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MAPP

Metrology for aerosol optical properties

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1 Overview

The overall aim of this project was to enable the SI-traceable measurement of column-integrated aerosol optical properties retrieved from the passive remote sensing of the atmosphere using solar and lunar radiation measurements. In order to validate and improve the current aerosol optical property retrievals using state-of-the-art inversion models, radiometers of the three largest aerosol monitoring networks were calibrated at NMI laboratories, and portable devices for the in-field calibration of network radiometers were developed. Comprehensive uncertainty estimates for the column averaged aerosol optical properties were developed, to underpin the measurements of the three global networks involved in this project. These aerosol uncertainty calculations were implemented in the inversion code GRASP which is a publicly available code to the scientific community to retrieve aerosol optical properties from solar irradiance measurements. The top of the atmosphere solar spectrum TSI-1 HSRS was validated with respect to traceable ground-based measurements to provide a validated and traceable solar spectrum to use for aerosol optical depth retrievals from calibrated solar irradiance measurements.

2 Need

Global climate assessments require harmonised and quality-controlled datasets. This implies measurements preferably traceable to the SI and cross-network wide implementations of calibration and post-processing procedures.

Atmospheric aerosols are minor constituents of the atmosphere, but a critical component in terms of impacts on the climate. Their properties have been recognised as **Essential Climate Variables (ECVs)** by the Global Climate Observing System (GCOS). As pointed out in all previous IPCC (Intergovernmental Panel on Climate Change) reports, aerosols continue to contribute the largest uncertainty to estimates and interpretations of the Earth's changing energy budget.

Long-term monitoring of aerosol ECVs including their uncertainties is needed for observing sensitive changes in the Earth climate system. Currently the three main global surface-based networks measuring column integrated aerosol optical properties in Europe, AERONET Europe, GAW-PFR and SKYNET Europe, operate over hundred stations with complex calibration strategies based on artefact reference devices. The lack of an appropriate metrological framework in the calibration, operation, and data processing of these networks rendered the whole monitoring concept very convoluted, time consuming, labour intensive, and uncertain.

Calibrations of reference radiometers have been performed at only two stations world-wide (Mauna-Loa, Hawaii or Izaña, Tenerife). The subsequent calibration of network radiometers were based on such reference artefacts relies on instrument stability and instrument redundancy and not on objective metrological concepts. The field calibration procedure of calibrating network radiometers at centralised calibration stations outdoors requiring perfect weather conditions meant long calibration times and therefore excessive downtime at monitoring stations.

Emerging technologies such as compact array spectroradiometers or portable Fourier spectrometers offer the potential for enhanced atmospheric products providing significantly more information on the radiative forcing impact on climate.

The need for an objective assessment of uncertainties of the retrieved aerosol properties is an inherent property of traceable measurements, which was lacking in all three main global aerosol monitoring networks. The databases providing access to the network data did not offer concurrent uncertainty estimates.

3 Objectives

The overall goal of this project was to enable the SI-traceable measurement of column-integrated aerosol optical properties (ECVs) for assessing the radiative forcing impact on the climate. These properties were retrieved from the passive remote sensing of the atmosphere using solar and lunar radiation measurements that were largely lacking traceability to the SI. The specific objectives of the project were:

1. To develop calibration methods and traceable devices for SI-traceable laboratory and in-field calibrations for radiometers measuring direct solar spectral irradiance and sky radiance, in the spectral range 310 nm up to 1700 nm with an expanded uncertainty of 1 %. Instruments measuring lunar irradiance were included into the scope of the objective following the expressed needs of the stakeholders.



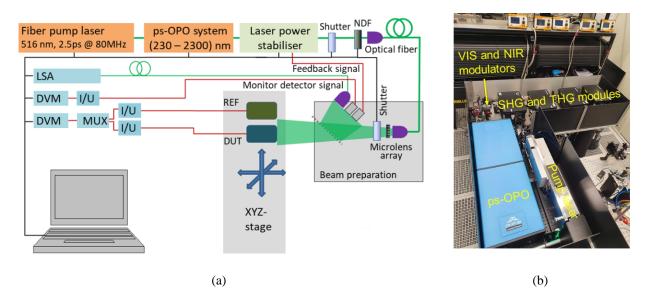
- 2. To validate the methods for zero airmass extrapolation by means of traceable ground-based solar spectral irradiance measurements and comparison to satellite-based solar extra-terrestrial spectra. Due to the growing importance of lunar irradiance measurements to retrieve AOD during night-time conditions (e.g. polar winter), validations of lunar irradiance phase models were also addressed.
- 3. To develop a comprehensive uncertainty budget for aerosol optical properties, such as aerosol optical depth, aerosol size distribution, and aerosol single scatter albedo, retrieved from remote sensing-based measurements of direct and scattered solar radiation, enabling its inclusion in the corresponding data archives of the aerosol monitoring networks, with the relevant calibration and traceability information.
- 4. To facilitate the take-up of the developed technology and measurement infrastructure developed in the project by the measurement supply chain (NMIs, DIs, and calibration laboratories), standards developing organisations (WMO CIMO, WMO GAW SAG) and end users (e.g. ground-based and space-based remote sensing and atmospheric science communities). In addition, to support the European Metrology Network for Climate and Ocean Observation.

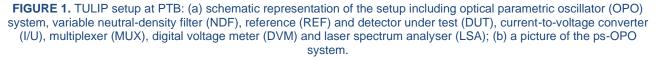
4 Results

4.1 To develop calibration methods and traceable devices for SI-traceable laboratory and in-field calibrations for radiometers measuring direct solar spectral irradiance and sky radiance, in the spectral range 310 nm up to 1700 nm with an expanded uncertainty of 1 %. Instruments measuring lunar irradiance were included into the scope of the objective following the expressed needs of the stakeholders.

The established calibration procedures for filter radiometers, also known as sun photometers, employed in the aerosol monitoring networks, such as AERONET, GAW-PFR, SKYNET, are based on comparisons with the reference instruments in each network, which in turn are calibrated at specific high-mountain locations, e.g., Izana, Mauna Loa, to derive the signals they would measure on top of the atmosphere. Objective 1 is aimed to enable calibration of such radiometers in SI units using laboratory procedures. To achieve this, the consortium partners have developed calibration and characterisation methods and instruments. The activities to achieve the objective were divided into four tasks carried out in Work Package 1.

Spectral irradiance calibrations





PTB has expanded the measurement capabilities of its narrowband tuneable laser-based setup TULIP (TUnable Lasers In Photometry) to enable absolute calibrations of sun photometers for their spectral irradiance responsivity up to 1.7 μ m with uncertainties in the range 0.1 % to 1 %. In particular, power stabilisation and



wavelength monitoring of the tuneable laser system and appropriate transfer standards for the near-infrared region have been developed. The TULIP facility, shown in Figure 1, is built around a laser system based on an optical parametric oscillator (OPO) operating in pulsed mode with a pulse length of 2.5 ps and a repetition rate of 80 MHz. The laser wavelength is automatically tuneable throughout the spectral range from 230 nm to 2300 nm. A high-accuracy laser spectrum analyser (LSA) is used to monitor the laser wavelength, which is stable within 10 pm during a typical measurement sequence. A spatially homogeneous, non-polarised radiation field with temporally stabilised irradiance values is generated by beam shaping optics based on a micro-lens array. The amplitude stabilisation of the output radiation from the laser system is achieved using two liquid crystal display (LCD)-based modulators inserted in the signal and idler beams of the OPO, before the second and third harmonic (SHG and THG) modules of the laser system. The feedback signals for the control circuits of the intensity modulators are taken from a Si and InGaAs photodiodes irradiated by a fraction of the radiation field formed by the micro-lens array. In this way, the irradiance values at the measurement plane are stabilised to a level of a few parts in 10⁴. Spectral irradiance responsivity calibrations are made in such a field by comparing the signal of a device under test (DUT) to that of a reference detector (REF) that are positioned consecutively to the same position in the measurement plane. The spectral irradiance responsivity of the reference detectors built of Si and InGaAs photodiodes for the visible and near infrared wavelengths, respectively, is obtained through a chain of calibrations from a primary cryogenic radiometer and from the calibrated area of precision radiometric apertures used with the reference detectors. The tuneable laser setup at PTB was used to calibrate the spectral irradiance responsivities of two instruments from GAW-PRF network provided by SFI Davos. One GAW-PFR radiometer was designed for measuring direct solar irradiance and another one for lunar irradiance. Also, three Cimel CE318T filter radiometers from AERONET Europe were calibrated at PTB for their spectral irradiance responsivities. The instruments were provided for the measurements by AEMET, UVa, and CNRS. Finally, two Prede POM filter radiometers from the SKYNET Europe provided by UV and CNR were calibrated at PTB as well.



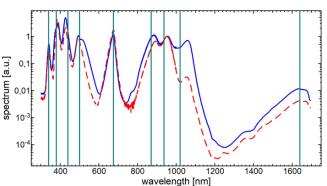


FIGURE 2. Left: LED-based source attached to an integrating sphere; CE318T radiometer is positioned in front of the output port of the sphere. Right: relative spectral distribution of the optical radiation emitted by the first version (red) and the second version (blue) of LED-source in arbitrary units. The green lines indicate the wavelengths of the radiometer channels.

In order to detect changes in the irradiance responsivity of the AERONET Europe reference radiometers, PTB, in cooperation with CNRS, developed a portable LED-based source flangeable to a port of an integrating sphere (Figure 2). The design of the LED unit comprises 9 surface mount device (SMD) light emitting diodes (LEDs) soldered to a printed circuit board (PCB). One LED is used for each band of the sun photometer, spread over the spectral range from 340 nm to 1640 nm. The temperature of the LEDs is controlled by a Peltier device attached to the other side of the PCB. The LEDs are connected in series and operated in stabilised current mode. Two versions of the LED devices have been built and studied within the project. The devices were characterised in terms of spectral characteristics, stability and reproducibility of the emitted radiant flux, as well as homogeneity of radiance distribution over the output port of the sphere. It turned out that the stability of the source is limited by the throughput stability of the integrating sphere. This again depends on the type of material the sphere is made of and the ambient conditions. A barium sulphate-coated sphere showed a significant dependence on air humidity when used with the LED devices. A PTFE-based integrating sphere was found to be unaffected by humidity and qualified to provide the stability of the monitoring source at the required level of 0.1 %.

Spectral radiance calibrations



To enable spectral radiance calibrations of sun photometers designed for sky radiance measurements, two approaches have been realised. First, VSL adapted its setup for spectral radiance responsivity measurements of two Prede POM filter radiometers from the SKYNET Europe provided by UV and CNR. The setup is based on a ns-pulsed OPO (EKSPLA NT242). The beam from the laser system is coupled into an integrating sphere. One port of the sphere has a monitor photodiode. The large output port of the sphere is used to measure the sun photometers (Figure 3). Measurements are made relative to a calibrated reference detector placed in front of the output port after the sun photometer measurements.

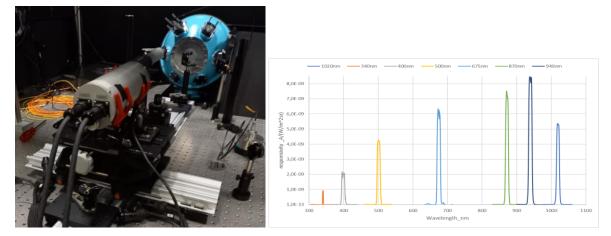


FIGURE 3. POM sun photometer installed in front of the output port of an integrating sphere at VSL and measured spectral radiance responsivities of POM_CNR. As radiation source, a ns-pulsed OPO system is used.

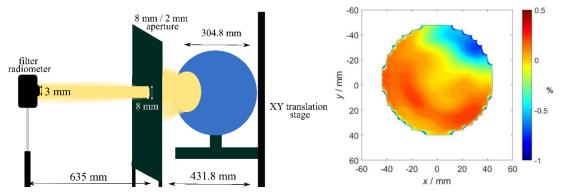


FIGURE 4. Setup at Aalto for measuring spatial homogeneity of the integrating sphere source and scan results at 500 nm.

In a second approach, an integrating sphere source was purchased by ULILLE for spectral radiance calibrations of sun photometers from AERONET Europe network. The source was characterised by Aalto in terms of the radiance homogeneity of the output port (Figure 4). Afterwards, the spectral radiance of the integrating sphere source was measured by PTB (Figure 5). ULILLE measured the radiance responsivity of five CIMEL sunphotometers during the field campaign organised at Izana, in September 2022.

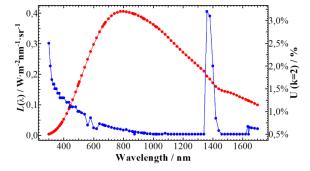


FIGURE 5. Spectral radiance of the source calibrated by PTB (red curve, left axis) and the corresponding calibration uncertainty (blue curve, right axis).



To enable spectral radiance calibrations of sun photometers at network calibration centers, a narrowband tuneable portable source was developed by CMI (Figure 6). The bandwidth of the source is 0.2 nm. The wavelength can be set with an uncertainty of 0.5 nm and tuned in the spectral range from 340 nm to 1640 nm. The source was used to measure the bandpass function of Cimel radiometers of CNRS.

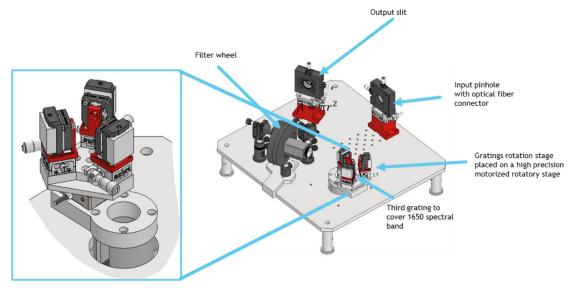


FIGURE 6. Design of a narrowband tuneable portable source.

Field-of-view properties

The field of view is one of the most important characteristics that must be known for a sun photometer. Methods for laboratory and in-field determination of angular response functions (ARF) of radiometers measuring direct solar spectral irradiance and sky radiance were developed by Aalto, SFI Davos, CNRS, UVa, CNR, and UV. Aalto and SFI Davos developed laboratory setups for ARF measurements. Aalto measured the ARF of the POM instrument by CNR and one PFR. SFI Davos characterised POM photometers of UV and CNR, a PFR and several Cimel radiometers (Figure 7). The data were used by CNRS, CNR, UVa and UV to develop the methodology for in-field measurements of the ARF of the instruments using the sun as a source. The ARF data of a reference Cimel sun photometer of CNRS was used to validate radiance and irradiance calibrations at PTB.

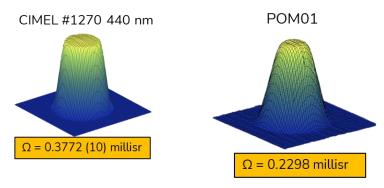


FIGURE 7. ARFs of two instruments measured by SFI Davos.

Linearity and temperature coefficients

The final aspects of instrument characterisations required to meet Objective 1 were the characterisation of the instruments in terms of radiometric linearity and temperature coefficients. The linearity checks were part of the instrument calibrations at PTB and VSL. The temperature characterisations were carried out at PTB for a CE318T radiometer from UVa. The instrument was characterised using a broadband source and a climate chamber over the entire temperature range from -30 °C to +50 °C. In addition, the instrument was heated and measured at two temperatures in the TULIP setup (Figure 8). The POM instrument of the CNR has also been measured in a climate chamber.



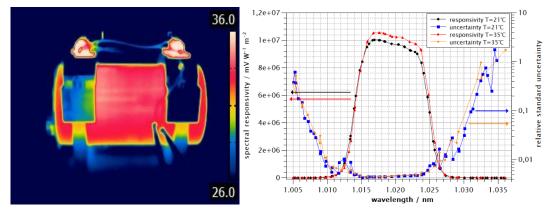


FIGURE 8. Temperature picture of the heated CE318T radiometer and the spectral responsivity of the Si 1020 nm channel with the respective relative standard uncertainties at 21 °C and 35 °C.

Conclusions

Thanks to the fruitful cooperation of the partners, Objective 1 was achieved. Various aspects of the calibration and characterisation of filter radiometers from AERONET-Europe, GAF-PFR and SKYNET-Europe networks with metrological traceability to the SI have been addressed and resolved. In summary, the main outputs of the activities include:

- Setup and procedures for spectral irradiance responsivity calibrations of filter radiometers at PTB in the spectral range from 310 nm to 1700 nm with an uncertainty of 0.1 % to 1 %.
- LED-based monitoring source for observing spectral responsivity changes of AERONET filter radiometers at the level of 0.1 %.
- Setup, transfer standard and procedures for spectral radiance responsivity calibration of sun photometers at VSL and ULILLE.
- ULILLE measured the radiance responsivity of 5 CIMEL sunphotometers.
- Setup for homogeneity characterisations of radiance sources at Aalto.
- Narrowband tuneable portable source for bandpass function measurements of sun photometers.
- Setups for field-of-view measurements at Aalto and SFI Davos. Procedure for in field measurements of this
 characteristics. Data on typical ARFs of different instruments from the AERONET-Europe, GAF-PFR and
 SKYNET-Europe networks.
- Data on temperature dependence of AERONET and SKYNET radiometers.
- 4.2 To validate the methods for zero airmass extrapolation by means of traceable groundbased solar spectral irradiance measurements and comparison to satellite-based solar extra-terrestrial spectra. Due to the growing importance of lunar irradiance measurements to retrieve AOD during night-time conditions (e.g. polar winter), validations of lunar irradiance phase models will also be addressed.

The crucial element in the determination of the atmospheric transmission and thereby of the aerosol optical depth, is the knowledge of the solar (or lunar) irradiance at the top of the atmosphere (ToA). The established methodology for obtaining the top of the atmosphere spectral irradiances is through in-situ calibrations of the reference radiometers based on zero airmass extrapolations (also called Langley-plot procedure) at pristine high altitude sites (Shaw, 1983; Toledano et al., 2018). Then, assuming stability of the radiometers, these are relocated to their respective calibration sites (for example PMOD/WRC, Davos, Switzerland for GAWPFR, or Observatoire de Haute-Provence, France, Valladolid and Izaña, Spain, in the case of AERONET-Europe) to transfer their ToA irradiance values to the network radiometers. This approach is currently the most accurate method for the determination of atmospheric transmission, since only relative measurements of the same instrument are required to obtain the ToA and the surface irradiances. The corresponding measurement uncertainty of the atmospheric transmission can be quantified by the variability of the retrieved ToA solar spectrum on subsequent half-days. As discussed by Toledano et al. (2018), the standard error of the mean of



the retrieved ToA irradiances can be significantly less than 0.5 %, as also shown later in this study. However, there are several disadvantages to this methodology:

- The Langley procedure to retrieve the ToA irradiances requires stable atmospheric conditions with constant AOD during at least one half day to extrapolate the irradiance measurements to zero airmass. The procedure assumes that the atmosphere varies randomly during the calibration period and a statistical approach is applied to retrieve a representative ToA irradiance from a set of measurements (Toledano et al., 2018). Systematic atmospheric variations (for example in the short ultraviolet due to ozone related photochemical effects), would go unnoticed and potentially falsify the results. Furthermore, there is a trade-off between the length of the calibration period and the observed degradation of the instrument which is difficult to quantify and varies for different instruments.
- The zero airmass extrapolation fails in spectral regions with saturated trace gas absorptions, as for example in the water vapour regions around 930 nm.
- Metrological traceability of the reference radiometers is lost after their relocation from the high altitude site to their respective nominal operation sites. The calibration validity is instead assessed through instrument redundancy, and assumptions about instrument stability and degradation.
- Since the irradiance measurements of the radiometers are not traceable to the SI, the solar irradiance measurements themselves cannot be used for quality control or comparisons between instruments.

Since the 1970's, stratospheric balloon, rocket and then satellite based experiments have been measuring the top-of-atmosphere spectral solar irradiance with negligible atmospheric absorption. In principle, these ToA solar spectra, given in SI units, typically in Wm⁻²nm⁻¹, therefore provide directly the necessary ToA irradiances needed to retrieve the atmospheric transmission from calibrated ground-based solar irradiance measurements. However uncertainties in the ToA solar spectra and of the ground- based spectral measurements have so far been too large to achieve the desired uncertainties in atmospheric transmission and thus AOD, when compared to the self-consistent approach based on the zero airmass extrapolation for each instrument.

The situation has considerably improved with the advent of fully SI-traceable characterised and calibrated satellite experiments such as TSIS-1 with a combined standard uncertainty of less than 0.25 % (Richard et al., 2020). Furthermore, a hybrid high spectral resolution solar spectrum based on TSIS-1 measurements has been constructed to provide ToA solar spectra with standard uncertainties between 0.3 % and 1.3 % over the spectral range from 202 nm to 2370 nm (Coddington et al., 2021). As discussed in Coddington et al. (2021), this hybrid solar spectrum TSIS-1 HSRS agreed to within 0.5 % with the ToA solar spectrum QASUMEFTS obtained from zero airmass extrapolations from SI-traceable ground-based solar irradiance measurements with the QASUME spectroradiometer in the spectral range 300 nm to 500 nm (Gröbner et al., 2017). Furthermore, Kouremeti et al. (2022) have shown that AOD retrieved from SI-traceable solar irradiance measurements with a precision filter radiometer at the spectral channels 368 nm, 412 nm, 500 nm, and 862 nm, agreed to within 0.01 when compared with the AOD derived using a Langley-plot based calibration of the same instrument.

In this project, we have obtained SI-traceable spectral solar irradiance measurements performed with solar spectroradiometers at the high altitude observatory in Izaña (IZO), Tenerife, during a three-week field campaign. The ToA solar spectra derived from zero airmass extrapolations were compared with the TSIS-1 HSRS solar reference spectrum over the range 300 nm to 2100 nm. IZO is a high mountain station at an elevation of 2373 m above sea level (a.s.l.) above a strong subtropical temperature inversion layer, which acts as a natural barrier for local pollution and low-level clouds. The site is a primary calibration site for instruments performing zero airmass extrapolations due to its stable atmospheric conditions during most of the year. Based on a long-term climatology of the site, the period of the campaign was selected so as to offer the highest probability for clear skies, stable total column ozone values and a minor probability of Saharan dust intrusions.

The following radiometers and spectroradiometers were deployed during this field campaign:

QASUME spectroradiometer

QASUME consists of a scanning double monochromator with a focal length of 150 mm and two 2400 lines/mm gratings resulting in a full width at half maximum (FWHM) of 0.86 nm. The whole system resides in a temperature controlled enclosure to allow outdoor operation under varying ambient conditions. The solar radiation is collected with a temperature stabilised diffuser connected via an optical fiber to the entrance slit of the monochromator. A portable lamp monitoring system allows for the calibration of the whole system while being deployed in the field. A collimator tube with a full opening angle of 2.5° is mounted on an optical tracker



to which the diffuser head can be fitted, allowing the measurement of direct solar spectral irradiance. The expanded relative uncertainty of spectral solar irradiance measurements is 1.4 % (k=2, representing a coverage probability of 95 % assuming a normal probability distribution). QASUME was calibrated every day using a portable lamp monitoring system with a set of three 250 W tungsten-halogen lamps in order to verify its stability and confirm its traceability to the SI. The calibrations were performed daily, and varied by less than ± 0.5 % over the course of the campaign.

QASUME-IR spectroradiometer

The QASUME-IR spectroradiometer consists of a single monochromator with focal length 300 mm. Two gratings are used to cover the extended spectral range from 550 nm to 900 nm (1200 lines/mm), and 900 nm to 1700 nm (830 lines/mm). The corresponding spectral resolution for these two regions is 1.6 nm and 2.4 nm (FWHM). The entrance optic consists of an integrating sphere, coupled to the entrance port of the spectroradiometer with an optical quartz fiber. The integrating sphere is connected to a collimator with the same dimensions as used for QASUME to allow measurements of the direct solar irradiance. Two temperature stabilised photodiodes (Silicon-based until 900 nm and InGaAs for the second spectral range) measure the dispersed radiation at the two output slits of the monochromator. The instrument was calibrated daily using the same portable system as used for QASUME, resulting in a similar relative expanded uncertainty of 1.4 % (k=2) apart from the spectral bands strongly affected by atmospheric humidity.

The responsivity of QASUME-IR was very stable throughout the campaign, with a variability of less than 0.4 % in the spectral range 550 nm to 1300 nm. Between 1300 nm and 1500 nm, the measured responsivity varied by 1.5 %, due to ambient changes in relative humidity that absorbed some of the radiation emitted by the transfer standard lamp in the portable monitoring system, before reaching the entrance optic of the spectroradiometer.

Precision Solar Spectroradiometer

The Precision Solar Spectroradiometer (PSR) is based on a temperature stabilised grating spectroradiometer with a 1024 pixel Hamamatsu diode-array detector, operated in a temperature controlled hermetically sealed nitrogen flushed enclosure. The spectroradiometer measures the solar spectrum in the 316 nm to 1030 nm spectral range with a FWHM of between 1.5 nm and 4 nm. The relative expanded uncertainty (k=2) is 1.8 % for direct normal solar spectral irradiance measurements in the central wavelength range between 400 nm and 900 nm, with slightly increasing uncertainties at either end of the spectrum.

BTS spectroradiometers

The BTS is a commercially available system composed of two array-spectroradiometers made by the company Gigahertz Optik GmbH. The spectral range from 300 nm to 1050 nm is covered with a 2048 pixel Si BTS2048-VL-TEC-WP with a nominal spectral resolution (FWHM) of 2.5 nm, while the spectral range from 950 nm to 2150 nm is measured with a BTS2048-IR-WP with a nominal spectral resolution of 8 nm (FWHM) with 512 pixel and an extended InGaAs detector.

Each spectroradiometer has a collimator to measure only direct solar irradiance with a diffusor entrance optic. The BTS2048-IR-WP has been calibrated by PTB a half year before the campaign with an estimated relative expanded measurement uncertainty of 3.3 % (k=2) up to 1600 nm and 4.3 % (k=2) above 1600 nm. The BTS2048-VL-TEC-WP was calibrated in the ISO 17025 calibration laboratory of Gigahertz-Optik, which is traceable to PTB and has been validated by PTB who confirmed the calibration uncertainty. An estimated expanded measurement uncertainty (k=2) of 3.5 % from 300 nm to 330 nm, 1.9 % (k=2) from 330 nm to 450 nm and 1.8 % (k=2) for the remaining spectral range was achieved. The calibrations were checked on-site using a commercially available mobile transfer standard based on a 250 W tungsten lamp called BN-LHSI-WP before and after the measurement campaign. The BTS2048-VL-TEC- WP calibration did not change significantly and therefore the laboratory calibration was used during the whole campaign.

Precision filter radiometer PFR N01

A precision filter radiometer, PFR N01, was operated during the measurement campaign. The four spectral channels of this radiometer were fully characterised and calibrated with respect to the SI at the tuneable laser facility of PTB in January 2021 (Kouremeti et al., 2022).

Solar-pointing Fourier Transform Infrared Spectrometer

A solar-pointing Fourier Transform Infrared Spectrometer (FTIR) was deployed at IZO during the campaign. This instrument was a scanning Michelson design supplied by Bruker Corp. (model 125M) coupled to a solar tracking system (also supplied by Bruker). It was used to provide high-resolution (0.01 cm⁻¹) measurements



of the solar spectrum between 500 nm and 5500 nm. Langley analysis based on Kiedron and Michalsky (2016) was used to separate the top-of-atmosphere and atmospheric contributions to the measurements across the near-infrared region between 950 nm to 2100 nm.

The high-resolution measurements provided by the FTIR instrument were used to test the validity of the gas absorption assumptions in the near infrared region. The figure below shows an example atmospheric optical depth between 1500 nm and 1700 nm, as determined from the gradient of the Langley fit to solar FTIR data taken on the 22nd September 2022. In addition to the high resolution data (black line), three spectrally broadened results are shown with the FWHM values of 2.4 nm, 4 nm and 6 nm, corresponding to the spectral resolution of the QASUME-IR and BTS spectroradiometers respectively. The baseline value is determined as the mean value of the high resolution data points within the respective bandwidths that are unaffected by gas absorption lines.

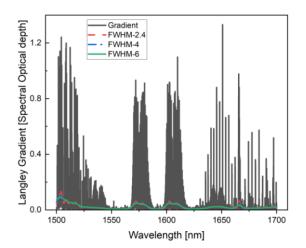


FIGURE 9. Example of high resolution atmospheric optical depths determined from Langley analysis on 22nd September 2022 (black line). Broadened results for spectral resolutions of 2.4 nm, 4 nm and 6 nm are also shown (red, blue and green lines respectively).

Validation of TSIS-1 HSRS solar spectrum

The atmospheric transmission is obtained from direct solar irradiance measurements using the Beer-Lambert law,

$$I(\lambda) = I^0(\lambda)e^{-\tau m} \tag{1}$$

Where *I* is the solar irradiance at the Earth surface, P the top-of-the-atmosphere (TOA) solar irradiance, *t* the optical depth of the atmosphere, and *m* the airmass.

The zero airmass extrapolation procedure as described in Gröbner et al. (2017) is used, by performing a linear regression of the logarithm of the spectral solar irradiance measurements with respect to airmass to retrieve the spectral solar irradiance value at airmass 0, representing the ToA solar irradiance at wavelength λ . This procedure assumes that during the duration of these measurements the atmospheric transmission remains constant, and that any remaining small atmospheric variations are uncorrelated and therefore random from one day to the next.

From the available measurements in the period 6 to 22 September 2022, between 8 and 15 half-days were selected from the data set of each spectroradiometer, to retrieve ToA solar spectra for each half day period by zero airmass extrapolation. Subsequently, the mean ToA solar spectrum and the corresponding standard error were computed from these retrievals for each spectroradiometer. Figure 10 shows the extrapolated ToA solar spectrum from TSIS-1 HSRS, convolved with a nominal 1 nm FWHM triangular line spread function, is also shown in the upper part of the figure.



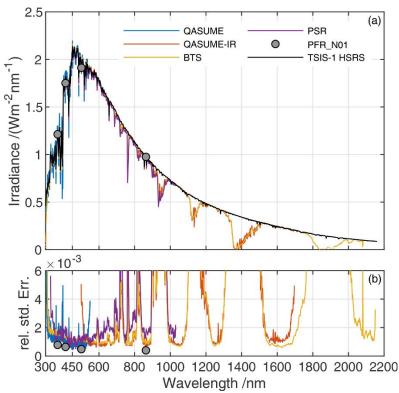


FIGURE 10. Top: Top of atmosphere solar spectra derived from zero airmass extrapolations by QASUME (blue), QASUME-IR (red), BTS (yellow), and PSR (violet) and TSIS-1 HSRS (green) convolved with a 1 nm FWHM triangular line spread function. The grey circles represent the solar irradiances measured with PFR N01. Bottom: Relative standard error of the mean of the ToA solar spectra shown in the top figure using the same colour scale.

The ratio between the extrapolated ToA solar spectra is shown in Figure 11. A 10 nm moving average is applied to the ratios to remove some of the high frequency spectral variability. The agreement between the extrapolated ToA solar spectra and TSIS-1 HSRS in the spectral regions unaffected by strong gas absorptions is mostly within 1 %. The slightly larger deviations seen with the array spectroradiometers PSR and BTS in the ultraviolet spectral region below 400 nm, likely come from unaccounted wavelength errors and line spread function related artifacts when convolving the high spectral resolution spectrum TSIS-1 HSRS with the spectrally varying line spread functions of the PSR and BTS spectroradiometers. The consistency between TSIS-1 HSRS and the ToA solar irradiances retrieved with PFR N01 is also excellent, with relative differences below 1 %.



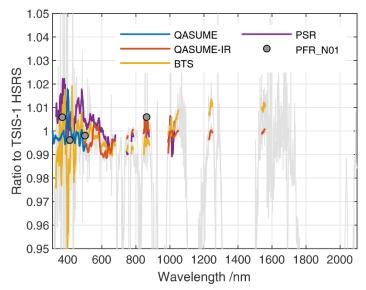


FIGURE 11. Spectral ratio of the zero airmass extrapolated ToA solar spectra from QASUME (blue), QASUME-IR (read), PSR (violet), and BTS (yellow) relative to the TSIS-1 HSRS solar spectrum convolved with the respective line spread functions of the spectroradiometers. The coloured lines are a 10 nm moving average of the spectral ratios in spectral regions with no or only weak atmospheric trace gas absorption, while the grey line represents the full spectral ratio of BTS to TSIS-1 HSRS. The grey circles represent the ratios of the spectral solar irradiances of PFR N01 with TSIS-1 HSRS convolved with the spectral filter transmissions of PFR N01.

Conclusions

The Top of Atmosphere solar irradiance spectrum was retrieved from the solar irradiance measurements using zero airmass extrapolation during cloudfree conditions and compared to the TSIS-1 HSRS solar spectrum. The agreement between the extrapolated ToA solar spectra and TSIS-1 HSRS was excellent and well within the combined uncertainties of the spectroradiometers over the full spectral range. Especially the measurements of QASUME/QASUME-IR between 300 nm and 1700 nm were within 1 % (peak to peak) of the TSIS-1 HSRS solar irradiance spectrum in the spectral regions not affected by trace gas absorptions. Based on this comparison, the measurements of QASUME could be used to reduce the stated relative standard uncertainty of the TSIS-1 HSRS ToA solar spectrum in the spectral range 308 nm to 400 nm from 1.3 % to 0.8 %. The objective was achieved.

4.3 To develop a comprehensive uncertainty budget for aerosol optical properties, such as aerosol optical depth, aerosol size distribution, and aerosol single scatter albedo, retrieved from remote sensing-based measurements of direct and scattered solar radiation, enabling its inclusion in the corresponding data archives of the aerosol monitoring networks, with the relevant calibration and traceability information.

Rational. The accurate characterisation of aerosol optical properties and surface properties is crucial for understanding atmospheric processes and their impact on climate. Remote sensing techniques have emerged as valuable tools for retrieving aerosol and surface properties from satellite, aircraft, and ground-based measurements. However, the estimation of uncertainties associated with these retrievals remains a challenge. In the framework of this project, we developed a methodology to estimate the dynamic errors for the aerosol optical properties retrieved from remote sensing ground-based measurements by diverse photometers and radiometers involved in the project.

The generation of the dynamic error estimates were realised using the Generalized Retrieval of Atmosphere and Surface Properties (GRASP) algorithm (Dubovik et al. 2021), which is based on the concept of statistical optimisation fitting of observations following the Multi-term LSM strategy specifically designed for retrieving aerosol and surface properties. The GRASP algorithm has the capability of providing dynamic error estimates, taking into account the effect of both, random and systematic measurement uncertainties propagations (Herrera et al., 2022). Furthermore, the study included the generation of full covariance matrices. It was demonstrated that analysing correlation patterns is very useful for optimising observation schemes and retrieval setups. Specifically, the identification of significant correlations between different retrieved parameters allows the evaluation of the needs and efficiency of additional ancillary data or a priori data desirable for improving the efficiency of retrieval and decreasing the retrieval accuracy.



In order to evaluate the accuracy and reliability of the realised developments a series of the synthetic sensitivity tests were conducted using simulated measurements perturbed by random and systematic errors. The analysis was realised using AERONET-like observations. In the frame of these tests, the synthetic proxy observations perturbed by 300 random-noise-generated realisations were inverted using the GRASP algorithm. Then, the retrieved parameters were compared to those used for the generation of the synthetic data, and the obtained error estimates were compared with actual deviations in the retrieved parameters from assumed values. This analysis was realised for the synthetic observations for three different aerosol types, including biomass burning, urban, dust, and mixtures. These different aerosol types were used to evaluate the performance of the GRASP algorithm in retrieving specific aerosol properties. Also, the main observed tendencies in the error dynamic agree with known retrieval experience. For example, the main accuracy limitations for retrievals of all aerosol types relate to the situations with low optical depth.

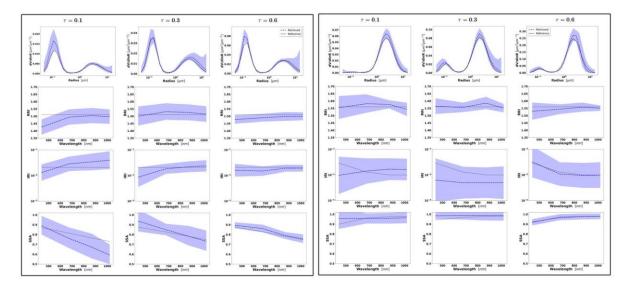


FIGURE 12. Aerosol properties retrieved from simulated sun/sky photometer data with assumed bias in AOD- and radiance-simulated data for biomass burning (left) and dust (right) aerosol for AOD(440)=0.1, 0.3 and 0.6 (left to right). Retrievals after adding positive bias +0.01 in AOD and +5% in radiances are represented in both cases. The solid lines are the simulated properties (SD, RRI, IRI and SSA), and the dashed lines are the retrieved parameters. The shaded area indicates the systematic error estimated by the GRASP algorithm.

The results of the different tests showed that the complete set of aerosol parameters for each aerosol component can be robustly derived with acceptable accuracy in practically all considered situations. Moreover, in the case of aerosol mixture, the algorithm successfully retrieves total aerosol properties even in cases where individual aerosol components are challenging to characterise. For example, Figure 12 illustrates the retrieval of biomass burning and dust for the three different aerosol loads (0.1, 0.3 and 0.6) when the total error estimate includes both random and systematic components. The retrieved properties are the size distribution, the real and imaginary refractive index and the single scattering albedo at the different wavelengths.

As a part of this objective, the utilisation of GRASP algorithm for deriving detailed aerosol properties and estimations of their errors was demonstrated for the observations from different instruments including Cimel, Prede, PFR, PSR, QASUME radiometers and photometers that are part of the database developed in the framework of this project by efforts of the UVa and GRASP SAS. The GRASP retrievals and the error estimates of the different aerosol properties were shown to be fully adequate in a comparative analysis with the aerosol products available from AERONET and SKYNET retrievals as can be observed in Figure 13.



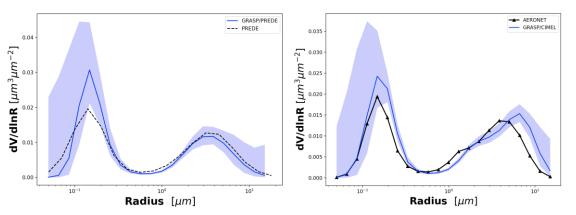


FIGURE 13. Comparison of retrieved size distribution by GRASP vs SKYNET (left) and GRASP vs AERONET (right). The shaded area indicates the systematic error estimated by the GRASP algorithm. The retrievals are based on the observations at Roma on September 12th, 2021.

In addition to the evaluation of the error bar estimates and the effects of systematic errors, the correlation structures of the error covariance matrices were analysed and illustrated. For example, Figure 14 shows the correlation matrices for AERONET-like conventional retrievals in two particular cases: observations of biomass burning (mostly spherical particles) and dust (mostly non-spherical particles) aerosols.

The matrices show the correlation structure between all retrieved parameters: 22 parameters (22x22) that represent the SD, followed by two blocks of 4x4 related to the RRI and IRI for four wavelengths (440 to 1020 nm) and a 1x1 block that is related to the sphericity fraction. The colors indicate the values of the correlation

coefficients $\rho_{ij} = \langle \Delta a_i \Delta a_j \rangle / \sqrt{\langle \Delta a_i^2 \rangle \langle \Delta a_j^2 \rangle}$, where the red color denotes positive correlations, and the blue color indicates negative correlations. The density of the colors indicates the values of the correlation coefficients changing from zero (the white color) to dense red or blue colors corresponding to values of 1 and -1 accordingly. For example, the underestimation of RRI accompanied by an overestimation in the size distribution of very fine particles has been widely discussed in studies by Dubovik et al. (2000, 2002a,b) and could be seen in the correlation matrix as strong negative correlations between size distribution and real refractive index. It should be noted that the presence of high correlations is an indication that adding information about one of the correlated parameters should improve the retrieval, not only for the constrained parameter itself but also for the parameters that are strongly correlated with this parameter.

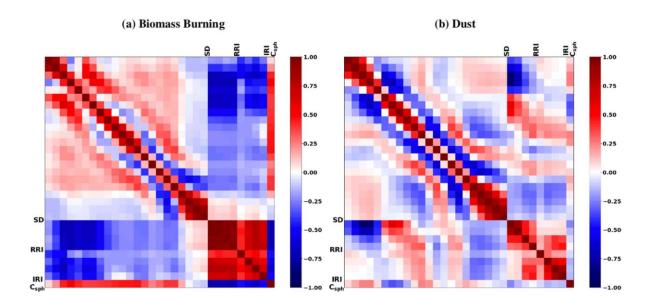




FIGURE 14. Correlation matrices of the estimated errors for aerosol retrieval from sun/sky radiometer observations (a) for biomass burning aerosols and (b) for desert dust using the GRASP algorithm. The values close to 1 or -1 mean stronger correlations between the properties, positive or negative, respectively.

Furthermore, having studied the correlation matrices allows us to obtain additional information on the synergy between different instruments and/or measurements used. For example, as it is known, the addition of polarimetric observations to the traditional set of AERONET observations results in a clear improvement in the retrieval of RRI and the size distribution of very fine particles (Li et al., 2009; Fedarenka et al., 2016). Indeed, the degree of linear polarisation is known to be very sensitive to the amount and especially the RRI of fine particles (Dubovik et al., 2006). All these tendencies and others could be seen in the correlation matrices and could help to improve the retrieval set-ups.

All the results, tests and description of conducted study can be found in the paper by Herrera et al. (2022), which is also a deliverable of this project. Moreover, an automatisation tool for the inversion of different measurements has also been realised, which allows a customised inversion for each instrument (Cimel, Prede, PFR, PSR, QASUME). For these inversions, the retrieved properties and their error estimates are provided as available products.

Conclusions

The uncertainties in aerosol optical properties have direct consequences in the estimation of the aerosol radiative forcing estimations. This was investigated by partner UoR, which performed radiative transfer calculations to determine the sensitivity of global direct aerosol radiative forcing to uncertainties in aerosol optical properties. The study has been submitted for publication to a peer-reviewed journal (Elsey et al., 2023). In this work a Monte-Carlo framework was built to calculate the impact of uncertainties in aerosol optical depth (AOD), single scattering albedo (SSA) and asymmetry parameter on the uncertainty in shortwave DARE and DARF. This framework used the results of over 2.3 million radiative transfer simulations to calculate global clear-sky DARE and DARF based on a range of aerosol optical property uncertainties, representative of existing and future global observing systems. The one-sigma uncertainty varied between ±0.23 to ±1.91 Wm-2 (5 and 42 %) for the top of atmosphere (TOA) clear-sky DARE and between ±0.08 to ±0.47 Wm-2 (9 and 52 %) for the TOA DARF. At the TOA, AOD uncertainty was the main contributor to overall uncertainty, except over bright surfaces where SSA uncertainty contributed most. Regionally varying uncertainties were applied to represent current measurement uncertainties, finding that aerosol optical property uncertainties represented 24 % of TOA DARE and DARF. Reducing regionally varying optical property uncertainties by a factor of two would reduce their contributions to TOA DARE and DARF uncertainty proportionally. Scaling to all-sky conditions, aerosol optical property uncertainty contributed to about 25 % total uncertainty in TOA, all-sky DARE and DARF. Compared to previous studies which considered uncertainties in non-aerosol variables, this study suggests that the aerosol optical property uncertainty accounts for a third to a half of total uncertainty. Objective 3 was achieved.

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5 Impact

The winter school SORBETTO-3 was held from 6 to 10 February 2023 at the ESA ESRIN site in Frascati, Italy. Contributions in the form of oral presentations were given by 8 MAPP partners. 30 participants were selected from the submitted applications.

7 peer-reviewed publications on different aspects of MAPP have been published and 7 more were submitted to peer-reviewed journals and are currently under review. MAPP related activities were disseminated through 16 presentations at standardisation & regulatory body meetings and 31 presentations at conferences and workshops.

Three private companies manufacturing sunphotometers and spectroradiometers for measuring solar radiation have joined the project as collaborators. Presentations describing the objectives of the project were given at steering committee meetings of ACTRIS in order to highlight the benefits that the project will bring to the aerosol remote sensing community in Europe. Two new collaborators have joined the project, increasing significantly the end-user up-take and stakeholder involvement (NASA, hosting the global AERONET network, and ESA participating at the field campaign at Izana in September 2022 for the lunar measurements). The collaborators were actively participating in the project and benefitting from the calibration procedure developed within the project.

Training was performed by the project partners on laboratory calibrations and solar measurements. A guest researcher from SFI DAVOS completed a one-month training at PTB in November 2021, focused on the characterisation and calibration of solar and lunar filter radiometers and one spectroradiometer. A training workshop for partners and project collaborators was held during the third project meeting at PTB.

Impact on industrial and other user communities

Industry has benefitted from the developments in novel radiometric technologies developed in this project, which are specifically aimed at promoting the use of emerging technologies for outdoor measurements of solar spectral irradiances covering the UV to near-infrared spectrum. The improved data analysis products and uncertainty estimates developed in this project have an impact on European small and medium-sized enterprises such as GRASP, which are now providing enhanced services tailored to the needs of their customers.

The users of network data such as climate scientists benefit from the comprehensive uncertainty analysis as they will utilise not only the aerosol measurement data but also an objective assessment of the reliability of each data point, which is crucial for producing reliable climate assessments and long-term trend analyses. Furthermore, as these data serve as fiducial reference for various satellite platforms and satellite dissemination platforms, their traceability and reliability have substantially enhanced their importance and usefulness. As an example, recent (Sentinel 5P) and planned (Sentinel 5, EarthCare) missions of the European Space Agency will benefit from these results.

This project supports many users involved with the European Research Infrastructure for the observation of Aerosols, Clouds and Trace gases (ACTRIS) by providing new traceability routes and therefore improved products and services.

Impact on the metrology and scientific communities

The NMIs and DIs have extended their services and collaboration with the scientific community by offering not only access to their laboratory infrastructure as in-kind contribution through the project MAPP, but also by providing essential capacity development and collective activities aimed at creating a common decentralised European platform for radiometric calibrations of atmospheric remote sensing instruments, which is the aim of the European Metrology Network for Climate and Ocean Observation. Specifically, the new procedures developed for radiance and irradiance calibrations of the network instruments have taken into account specific user community needs (wide temperature range, irradiance and radiance level changes over several orders of magnitude) which are usually not offered by metrological institutes, thereby enlarging their portfolio and developing new capabilities to serve the end user needs.

The project has standardised measurements of atmospheric aerosol optical properties from sunphotometers and solar spectroradiometers by establishing and validating consistent calibration and characterisation schemes, based on a mix of outdoor and laboratory-based calibration procedures. The project developed the initial tools and methods in synergy with the end-user laboratories of each aerosol monitoring network and established long-term collaborations needed to effectively implement these new techniques in a routine network calibration routine. The mid to long-term strategy is to support a ground-breaking change in the way



calibrations will be performed and to transfer best laboratory practices from the metrological community to the end-user laboratories.

Impact on relevant standards

The project participated in ACTRIS steering meetings and in particular CARS (ACTRIS Centre for Aerosol Remote Sensing) which are responsible for the calibration of radiometers of the European Branch of AERONET. The MAPP results were discussed at these meetings and will feed into a new traceability scheme for the ACTRIS/AERONET network which is under discussion.

Similarly, the Scientific Advisory Group on Aerosols, as well as the Expert Team of Atmospheric Composition Measurement Quality (ET ACMQ) were regularly informed of the progress of MAPP. Possible modifications to the Aerosol Chapter of the former CIMO Guide to Instruments and Methods of Observations to include the new developments of MAPP will be discussed for the next revision lifecyle of this document.

The outputs of this project play a major role in revising existing and setting new standards for the aerosol column-averaged and aerosol remote-sensing components of the Earth Observation community at large. While these changes will occur in a mid-term perspective (5-10 years), their impact will be significant on demonstrating traceability of aerosol remote sensing at a global scale.

Longer-term economic, social and environmental impacts

Some of the major results of this project are advanced calibration services, upgraded data analysis tools, and novel measurement instruments. Several European small and medium-sized businesses were involved in these activities, which thereby have gained a competitive advantage in this specialised global market.

The project has provided a framework for assessing the radiative impact of one of the most uncertain and variable drivers of the climate system through improved measurements and associated modelling. This will support societies in adapting to a changing climate and choosing climate mitigation strategies. Choosing the most effective climate mitigation strategy provides the best long-term benefits to citizens at a minimum cost.

Various geoengineering proposals and ideas aiming to reduce the direct radiative forcing (e.g. through injecting sulphate aerosols in the stratosphere) have been proposed in the past decade, based on the rationale that even aggressive reductions in net emissions of greenhouse gases will be insufficient to limit global climate risks. The success of such proposals depends on the proposed amount of solar dimming that is directly proportional to the aerosol load in the atmosphere. Thus, measurement accuracy of columnar aerosol properties is the key factor to address if such model-based scenarios could be considered realistic. The project results support these investigations, as aerosol measurement uncertainty will be a crucial factor in evaluating issues such the aerosol lifespan, regional aerosol concentration distribution, aerosol transport and others.

In addition, the project will have a long-term impact on public awareness bodies and private and public sectors that are dealing with a number of issues related to health; indirect effects such as aerosol related health effects (additional lower atmosphere aerosol pollution), ozone depletion and UV radiation changes, regional warming and effects on the hydrologic cycle and regional weather changes.

6 List of publications

- Angular responsivity of ground and space-based direct solar irradiance radiometers. Hülsen, G., Gröbner, J., Pfiffner, D., Gyo, M., Kouremeti, N. and Föller, J. 2022 Journal of Physics: Conference Series. 2149 012001. <u>https://doi.org/10.1088/1742-6596/2149/1/012001</u>
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