

Aerosol absorption measurements at the ALOMAR subarctic station

Medidas de absorción debida a los aerosoles en la estación subártica ALOMAR

Elena Montilla-Rosero^(1,*), Edith Rodríguez^(1,2), Sandra Mogo^(1,3,S), Victoria Cachorro^(1,S),
Rubén Rodrigo⁽¹⁾, Ángel de Frutos^(1,S)

1. Atmospheric Optics Group, University of Valladolid, Prado de la Magdalena s/n, 47011, Valladolid, Spain.
2. Finnish Meteorological Institute, Erik Palménin Aukio, P.O Box 503, Helsinki, Finland.
3. Department of Physics, University of Beira Interior, 6201-001, Covilhã, Portugal.

*) Email: montilla@goa.uva.es

S: miembro de SEDOPTICA / SEDOPTICA member

Recibido / Received: 16/11/2010. Versión revisada / revised versión: 08/02/2011. Aceptado / Accepted: 09/02/2011

ABSTRACT:

The global study of atmospheric aerosol is one of the key factors in regards to climate change and those effects. Over the last 4 years a strong research work on aerosols properties characterization have been carried out into the Atmospheric Optics Group of the University of Valladolid (GOA-UVa) in Spain. We present the results about aerosol absorption coefficient measurements with two different techniques: the “integrating sphere photometer” and the particle soot absorption photometer (PSAP, Radiance Research). This data was acquired into the summer campaign 2008, made at north of Norway, like a result of the participation of GOA in the POLARCAT project, lead by the Norwegian Institute for Air Research, and included in the Fourth International Polar Year. Both methods are filter based and provide *in-situ* measurements that could be combined with optical column measurements for a better characterization of local aerosol. It contributes especially to the investigation of pollution events and to establishment the effects of the population over any local aerosol climatology. Based on this research background and the affordable and reliable instrumentation described in this work, the research can continue in Colombia in close collaborations with Spanish research groups.

Key words: Aerosol, Light Absorption, Absorption Photometer Method.

RESUMEN:

El estudio de los aerosoles atmosféricos a nivel mundial, es uno de los factores clave para entender el cambio climático y sus efectos. En los últimos 4 años, el Grupo de Óptica Atmosférica de la Universidad de Valladolid (GOA-UVa, España), ha venido realizando un importante trabajo de investigación respecto de la caracterización de las propiedades de los aerosoles. En este trabajo se presentan los resultados de las mediciones del coeficiente de absorción debida a los aerosoles con dos técnicas distintas: el fotómetro de esfera integradora y el fotómetro de absorción PSAP (Radiance Research). Estos datos fueron obtenidos durante la campaña de verano 2008 realizada al norte de Noruega, como resultado de la participación del GOA-UVa en el proyecto POLARCAT, dirigido por el Instituto Noruego para la investigación del aire y que está incluido en el cuarto año Polar Internacional. Los dos métodos utilizados están basados en las medidas sobre filtros y proporcionan medidas *in situ* que pueden ser combinadas con medidas sobre la columna atmosférica, para una mejor caracterización de los aerosoles locales. Este estudio contribuye especialmente a la investigación de los eventos de polución y en el establecimiento de los efectos de la población sobre la climatología local de aerosoles. Teniendo como base estos antecedentes de investigación y, la posibilidad de adquirir la asequible y fiable instrumentación descrita en este trabajo, la investigación puede continuar en Colombia en estrecha colaboración con los grupos de investigación españoles.

Palabras clave: Aerosol, Absorción de Luz, Fotometría de Absorción.

REFERENCES AND LINKS

- [1] IPCC – 2001, *Climate Change 2001: The Scientific Basis*, J. T. Houghton, Y. Ding, D. J. Griggs, M. Noguer, P. J. van der Linden, X. Dai, K. Maskell and C. A. Johnson, Edts., Contribution of Working Group I to the Third Assessment Report of the Intergovernmental Panel on Climate Change, pp. 881 (2001).
- [2] WMO – 2003, *Aerosol Measurement Procedures, Guidelines and Recommendations*, N° 153, World Meteorological Organization, Geneva (2003).
- [3] T. Anderson, R. Charlson, N. Bellouin, O. Boucher, M. Chin, S. Christopher, J. Haywood, Y. Kaufman, S. Kinne, J. Ogren, L. Remer, T. Takemura, D. Tanré, O. Torres, C. Trepte, B. Wielicki, D. Winker, H. Yu., “An “A-Train” strategy for quantifying direct aerosol forcing of climate”, *Bull. Am. Met. Soc.* **86**, 1795-1809 (2005).
- [4] J. Sciare, K. Oikonomou, H. Cachier, N. Mihalopoulos, M. O. Andreae, W. Maenhaut, R. Sarda-Estève, “Aerosol mass closure and reconstruction of the light scattering coefficient over the Eastern Mediterranean Sea during the MINOS campaign”, *Atmos. Chem. Phys.* **5**, 2427-2461 (2005).
- [5] O. Schmid, P. Artaxo, W. P. Arnott, D. Chand, L. V. Gatti, G. P. Frank, A. Hoffer, M. Schnaiter, M. Andreae “Spectral light absorption by ambient aerosols influenced by biomass burning in the Amazon Basin. I: Comparison and field calibration of absorption measurements techniques”, *Atmos. Chem. Phys.* **6**, 3443-3462 (2006).
- [6] A.-C. Engvall, R. Krejci, J. Ström, R. Treffeisen, R. Scheele, O. Hermansen, J. Paatero, “Changes in aerosol properties during spring-summer period in the Arctic troposphere”, *Atmos. Chem. Phys.* **8**, 445-462 (2008).
- [7] S. Mogo, V. E. Cachorro, M. Sorribas, A. de Frutos, R. Fernández, “Measurements of continuous spectra of atmospheric absorption coefficients from UV to NIR via optical method”, *Geophys. Res. Lett.* **32**, L13811 (2005).
- [8] T. C. Bond, T. L. Anderson, D. Campbell, “Calibration and intercomparison of filter-based measurements of visible light absorption by aerosols”, *Aerosol Sci. Tech.* **30**, 582-600 (1999).
- [9] D. Delene, J. A. Ogren, “Variability of aerosol optical properties at four North American surface monitoring sites”, *J. Atmos. Sci.* **59**, 1135-1150 (2002).
- [10] G. Hagler, M. Bergin, E. Smith, J. Dibb, “A summer time series of particulate carbon in the air and snow at Summit, Greenland”, *J. Geophys. Res.* **112**, D21309 (2007).
- [11] R. W. Bergstrom, “Predictions of the spectral absorption and extinction coefficients of an urban air pollution aerosol model”, *Atmos. Environ.* **6**, 247-258 (1972).
- [12] C. Tomasi, V. Vitale, A. Lupi, C. Di Carmine, M. Campanelli, A. Herber, R. Treffeisen, R. S. Stone, E. Andrews, S. Sharma, V. Radionov, W. Von Hoyningen-Huene, K. Stebel, G. H. Hansen, C. L. Myhre, C. Wehrli, V. Aaltonen, H. Lihavainen, A. Virkkula, R. Hillamo, J. Ström, C. Toledano, V. Cachorro, P. Ortiz, A. de Frutos, S. Blindheim, M. Frioud, M. Gausa, T. Zielinski, T. Petelski, T. Yamanouchi, “Aerosols in polar regions: A historical overview based on optical depth and in situ observations”, *J. Geophys. Res.* **112**, D16205 (2007).
- [13] R. W. Bergstrom, P. Pilewskie, P. B. Russell, J. Redemann, T.C. Bond, P. K. Quinn, B. Sierau, “Spectral absorption properties of atmospheric aerosols”, *Atmos. Chem. Phys.* **7**, 5937-5943 (2007).
- [14] R. Draxler, G. Rolph, “Hybrid single-particle Lagrangian integrated trajectory”, Model access via NOAA ARL READY, <http://www.arl.noaa.gov/ready/hysplit4.html>, NOAA Air Resources Laboratory, Silver Spring, MD (2003).

1. Introduction

Aerosols influence the atmospheric energy budget through direct and indirect radiative effects. Direct effects include the scattering and absorption of radiation and the subsequent influence on planetary albedo and the climate system. Indirect effects involve the influence of anthropogenic aerosol on available cloud

condensation nuclei (CCN). Cloud lifetimes and precipitation frequencies can also be affected. Scientific evidence indicates that in regions with high anthropogenic aerosol concentrations, aerosol forcing is of the same magnitude, but opposite in sign to the combined effect of all greenhouse gases [1]. Furthermore, as uncertainties are strong, long-term monitoring

of aerosols properties will be crucial in determining the role that aerosols play in climate, in documenting changes in the regional air quality and in providing a scientific basis for policy decisions regarding control strategies [2].

In atmospheric measurements, one of the main goals is an appropriate characterization of the nature and impact of aerosols and the inclusion of its effects on existing models in order to reduce uncertainties in climate prediction [3]. An important contribution to these uncertainties has been associated with the determination of physical and optical properties of the aerosol particles, *i.e.* the size distribution, the chemical composition and the shape (properties historically related to studies of air pollution and quality air), as well as the light scattering coefficient (σ_s), the light absorption coefficient (σ_a) and the light extinction coefficient ($\sigma_e = \sigma_a + \sigma_s$), the single scattering albedo (SSA), the asymmetry parameter (g) and the aerosol optical depth (AOD), these properties are closely related to the radiative transfer studies.

Several factors difficult the determination of light absorption by atmospheric aerosols, even on local level, in addition to the large space-time variability of the particles, the technical difficulty of measure is included. As long as reliable techniques for *in situ* measurements of light scattering have been available for several decades [4], the light absorption by particles is a more elusive property, because during the absorption process, the photons are transformed in heat and the difficulty for direct measure is greater [5]. The visible light absorption in the atmosphere is predominantly due to aerosol particles, and black carbon is responsible for more than 90% of this absorption (BC, the term used to refer to the carbonaceous absorbing particles).

The Arctic region is an interesting area to make measurements of aerosol properties, due to its high sensitivity to changes introduced by the aerosol and their environmental impact. The study of the microphysical, the optical and the radiative properties of aerosols in this area could be an early warning of global climate change. The interactions among solar radiation, aerosols and clouds increase the radiative

impact of atmospheric aerosols in this region [6]. As part of the International Polar Year (IPY) activities, the GOA-UVa carried out an intensive campaign at the subarctic station of ALOMAR (Arctic Lidar Observatory for Middle Atmosphere Research) in the summer 2008.

In this work, the absorption data obtained between 13 June and 25 August of 2008 during this campaign are presented. Two measurement techniques were used: the particle absorption soot photometer PSAP (Radiance Research), and the integrating sphere photometer (FEI). The FEI provides measurements of continuous spectra from the ultraviolet to the near infrared wavelengths. Finally, backtrajectories of air masses are analyzed for most relevant events and these findings give tentative evidence about predominant sources of aerosol particles.

2. Methods and instruments

The sub-Arctic station of ALOMAR (69° 16' 42" N, 16° 00' 31" E, 380 m a.s.l) is located about 300 km at the north of the Arctic circle, on the Andøya island in the Vesterålen archipelago. The measurement site is well suited for tropospheric measurements due to the absence of important local and regional pollution sources.

The *in situ* measurements were performed at low relative humidity (RH 40%) which was controlled using a heater tape. An aerosol inlet system was mounted above the roof of the building (about 7 m above ground level). Aerosol samples were collected at the top of a stainless steel intake stack with a cut-off diameter of 10 μm and a height of 2 m above roof level. The stack was fitted with a rain cap, and sampling occurs from the underside of the cap through a metal screen designed to prevent insects from entering the system. Inside of the building, the aerosol flow was divided into several flows using a distribution system, and the flow rate was calibrated every day with a Gilibrator Gilian®. The three stage cascade impactor (Dekati® PM10) was installed to collect particles on Ø47 mm Millipore filters with homogeneous size distribution pores of 0.2 μm . The impactor cutpoints are 10, 2.5 and 1 μm . The sample flow rate was 15 l/min. The size-resolved filters were collected once daily during the campaign period,

except on rainy days. The impactor DEKATI is shown in Fig. 1.

Filter-based techniques are used in this work for the measurement of aerosol light absorption. The aerosols are deposited at a known volumetric flow rate V [m³] on a filter area A [m²]. If the particles would not scatter light, their absorption coefficient σ_a , in units of m⁻¹, could be measured with a simple optical transmission measurement on the filter. The transmission measurement consists of measuring the optical intensity I_0 transmitted through the filter blank and the optical intensity I transmitted through the loaded filter, which are connected through Beer's law [7] and the absorption coefficient can be determined as

$$\sigma_a = -\frac{A}{V} \ln \left[\frac{I}{I_0} \right]. \quad (1)$$

The Particle Soot Absorption Photometer (PSAP, Radiance Research S/N 96, Seattle WA) is a commercially available, filter-based light-absorption instrument (Fig. 1). This device provides measurements at the wavelengths of 470, 522 and 660 nm with a temporal resolution of 1 second. The PSAP measures the temporal change of the transmittance, while a fiber filter (Pallflex) is being loaded. In this case, the limit transmittance was set to 75%, and the sample flow rate was set to 1.5 l/min. The PSAP data had to be corrected for particles scattering. The scattering coefficient was measured with an integrating nephelometer TSI. This correction was applied based on the method of Bond *et al.* (1999) [8].



Fig. 1. DEKATI impactor and PSAP, commercial filter-based instruments.

The FEI was developed for measuring the spectral absorption coefficient from the UV to the near infrared wavelengths with a spectral resolution of 10 nm. The system uses a filter-based method. The integrating sphere collects or integrates the scattered light, so that the absorption alone reduces transmitted light. The light absorption is determined by measuring and comparing the intensity over filters with and without particles deposited. These filters are dissolved totally to produce liquid suspensions of the deposited particles. Figure 2 shows schematic diagram of the FEI and Fig. 3 shows the FEI setup.

Key components of the FEI are: (1) A 6 inch integrating sphere manufactured by Gigahertz-Optik® with a spectral reflectance of 98% that cover the wavelength range from 250 to 2500 nm. (2) The light source contains a 150 W Xenon lamp integrated in APEX illuminator (ORIEL®) equipped with a transmission filter, and an automated scanning monochromator (ORIEL® Cornerstone™ 130). (3) The signal is detected by P-9710-1 Gigahertz Optik photometer. Since only ratios of the signals with and without samples are used, instrument errors cancel.

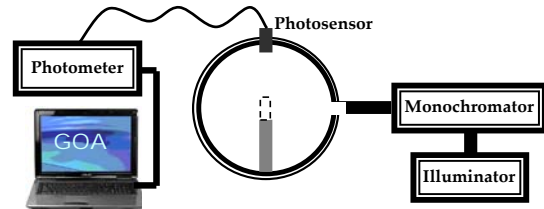


Fig. 2. Schematic diagram of the FEI. The dashed line into the sphere is the mounting for a liquid sample.



Fig. 3. FEI setup.

3. Results and discussion

The hourly average values of σ_a measured by PSAP range between 0.01 to 2.81 Mm^{-1} , 0.01 to 2.28 Mm^{-1} and 0.01 to 1.83 Mm^{-1} at 470, 522 and 660 nm, respectively. For the same wavelengths, the daily means values of σ_a are 0.39, 0.35 and 0.32 Mm^{-1} . Table I shows a brief statistics of daily σ_a values during the campaign period.

The maximum value registered was 1.72 Mm^{-1} at 522 nm. These values are particularly low, and close to the lower detection limit. However, these values are comparable to those measured at other Arctic sites, including measurements in Barrow, Alaska during summer time [9] and measurements made in Greenland [10]. The absence of local pollution sources and the low influence of carbonaceous particles originated in areas close to ALOMAR station could explain the low σ_a values.

Table I
Statistics of σ_a daily values measured by PSAP

λ [nm]	Mean Value [Mm^{-1}]	SD	Minimum Value [Mm^{-1}]	Maximum Value [Mm^{-1}]
470	0.42	0.30	0	1.50
522	0.47	0.34	0	1.72
660	0.35	0.25	0	1.22

The comparison of absorption data time series by PSAP and FEI at 470, 520 and 660 nm is presented in Fig. 4. The correlation between these measurements is shown in Fig. 5. The best correlation was obtained at 520 nm ($R=0.95$), and the correlation is the lowest at 470 nm ($R=0.92$). These results reflect the good agreement between the PSAP and the FEI instruments. Moreover, this agreement confirms the good response of both instruments. The stronger discrepancy between the instruments was registered on 29 July. This day, the PSAP measured a high value of σ_a , which was even higher according to the FEI measurement. Therefore, these days (i.e. with high absorption) should be studied in more details on the following paragraphs.

The comparison of both instruments shows that for the low and moderate levels of absorption, they have a good agreement. For

these levels of absorption at three wavelengths of PSAP, the differences are between 5% and 6%, while the differences increase up to 15% for higher absorption levels.

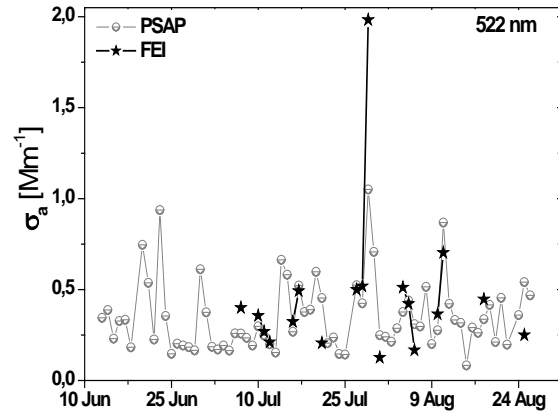


Fig. 4. Time series comparison of the daily average of σ_a as measured by the PSAP and the FEI.

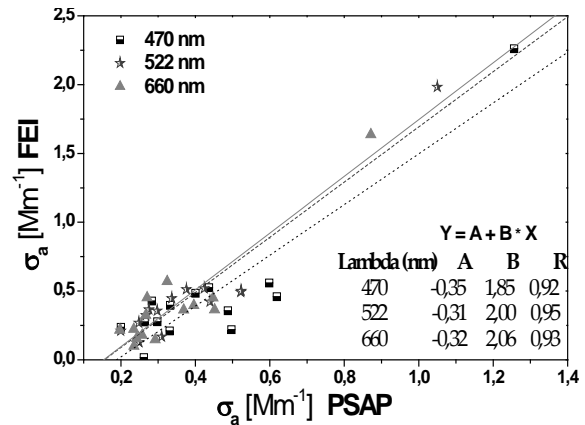


Fig. 5. Scatter plot comparison of σ_a daily average measured by the PSAP and the FEI.

Spectral absorption coefficients measured by FEI are shown in Fig. 6 for three representative days. These three days correspond to the highest values, the medium values and the lowest values of σ_a , observed during the campaign period. The absorption coefficient decreases with increasing wavelength, this behaviour is known as the gray absorption and this is a typical characteristic of absorbing carbon [11]. Between 320 and 400 nm the σ_a values decrease more sharply in the two lower spectra, showing that for certain aerosol compositions, the absorption in the near ultraviolet presents more variability than in the visible.

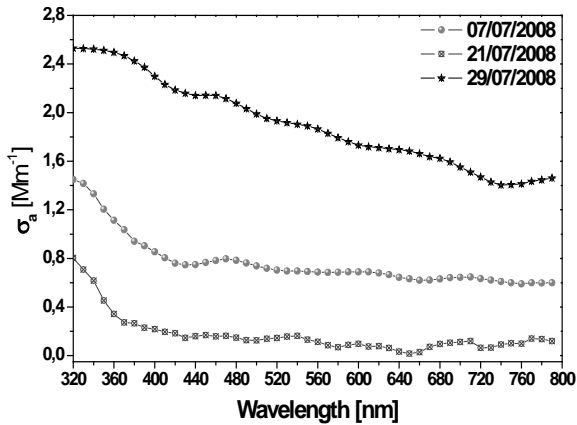


Fig. 6. Spectra of σ_a given by FEI for three representative days (7, 21 and 29 July 2008)

From FEI measurements in the near ultraviolet to the visible, the spectral dependence (α_a) of σ_a was derived. For most of the days α_a is an inverse function of wavelength (λ^{-1}). For the days with high absorption, the spectral dependence in the UV-A domain is highly variable. This could be explained by the high variability of the aerosol shape and composition. More detailed studies that include measurements of chemical composition and size distribution could help to clarify this behaviour.

During the campaign period, the absorption coefficients are lower than 2 Mm^{-1} for daily average values. These values are considered normal for this type of study area [12]. The largest absorption coefficient was registered on 29 July with different measurements devices. For this day, the daily average value of σ_a was of 1.1 Mm^{-1} , as measured by PSAP. According to Bergstrom *et al* (2007) [13] the spectral dependence parameter (α_a) is greater than 1 for carbonaceous particles, in this case, α_a is equal to 1.05. Furthermore, the analysis of HYSPLIT backtrajectories together with satellite images from MODIS (Fig. 7 and Fig. 8) [14], show that the presence of fine mode particles with high content of BC originated in biomass burning can be confirmed.

4. Conclusions and outlooks

As a part of the activity in the International Polar Year (IPY-2007-2008), the Atmospheric Optics Group carried out an intensive campaign during the summer 2008 to study atmospheric aerosols

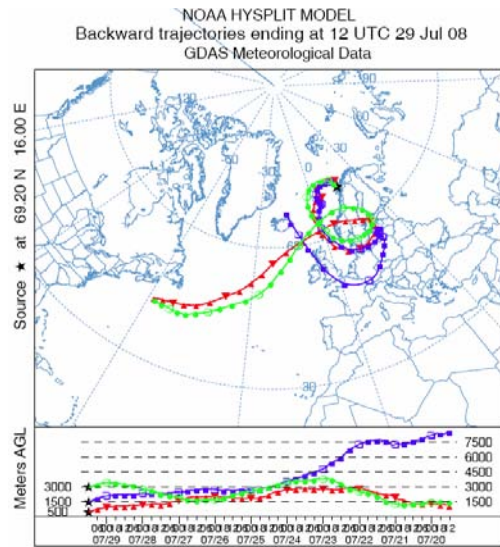


Fig. 7. Air mass backtrajectories from HYSPLIT model.



Fig. 8. MODIS image for the ALOMAR station at 29 July 2008.

in the sub-Arctic station ALOMAR at north of Norway. Two different filter-based instruments were used to measure the aerosol absorption coefficient (σ_a). These instruments are the commercial photometer PSAP, and the integrating sphere photometer (FEI). For this work, the FEI, which measures continuous spectra in the UV and the NIR with a good spectral resolution, has been implemented and calibrated. The bibliographic references about the measurement of continuous spectra of absorption are very scarce. One of the main objectives of this work was to show the first measurements of this kind in the research site.

The implementation of the FEI technique gives the possibility to start with a database about aerosol characterization. The technique is

planned to be applied in different locations and future results will be found at different places as in Colombia where the technique will be implemented as part of the outlook of this first experience.

Acknowledgments

This work was supported by Complementary Action CGL2008-01571-E, the project CGL2008-05939-C03-01 for CICYT and GR-220 of Junta de Castilla y León. The Programme Alban for the scholarship No. E06D101060CO is also gratefully acknowledged. The authors would like to thank the ALOMAR staff for their technical support and the INTA team for participating in this campaign. The presentation of this work in the V Workshop Lidar measurements in Latin America at Buenos Aires, Argentina (November, 2009) was supported by NASA GODA group.