Optical properties of tropospheric aerosols derived from lidar and sun photometer measurements at ALOMAR (69N) in 2005 and 2006

Propiedades ópticas de los aerosoles troposféricos obtenidos con lidar y fotómetro solar en ALOMAR (69N) durante los años 2005 y 2006

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REFERENCES AND LINKS


ABSTRACT:

We report the results of Sub-Arctic aerosol properties, obtained during three field campaigns conducted in 2005 and 2006. These have been carried out at the Arctic Lidar Observatory for Middle Atmosphere Research (ALOMAR), Andøya Rocket Range, Norway (69N, 16E, 380 m asl), and were supported by the eARI (enhanced Access to Research Infrastructure) program of the European Union. The instruments used for this study are the ALOMAR tropospheric Lidar and a Cimel sun photometer. Data in situ from cascade impactors are also included to know the levels of total suspended particles and major ionic components during the measurement period in 2005. Back trajectory analysis is presented. The evolution of the vertical distribution of the aerosols is analyzed, in order to study aerosol stratification and height patterns for the long-range transported aerosols to this coastal Sub-Arctic site.

Keywords: Tropospheric Lidar; Sun photometer; Arctic; Aerosol transport; AOD.

RESUMEN:

Se reportan los resultados de las propiedades de los aerosoles sub-árticos obtenidos durante tres campañ as en 2005 y 2006. Realizadas en el observatorio ártico de Lidar para la investigación media de la atmósfera (ALOMAR), Andøya Rocket Range, Noruega (69N, 16E, 380 m), apoyados por el programa eARI (acceso a grandes infraestructura de investigación) de la unión europea. Los instrumentos usados para este estudio son un fotómetro solar CIMEL y el lidar troposférico de ALOMAR. Se tendrán también en cuenta los datos in situ de los impactores de cascada para conocer el nivel de partículas suspendidas y los componentes iónicos durante las campaña de 2005. Se presenta también el análisis de las trayectorias. La evolución de la distribución vertical de los aerosoles se analiza, para estudiar los patrones de la estratificación y de altura de los aerosoles transportados a este sitio costero sub-ártico.

Palabras clave: Lidar Troposférico, Fotómetro Solar, Artico, Transporte de Aerosoles, AOD.
1. Introduction

Tropospheric aerosols have an important role in climate for their contribution to cloud formations and sunlight attenuation affecting critically the global radiative balance. Depending on various generating sources, tropospheric aerosols may vary greatly in both time and space. Several models for the study of the optical properties of the tropospheric aerosols have been developed. However, the optical properties of aerosols, governed by physical parameters such as particle density and size distribution, have not yet been well characterized in the Arctic zone.

In the Arctic, long term observations, especially in-situ and a few aerosol lidar measurements, are carried out at Ny-Ålesund/Svalbard (79°N) [1]. The location of ALOMAR observatory, north of the Arctic Circle and on an island, a few hundred meters from seashore and about 30 km off the continent, it is ideal for investigations related to Arctic phenomena such as noctilucent clouds, polar stratospheric clouds and Arctic haze. For air masses from the north and north-west ALOMAR can represent a reference station for almost unpolluted, clear air. Air masses passing above ALOMAR with back-trajectories from the north-east transport continental air from the urban centres in northern Russia, whereas western and south-western back-trajectories have a strong maritime influence. ALOMAR’s year-round capability is essential for long term studies to also include intra-annual variations.

Previous studies report synergetic use of lidar and sun-photometer [2-4]. The lidar and sun-photometer can provide information regarding microphysical properties of the aerosols by combining observations at different optical wavelengths. We present the results of Arctic campaigns in 2005 and 2006. With the simultaneous observations by Cimel sun
photometer, lidar and impactors cascade we can know the AOD and the levels of total suspended particles and major ionic components during the measurement period in 2005. It is extending the former 2002 and 2003 campaigns conducted by the Group of Atmospheric Optics of Valladolid [5].

2. Instruments

2.1. The Sun photometer

The Cimel Electronique CE-318 sun photometer is an automatic sun and sky radiometer, with spectral interference filters centered at selected wavelengths: 340, 380, 440, 500, 670, 870, 1020 and 1640 nm for aerosol measurements. The filter band pass has 2 nm FWHM in the UV and 10 nm FWHM for the visible and infrared regions. The Cimel sun photometer is the standard instrument of AERONET network [6]. Direct sun measurements are performed at these wavelengths (1.2° field of view) to determine aerosol optical depth (AOD) and another channel at 940 nm is used for water vapor content retrieval. Sky radiance measurements are acquired at 6 wavelengths (from 440 to 1640 nm) in the solar almucantar and principal plane (for details, see [6,7]). Inversion algorithms can then be applied to the sky and direct sun measurements to retrieve aerosol size distribution, single scattering albedo, phase function and complex refractive index [8,6].

The Cimel sun photometer was calibrated at El Arenosillo within the Spanish Network for Aerosol Measurements (RIMA), in close collaboration with AERONET and according to the AERONET protocols. The direct sun channels were calibrated by inter-comparison with a master instrument. For the sky channels calibration an integrating sphere with a parabolic primary mirror (focal length 125 cm) was used. The effective focal length of the lidar is reduced in the focal box to about 60 cm. Also in the focal box the spectral selection (<450 nm, 450-600 nm, >600 nm) and the selection of the parallel- (p) and cross-polarized components (s) at 532 nm are performed. The two wavelength bands <400 and >600 nm are guided via optical fibers to a spectral analyzer separating elastic and different inelastic channels. These spectrally (and by polarization) separated channels are then additionally filtered using narrow band interference filters, what minimizes crosstalk and background noise. Behind the filters the beam is focussed on photomultiplier tubes or an APD, and finally the electronic output is analyzed by a combination of one transient recorder and one photon counting device for each channel. At the moment only five detection channels are used (355o, 532p, 532s, 1064o and 387o), but the data acquisition electronics shall be extended to detect simultaneously three additional Raman channels (two H2O Raman at 408 and 660 nm and one extra N2 Raman at 608 nm).

During the reported period, the only Raman signal available was too weak to provide valuable extinction profile throughout the troposphere. Moreover, the altitude of full overlap was most of the time around 1400 m asl. Therefore the lidar data are presented here only to characterize the aerosol vertical distribution [10].

2.2. The tropospheric lidar

The newly setup tropospheric Lidar at ALOMAR, in operation since July 2005, is an aerosol backscatter lidar that provides elastic and inelastic atmospheric returns in the ultraviolet, visible and near infrared.

Since daytime capabilities are particularly relevant in the summer Arctic, the interference filters of the receiving optics are chosen rather narrow (0.2 nm). Crucial for the daylight capability is also the powerful laser (1020 mJ at 30 Hz repetition rate), which has been used before for a mesosphere system. Part of the original 1064 nm light is transformed into its second (532 nm) and third harmonics (355 nm). The light at the latter two wavelengths is linearly polarized. A refractive beam-widening telescope expands the outgoing beam to about 5 cm and reduces the beam divergence to less than 140 μrad. The telescope is a Newtonian type with a parabolic primary mirror (focal length 125 cm). The effective focal length of the lidar is reduced in the focal box to about 60 cm. Also in the focal box the spectral selection (<450 nm, 450-600 nm, >600 nm) and the selection of the parallel- (p) and cross-polarized components (s) at 532 nm are performed. The two wavelength bands <400 and >600 nm are guided via optical fibers to a spectral analyzer separating elastic and different inelastic channels. These spectrally (and by polarization) separated channels are then additionally filtered using narrow band interference filters, what minimizes crosstalk and background noise. Behind the filters the beam is focussed on photomultiplier tubes or an APD, and finally the electronic output is analyzed by a combination of one transient recorder and one photon counting device for each channel. At the moment only five detection channels are used (355o, 532p, 532s, 1064o and 387o), but the data acquisition electronics shall be extended to detect simultaneously three additional Raman channels (two H2O Raman at 408 and 660 nm and one extra N2 Raman at 608 nm).

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2.3. Cascade impactors

A six-stage cascade impactor was used to measure the particle size distribution. The aerodynamic cut-off diameters are 0.45 μm, 0.73 μm, 1.73 μm, 4.21 μm and 9.76 μm allowing the description of six fractions of particulate matter. In order to avoid a
situation where the particles bounce off the impactor, it was made of electrostatic nylon. The design of the cascade impactor has been described in detail previously [11]. The collection surfaces were polycarbonate membrane filters with 0.2 \( \mu \text{m} \) pores and a constant airflow rate of 16.5 l/min was used.

The filters were weighed before and after collection with 24 h of filter stabilization and acclimatization in the weighing room. Special care was taken in order to remove electrical charges from the filters.

The samples were analyzed for major ions using ion chromatography. The filters were treated with deionized water and the soluble matter ultrasonically extracted. Samples were placed in the ultrasound bath for half an hour. Then, levels of Cl\(^-\), NO\(_3\)-, SO\(_4^{2-}\), Mg\(^{2+}\), Ca\(^{2+}\), K\(^+\), Na\(^+\) and NH\(_4^+\) were assayed by means of ionic chromatography with conductometric detection. This procedure allowed the mass ions of the six well established sizes to be obtained. The average error in the measurements does not exceed 5%.

The sampling was possible only under special conditions avoiding unstable weather and the very frequent fog events which implied less measurement days and more damaged samples. We had a total of 13 samples were analyzed in ALOMAR (380 m ASL) in 2005 from 24 Jan to 05 Apr and from 11 Jun to 16 Aug.

### 3. Results

Figures 1 and 2 show the temporal evolution of spectral AOD for 550 nm and 1020 nm and the derived Ångström exponents for the range of wavelengths 440-870 nm as suggested by the AERONET protocols, during the campaigns in 2005 and 2006 respectively. We will analyze representative cases from each campaign.

![Fig. 1. Series of Aerosol Optical Depth measured at two wave-lengths (500 nm and 1020 nm, left scale) and Ångström exponent (right scale) measured with CIMEL sun photometer at ALOMAR during summertime 2005.](image)

#### 3.1. Impactor

In ALOMAR the total atmospheric particulate (TSP) values registered range from 3.97 \( \mu \text{g/m}^3 \) to 101.87 \( \mu \text{g/m}^3 \) and the average for all measured days is 29.09 \( \mu \text{g/m}^3 \) (std=28.86 \( \mu \text{g/m}^3 \)). It can be observed predominance of the coarse mode in winter and the fine mode in summer.

The total atmospheric particulate fraction was calculated as the addition of the average ionic concentrations of the cascade impactor. The anthropogenic ions, NO\(_3\)- and SO\(_4^{2-}\), are important species found in the atmospheric particulate fraction, with 0.31 and 0.35 \( \mu \text{g/m}^3 \), respectively. Table I summarize the mean levels of total suspended particles and major ionic components during the measurement period.

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<th>ALOMAR</th>
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<tr>
<td></td>
<td>min</td>
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<tr>
<td>TPS</td>
<td>3.97</td>
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<tr>
<td>Cl(^-)</td>
<td>0.09</td>
</tr>
<tr>
<td>NO(_3)^-</td>
<td>0.08</td>
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<tr>
<td>SO(_4^{2-})</td>
<td>0.04</td>
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<tr>
<td>Na(^+)</td>
<td>0.10</td>
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<tr>
<td>NH(_4^+)</td>
<td>0.03</td>
</tr>
<tr>
<td>K(^+)</td>
<td>0.01</td>
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<tr>
<td>Ca(^{2+})</td>
<td>0.01</td>
</tr>
<tr>
<td>Mg(^{2+})</td>
<td>0.04</td>
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</table>
In order to determine the marine influence on the composition of aerosols in ALOMAR, sea-salt ratios were calculated using Na+ as reference, assuming all Na+ to be of marine origin [12]. These sea-salt ratios are given in Table II. From Table II that the sea-salt ratios for SO$_4^{2-}$, K$^+$ and Ca$^{2+}$ are higher than the seawater ratios, which indicates incorporation of non-marine constituents in aerosols. The Cl$^-$ /Na$^+$ ratio is similar to the marine aerosol ratio, denoting the marine source as the only one for the Cl$^-$ ion in the atmospheric particulate fraction.

**TABLE II**

<table>
<thead>
<tr>
<th>Component</th>
<th>Cl$^-$/Na$^+$</th>
<th>SO$_4^{2-}$/Na$^+$</th>
<th>K$^+$/Na$^+$</th>
<th>Ca$^{2+}$/Na$^+$</th>
</tr>
</thead>
<tbody>
<tr>
<td>sea water</td>
<td>1.8</td>
<td>0.25</td>
<td>0.037</td>
<td>0.038</td>
</tr>
<tr>
<td>ALOMAR</td>
<td>1.9</td>
<td>1.96</td>
<td>0.152</td>
<td>0.127</td>
</tr>
</tbody>
</table>

Fig. 3. Aerosol Optical Depth measured at two wavelengths (500 nm and 1020 nm) and Ångström exponent measured by CIMEL sun photometer at ALOMAR from July 1 to 3, 2005.

### 3.2. Observations from July 1 to 3, 2005

The statistical results for aerosol optical depth and Ångström exponent for the first days of July 2005 give an average AOD of 0.048 with standard deviation (STD) of 0.008 at 500 nm and an average AOD of 0.013 (STD 0.003) at 1020 nm. The Ångström exponent is 1.86 (STD 0.09) (Fig. 3). During the end of June and beginning of July 2005, a stable high pressure system was centered between northern Scandinavia and the Svalbard archipelago, remaining almost for one week. A warm and dry air mass arrived at our site from the south east, with temperatures around 18-20°C at noon. This is consistent with the mostly cloudless lidar observations (Figs. 4-5, top panels) and the rather poor and stable aerosol stratification (bottom panels) observed above 1400 m asl (full-overlap lower altitude). According to the HYSPLIT back-trajectories (Fig. 6) air masses arriving above ALOMAR on July 2 have had a long residence time over sea. Moreover the lowest back-trajectory is fully confined to the Arctic. This is quite consistent with the low observed AOD values, while the very high values of Ångström exponent are rather surprising for such maritime back-trajectories.

Fig. 4. Lidar observations on July 1, 2005. The temporal resolution is 8 minutes. The top panel shows the time-series of range-corrected signals (RCS) for the 532 nm (p) channel. The bottom panel show the corresponding altitude log-derivative. In both panels, saturated colours are used.

Fig. 5. Diurnal cycles of Lidar observations from July 2 to 3, 2005. The conditions are the same as in Fig. 1.
3.3. Observations on August 11, 2005

The AOD is higher than in the last period, with average value at 500 nm of 0.12 (STD 0.02) and 0.062 (STD 0.014) at 1020 nm. The average Ångström exponent is 1.26 (STD 0.18) (Fig. 7). Unlike the case shown previously, the lidar observations (Fig. 8) show the presence of a strongly marked transition from aerosol rich to aerosol poor air parcels with increasing altitude (deep blue area in the bottom panel), what is consistent with higher AOD values. The back-trajectories (Fig. 9) present about five days residence time exclusively over land, what is consistent with higher AOD.

3.4 Observations on April 17, 2006

In the spring season the AOD is lower than in summer time. The Arctic troposphere is extremely clean. The long residence time of aerosols was apparent from the pronounced narrow size distribution centred in of minimum efficiency for particulate removal in the atmosphere. The average AOD got at 500 nm of 0.045 (STD 0.06) and 0.015 (STD 0.015) at 1020 nm. The average Ångström
The lidar presents very slow temporal change, with only minor change in aerosol vertical distribution. The south east air mass (Fig. 12) was mild and dry and the measurements were carried out from 16th in the evening until the morning of day 18th, still with mild temperatures (+3°C) but with pressure decreasing from 960 to 945 hPa.

Fig. 10. Aerosol Optical Depth measured at two wavelengths (500 nm and 1020 nm) and Ångström exponent measured by CIMEL sun photometer at ALOMAR on April 17, 2006.

Fig. 11. Time series of Lidar observations on April 17 2006. The conditions are the same as in Fig. 1.

Fig. 12. 7-days back-trajectories ending above ALOMAR on April 17, 2006 at 12h UTC.

3.5 Observations from August 27 to 28, 2006
The Cimel observations of aerosols at ALOMAR for these days have several similarities with the previously considered time series. The relatively small variation of AOD is consistent with the stable anticyclonic conditions prevailing in the region, while the AOD average values of 0.08 (STD 0.04) at 500 nm and of 0.02 (STD 0.03) at 1020 nm (Fig. 13) are quite typical for summertime at this location.

The rather high values of the Ångström exponent are indicative of prevailing small particles. The 7-days back-trajectories (Fig. 15) are mostly confined over land, what is consistent with relatively high AOD and with the persistent presence of a marked aerosol layer transition between 2 and 3 km asl in the lidar time series (Fig. 14).

Fig. 13. Aerosol Optical Depth measured at two wavelengths (500 nm and 1020 nm) and Ångström exponent measured by CIMEL sun photometer at ALOMAR from August 27 to 28, 2006.
6. Conclusions

As a continuation of the former campaigns of measurements at ALOMAR and the related study of the aerosols in the Arctic, the Group of Atmospheric Optics from Valladolid performed new measurement campaigns during the summers 2005 and 2006. In this paper we present case studies regarding the aerosol dynamics and characterization by the simultaneous and collocated measurements of a CIMEL sun photometer and a tropospheric backscatter lidar, and the levels of total suspended particles and major ionic components during the measurement period in 2005 measures with the cascade impactor. The observed AOD variations are consistent with the Lidar observations and with the back-trajectories of air masses. The observed Ångström values are found to be quite typical for a remote continental location likely with low contribution of coarse maritime aerosols.

The interpretation of the high Ångström exponents even when air masses were expected to bring aerosols of maritime type is not clear yet. In situ analysis of particle size distribution and chemical analysis would be needed to help clarifying the likely origin of the predominant fine particles observed above ALOMAR. This issue shall be further investigated in upcoming campaigns.

As more data are available, the observed aerosol stratifications must also be further investigated, related more about the both to the season and to the airmass origin. Since July 2006 a Cimel sun photometer operates at Alomar on a permanent basis. It may provide, together with the Lidar, a climatological approach to the aerosol properties in this Arctic region.

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