

Lunar AOD measurements are also being performed with the lunar photometer PFR-L manufactured at PMOD/WRC. The instrument is able to provide lunar irradiance measurements with an uncertainty of less than 0.8%. Previous comparisons in the frame of the Izaña 2022 campaign showed some differences attributable either to the RIMO model or the SI-calibrated sun-photometers. Therefore, during the first year of HARMONIA, N. Kouremeti from PMOD/WRC was granted a two month VM whose final objective was the improvement of the lunar AOD retrievals ("Improving Lunar PFR aerosol optical depth retrievals"). Within this VM grant, a lunar campaign has been held at Rethymno station (University of Athens, Greece) during two lunar phases in Summer 2023, in which the reference PFR-L002 was deployed alongside two other new lunar PFR radiometers, and a PFR sun-photometer. Results are still being examined.

Recently, Uchiyama et al. (2019) also developed a method similar to that of Barreto et al. (2019), to be applied on new lunar sky-radiometers Prede POM01L from the SKYNET network. The implementation of the proposed method on the ESR-SKYNET system was performed during a STSM granted to PhD student G. Kumar from University of Valencia (hosted by CNR at Rome, 1-6 July 2023; https://harmonia-cost.eu/stsm-on-lunar-aod-from-prede-sunphotometer/). Data from two recent field campaigns listed in the HARMONIA census has been used for the comparison between both AERONET and SKYNET algorithms: the QUATRAM3 field campaign, held in Rome during September 2021; and the Izaña field campaign, from the MAPP project, held in the Izaña AEMET observatory (Tenerife, Canary Islands) during September 2022.

As an example, in Figure 4.4.2, the differences between the AOD downloaded from AERONET and calculated for the Prede POM radiometers, implemented during the STSM, are presented simultaneously to the WMO prescribed thresholds (0.01/m±0.005, where m is the optical mass) for the Rome campaign, and for the 4 channels available. The RMS differences for every available channel is less than 0.008, and maximum standard deviation is 0.090. This clearly indicates good agreement between the methodology implemented during the STSM, and AERONET.

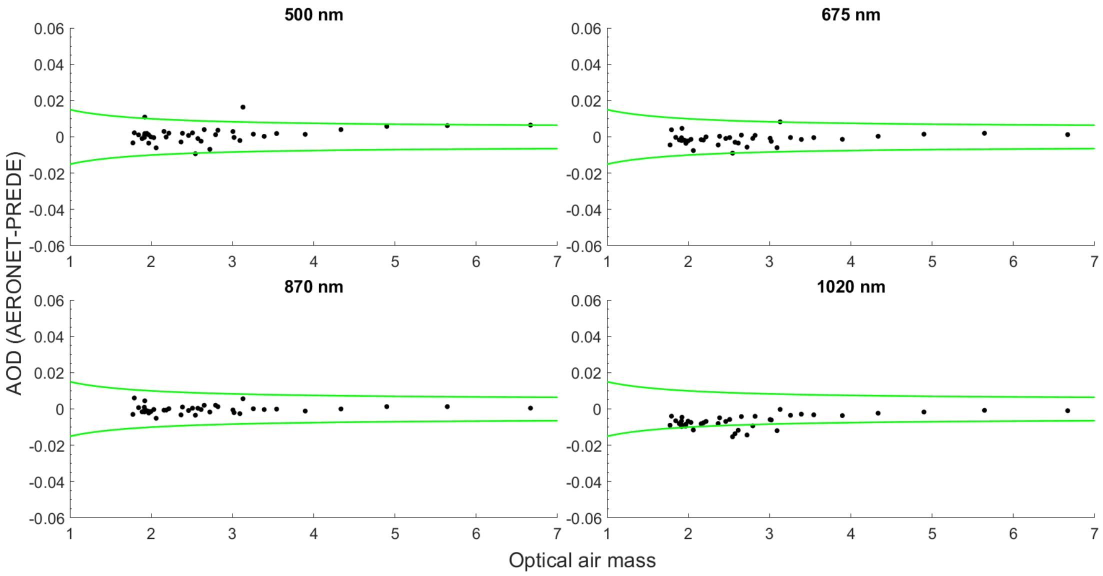

Figure 4.4.2: Representation of the AOD differences between new implemented methodology in ESR-SKYNET and standard AERONET algorithms. The green lines superimposed are the WMO recommended limits. Figures refer to the Rome 2021 campaign.

Further improvement of the recent ESR/SKYNET algorithms, to be developed within HARMONIA, include the validation/improvement of the lunar calibration method in different environments and lunar phases, correction of straylight effects, estimation of uncertainties and the improvement of the cloud screening method. For this, during the next years we expect not only to use new existent datasets declared in the HARMONIA census, but also to perform new experiments in high altitude stations and near sea level light-polluted environments. Collaboration between different members will be considered for the improvement and comparison of the AERONET, GAW/PFR and ESR/SKYNET lunar AOD algorithms.

4.5 Harmonization of algorithms between low-cost instruments and sky-cameras and well established references.

Low-cost sensors (LCS) for measurement of environmental parameters and atmospheric composition have increased their popularity during recent years, as it can be deduced by analysis of the number of publications in peer-reviewed journals (WMO, 2020). Its use has increased due to some advantages such as lower price, smaller size and lower power consumption, that makes them very interesting for some applications: UAV operation, mobile ground platforms, extensive networks. However, their measurements are not always reproducible, nor traceable. In order to tackle this limitation, it is needed to perform comparisons with well-established references, in indoor (well controlled) or outdoor (realistic) conditions, and to adapt well known algorithms for the harmonisation with other reference instruments or networks.

The appearance of new optical and electrical technologies at lower prices have made possible the manufacture of hundreds of LCS systems for environmental control, mainly for in-situ pollution control. In relation to our field of interest (aerosol sun-photometry) we have considered only manual sun-photometers such as Microtops or Calitoo; sky cameras; and other types of sensors such as the photometer model ZEN-R52.

The Microtops II and Calitoo instruments are light weight, affordable, and portable sun-photometers that must be manually pointed at the sun in order to retrieve the AOD (Figure 4.5.1). Absence of automatic mobile parts are the reason for their affordability; but the internal construction of the sensor does not differ greatly from more advanced reference sun-photometers. In the case of the Microtops II, previous comparisons showed a good correspondence with AOD measurements (from co-located Cimel CE318 AERONET instruments) and ozone content (from co-located Brewer spectrometers) when a fresh calibration is provided. In fact, the instrument is the reference sun-photometer for the MAN network (Smirnov et al., 2009). The Calitoo sun-photometer is an even more affordable instrument that can be used, for example, for public science and teaching. Within HARMONIA, several members have used these portable sun-photometers in field campaigns as CHARADMExp, or during long-term experiments like in Svalbard (see section 2.1). Possibility to adapt current reference algorithms on these instruments data could be done within HARMONIA.

Figure 4.5.1: Microtops II and Calitoo portable sun-photometers (source: https://solarlight.com/ and http://www.calitoo.fr/).

All-sky cameras are another type of instrument considered low-cost whose recent development has led to important advantages, although there is still room for improvement. Although all-sky cameras have been traditionally devoted to cloud fraction estimation, the sky radiance distribution also contains useful information for aerosol properties determination. The radiometric calibration of the sky cameras is not straightforward nor easily traceable; however, the normalised sky radiances (NSR) in arbitrary units, which are easier to obtain, measured along different angles in the sky, can also be used for aerosol inversion. To test this possibility, Román et al. (2022) inverted the NSR with an adaptation of the GRASP algorithm (Dubovik et al., 2021) also used for the inversion of other reference instruments, contributing to the harmonisation of algorithms and products. A sensitivity study was conducted, using synthetic data from GRASP forward module, in order to estimate the uncertainties involved; and an intercomparison against aerosol AERONET products, like AOD, was performed using a 2-year dataset from a Cimel CE318 photometer installed in the Valladolid permanent station (census section 2). Current research also explores the mult-pixel approach of GRASP applied to daily camera radiances, including a constraint on the temporal variation of certain aerosol properties that is expected to improve the temporal variability of the 1-pixel method. This analysis has been undertaken as a collaboration between different members from HARMONIA cost action.

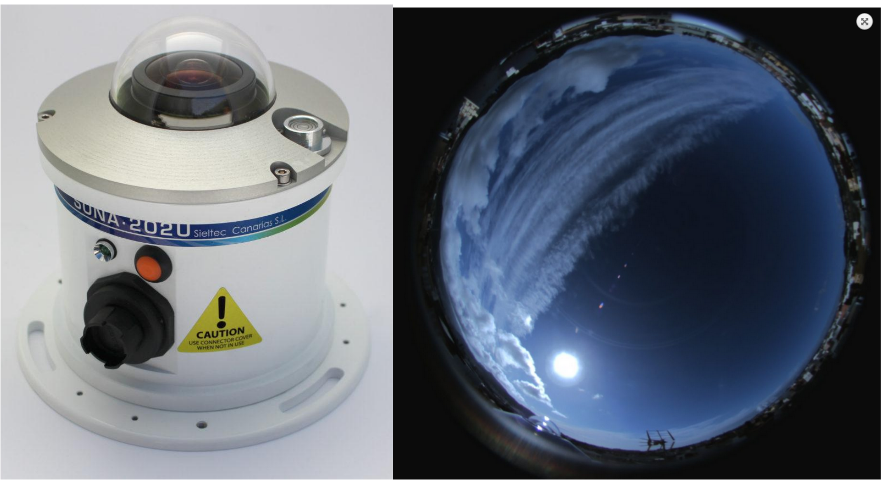

Figure 4.5.2: Example of current technology (a) all-sky camera; (b) RGB image (source: https://www.sieltec.com.es/sona/).

Another LCS considered is the ZEN-R52, manufactured by Sieltec Canarias (Figure 4.5.3). It is a photometer without mobile parts that measures sky radiance at the zenith direction (ZSR) in 4 aerosol channels: 440, 500, 675, and 870 nm. The original algorithm to retrieve the AOD with the ZEN-R52 consisted of comparing the spectral measurements with a previously built Look Up Table (LUT), that was location sensitive (Almansa et al., 2020). In order to improve the quality, homogeneity and spatial representativity of the retrievals, the GRASP algorithm (Dubovik et al., 2021) has been adapted to the ZEN-R52 data type, and new aerosol properties and associated uncertainties can now be obtained (Herrero-Anta et al., 2023). The comparison to an AERONET Cimel CE318 used as a reference at the Valladolid permanent site (census described in section 2) shows good results, with an uncertainty of 0.02 - 0.03 for the spectral AOD. For the calibration of the photometer, the ZEN measurements were compared with the sky radiance obtained with the GRASP forward module, fed with input aerosol data independently obtained by the colocated Cimel CE318 instrument from AERONET. This algorithm improvement has been the result of a collaboration between the University of Valladolid, GRASP SAS, AEMET, and Cimel Electronique, within HARMONIA.

Figure 4.5.3: Sieltec ZEN-R52 photometer (source: https://www.sieltec.com)

4.6 Recent proposals of cloud screening methods for sun-photometric databases.

Cloud screening algorithms have been shown to be important sources of discrepancy between aerosol properties retrieved by AERONET, PFR/GAW, SKYNET and other networks (Kazadzis et al., 2018). The different methods rely on sets of criteria about the temporal variability of the irradiance or AOD measurements. Harmonisation of these algorithms is envisaged, but new proposals are also convenient for a better rejection of cloud contaminated data. Although some types of cloudiness are easily rejected, others such as cirrus, are more complicated to be identified, and need more complex approaches.

With this objective, a STSM was granted to PhD. V. Schenzinger from Institute for the Biomedical Physics Institute at Innsbruck ("Clearing cirrus clouds from AOD measurements", period 19/03/2023 to 01/04/2023, hosted by PMOD/WRC; https://harmonia-cost.eu/625-2/). In this STSM, the recent proposal from Schenzinger and Kreuter (2021) was applied to PFR instruments from Davos and Izaña stations, and compared to the current cloud screening method used in PFR/GAW and AERONET networks.

The Schenzinger and Kreuter (2021) method is a nearest-neighbour clustering algorithm in the context of Machine Learning applied to outlier detection, that takes into consideration the AOD at 500 nm, its temporal variation, and the Angström exponents α and γ, similar to the δα used in the Gobbi et al. (2007) method. It was developed and tested initially using PFR data from the Innsbruck station for the identification and flagging of cirrus clouds, with good results. To contribute to the harmonisation objective, the method was applied in this STSM on the Davos station data (low AOD) and Izaña observatory data (low AOD, but with occasional strong dust episodes that make the situation more challenging for the proposed algorithm validation). The database used in the analysis consisted of one full year (2019) of AOD measurements from Davos (PFR and CIMEL) and Izaña (PFR).

The results of the analysis showed that the algorithm performs very well, even in different environments than originally tested; but in low AOD conditions, thin clouds did not reliably get identified, flagging more data points than the original algorithm. Therefore, more work is needed in order to improve this method so it can be operatively applied in different environments.

Other alternatives for cloud-screening can be adopted from solar radiation measurements, specifically the Long and Ackerman (2000) algorithm. This method has been long used for cloud screening pyranometers datasets with a 1-minute resolution. Combined use of the sun-photometric and solar radiation cloud screening algorithms has been recently implemented by the Deutsche Wetterdienst (DWD) (example case is shown in Figure 4.6.) but still needs some more development for an operative application that is desired to be undertaken within a collaboration between HARMONIA members.

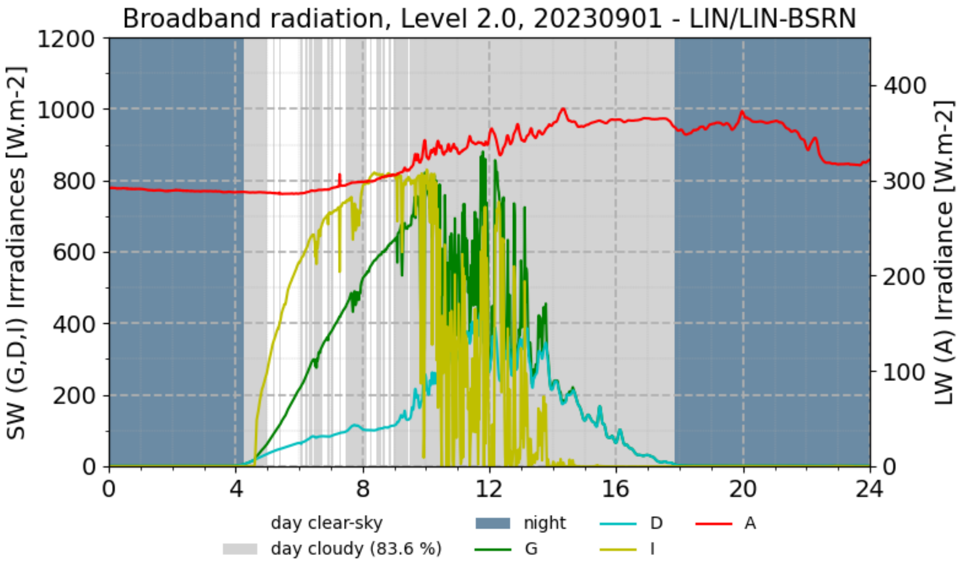

Figure 4.6: Short and long wave integrated irradiances for an example day (01/09/2023) in the Lindenberg BSRN station. In grey bands the data screened by Long and Ackerman (2000) method, also used to cloud screen sun-photometers data.

Unfortunately, not all the sun-photometric sites are equipped with auxiliary solar radiation measurements, which makes the previous approach of limited utility. To pursue further synergy between solar radiation and sun-photometric algorithms without a need for third instruments, other solar radiation cloud screening methods could be explored. For example, the Xia et al. (2007) method is a simplified approach, based on Long and Ackerman (2000), that only makes use of global solar irradiance. In this case, its adaptation to direct irradiance measurements taken by the sun-photometers, could be interesting. Auxiliary solar radiation measurements would not be needed in this case.

4.7 Improvement of the gas absorption correction.

The retrieval of AOD and other aerosol properties is performed in atmospheric windows where the effect of absorption from atmospheric gases (NO2, CO2, H2O, O3...) is inexistent or negligible. However, most aerosol channels suffer from small effects from gases absorption whose contribution needs to be estimated. To improve the gas absorption contribution, we need: a) improved methods for the gas absorption calculation; b) accurate measurements of the gas columnar content; c) increased temporal resolution of the gas component.

For example, a recent collaboration between HARMONIA members has determined the effect of columnar NO2 on the aerosol optical properties retrievals, namely the AOD, Angström Exponent, and SSA (Drosoglou et al., 2023), for both AERONET and SKYNET products, by using a 5-year database at the Rome permanent site from the HARMONIA census (section 2). The columnar NO2 data was obtained by a colocated Pandora instrument, although satellite based measurements (TROPOMI) were also tested. Although it is evident that the correction will be small for background sites with a low columnar NO2, the effect on AOD was not negligible for areas with high NO2 columnar content (>0.7 DU), producing biases as high as 0.009-0.012 for AERONET, and 0.018 for SKYNET. Therefore, measurement and algorithm improvements are of great interest in order to correct the aerosol optical properties products produced by international networks. More research is needed in order to produce harmonized results also in the gas absorption estimation.

5. Needs and recommendations

The information gathered during the census on instruments, database, and processing in the campaigns, long-term measurements and permanent observatories, needs to be available to the scientific community. To this purpose it has been recommended by WG members and invited experts to integrate the information in a catalogue, with user friendly characteristics and a public use. This suggestion will be evaluated taking into account the possible contributions and capabilities from the community.

The review of recent published or ongoing analysis about algorithm and measurements synergies and harmonisation led us to identify and propose several possible lines of improvement work that will be explored during the following years of HARMONIA, thanks to sharing the databases listed in the created census, establishing collaborations between members, and granting mobility opportunities. Specifically, we have identified the following research lines towards aerosol properties retrieval improvement:

1) Improvements towards a better performance of the techniques and the reduction of associated uncertainty. It includes improvements related to calibration issues and post-processing of data, because they are a key aspect for the accurate and reliable retrieval of aerosol properties in international networks. Particularly, we have described in detail current activities in the validation of the in-situ improved calibration technique used by SKYNET. It has been also shown that in polluted atmospheres the high concentration of some gases can lead to non-negligible biases in the reported aerosol data. Therefore, it is also needed to improve the treatment of the absorption processes not only in the VIS but also in the UVB and NIR regions for the corresponding aerosol retrieval. It is also important to validate night time AOD obtained by different types of lunar and stellar instruments, as the lunar algorithms still have room for improvement and have not been completely developed, so the provided data will be very important to fill gaps in our knowledge of the aerosol dynamics during night-time.

2) Assessment of new instruments including low cost sensors (short and long term comparisons). The harmonization of algorithms between low-cost instruments and sky-cameras and well established references is needed, as the use of low-cost instruments is spreading rapidly for environmental applications, providing products that are still not still completely assessed.

3) Improvements towards enhanced products with different instrumentation. For example, we have described current activities on the GRASP algorithm application to PFR and SKYNET instruments, enhancing the current outputs of the PFR sun photometers (providing in addition to AOD, the effective radius, etc) or the uncertainty estimation of SKYNET retrievals. Other possibilities include the application of GRASP to other inputs such as LIDAR, where fine, coarse mode properties profiles can be derived; or to multi-instrument synergies (spectroradiometer based, FTIR).

4) The need to work more on Artificial Intelligence (AI) and Machine Learning (ML) techniques. The AI and ML based techniques are becoming popular tools in many research fields. Some particular proposals have included its use for cloud screening of sun-photometric databases, as the existent methods are not always completely consistent, especially in situations where cirrus are dominant. But in general, the AI techniques have an important potential in other applications in the aerosol retrieval field too, such for example, aerosol classification.

5) The need to trace the instruments calibration to SI. Recent attempts have been tried to link the current calibration methods from international networks with SI sources, with variable results. Future recommendations include the need for the improvement of the traceability of network calibrations and products.

6) The need to link sun photometer uncertainties with actual user needs. They include the satellite and modelling communities, that rely on the products delivered by international networks of sun and sun/sky radiometers, such as AERONET, GAW/PFR and SKYNET.

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