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Aerosol characterization at the Pierre Auger Observatory

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Abstract: The Pierre Auger Observatory is a hybrid facility composed of surface and fluorescence detectors (denoted as SD and FD, respectively), where the FD system sets the energy scale in cosmic ray shower reconstruction. Atmospheric components attenuate the fluorescence light emitted by the de-excitation of atmospheric nitrogen, previously excited by the charged particles of the shower. Amongst these components, atmospheric aerosols are the ones with the largest fluctuations, being responsible for one of the major uncertainties in shower reconstruction with FD data. We present a detailed characterization of aerosols. They are collected at the Observatory and analyzed, in morphology and elemental composition, with experimental techniques used for the first time in a cosmic ray observatory: gravimetry, PIXE and SEM/EDX. An analysis of wind trajectories using the program HYSPLIT is used to understand the sources and the evolution of aerosols. The aerosols are further characterized by the FRAM, an optical telescope employed to perform CCD photometry of selected Landolt fields, in which we observe sets of precisely measured standard stars at various wavelengths. Using this photometric information we then compute the Ångström coefficients that also characterize the size of aerosols.

Keywords: Pierre Auger Observatory, atmospheric aerosols, direct sampling, gravimetry, PIXE, SEM/EDX, HYSPLIT, FRAM

1 Introduction

The Pierre Auger Observatory, conceived to measure ultra-high energy cosmic rays, has a hybrid design, consisting of surface and fluorescence detectors. The FD system is composed of 27 telescopes distributed in four stations [1]. The FD detects fluorescence light (300-450 nm) produced by the interaction of the cosmic ray showers with atmospheric nitrogen. This light is attenuated on its path from the shower to the telescopes by atmospheric constituents, with aerosols (suspended particles) playing a role of major importance. Since the aerosols are highly variable in space and time, a continuous monitoring of them is necessary. A large network of aerosol monitors is installed at the Observatory. Some of them, like the lidars (4 elastic ones and a Raman one) or CLF and XLF (Central and eXtreme Laser Facilities), obtain the aerosol optical depth as a function of height [2]. On the other hand, the FRAM (F/Photometric Robotic Atmospheric Monitor) is a star monitor that measures the total aerosol optical depth from the top of the atmosphere to the ground. Elastic lidars and the FRAM can also measure the optical depth from the shower to the FD, as part of a rapid monitoring program [3]. To complement the setup, the HAM (Horizontal Attenuation Monitor) has been used to measure the horizontal attenuation length between the FD sites almost at ground level and the APF (Aerosol Phase Function Monitors) give the aerosol phase functions (describing the angular distribution of aerosol diffused light) [4]. For these devices, the aerosols work just as a medium interacting with the fluorescence light. But their measurements do not specify what the properties of the aerosols are (shape, size, composition), apart for data from the APF and FRAM that give information on their mean size. Their characteristics remain hidden and some assumptions are made to model their interaction with radiation. Therefore, a detailed aerosol characterization project, by direct measurement and analysis, is being performed at the Observatory to improve and complement the information supplied by the other monitors. Knowledge of aerosol characteristics permits one to infer their origin (sources, evolution). This can be correlated with a study of air mass trajectories which evaluates the behavior of aerosols in space and time.

2 Instruments and techniques for aerosol measurements

2.1 Direct aerosol sampling using an Andersen-Graseby 240 (A-G 240)

By using an A-G 240 dichotomous sampler, direct sampling of aerosols takes place at the Auger Observatory. This instrument is installed at Coihueco FD station (35°06'52.9" S, 69°36'02.7" W, 1712 m a.s.l.), at 6.3 m above ground level (AGL). It has a pump that drives air into it, sweeping along

atmospheric particulate matter PM10 (with aerodynamic diameters d$\leq$10μm). The air is divided in two fluxes, one carrying fine particles (d$\leq$2.5μm) and the other carrying the coarse ones (2.5$<$d$\leq$10μm), which are deposited in two filters of polycarbonate (Millipore®HTTP, diameter 37 mm, pore 0.4 μm). The sampling period is 24 h. The operational, or actual, flow rate Q_a is 16.7 l/min. Q_a was corrected to U.S. Environmental Protection Agency reference conditions (298 K and 760 mm Hg), to obtain the standard flow rate Q_{std} used in calculations of mass and elemental concentrations. The aerosols captured in the filters are later analyzed by different experimental techniques: Gravimetry, PIXE, SEM/EDX (see Sec. 3.1, 3.2 and 3.3).

2.2 Concentration measurements using a Grimm 1.109

A portable laser aerosol spectrometer and dust monitor Grimm 1.109 was installed in the FD building of Coihueco in November 2010, at 1.715 m AGL, to perform local superficial aerosol concentration measurements. It operates with a dual technique: a) Continuous measurements of particle or mass concentration (particle/liter, μg/m^3), in fixed time intervals, for size channels from 0.22 to 32μm. The principle of operation is based on an internal laser (655 nm) and a model for the light dispersion produced by aerosols contained in the flux of air driven into the apparatus by a pump (flow rate 1.2 l/min) b) Collection of particles in a filter for later analysis.

2.3 Ångström coefficient measurements using FRAM

FRAM is a small optical telescope located about 20 m from the FD building at Los Leones (35°29'45.2" S, 69°26'58.9" W, 1430 m a.s.l.). The Schmidt-Cassegrain telescope with diameter of 300 mm is equipped with a type G2 CCD camera from Moravian Instruments and uses the Johnson & Bessel set of UBVRI filters. It is equipped with a wide-field camera (Moravian instruments G4) using a Nikkor 300 mm/f 2.8 camera lens with diameter of 12.5 cm. The telescope uses the equatorial Bisque Paramount ME mount and operates in robotic mode driven by the custom-made RTS2 software package [5]. It regularly observes selected fields of standard stars, so called Landolt fields, in various filters to derive extinction coefficients (or optical depths) at various wavelengths. The goal is to obtain the Ångström coefficient γ, used for parametrization of wavelength λ dependence of the aerosol optical depth τ_A: $\tau_A(\lambda) = \tau_{A0} \times (\lambda_0/\lambda)^\gamma$, where λ_0 is the reference wavelength and τ_{A0} is the aerosol optical depth measured for this wavelength. The Ångström coefficient is used in the cosmic ray shower reconstructions.

3 Instruments and techniques for aerosol analysis. Results.

3.1 Concentration analysis using Gravimetry

Filters are weighed with a Microbalance M3 (precision $\pm$1μg) before and after collecting aerosols with the A-G 240 at Coihueco, to obtain the mass of particulate matter deposited during the samplings. Before weighing, filters are conditioned (humidity 50% and temperature 25°C during at least 24 h) and irradiated with an alpha source (^{238}U) to eliminate static charge on them during weighing. The PM2.5 and PM2.5-10 concentrations (in μg/m^3), are calculated as the ratio between the collected mass and the volume of air that passed through the sampler during the period of measurement. The PM10 concentration in the ambient air is computed as the sum of PM2.5 and PM2.5-10 concentrations. The total volume of air sampled is corrected to standard conditions (V_{std}) and it is determined from the standard total flow rate Q_{std} and the sampling time (24 h). An analysis performed for a total of 36 days of measurements in the period June 2008-February 2009, gave a mean PM10 of 10.3μg/m^3 (standard deviation 6.5μg/m^3). PM2.5 and PM2.5-10 represent 31.1 % and 68.9 % of PM10. There is a trend towards increasing concentrations with warmer seasons. During the coldest days of the winter, very low concentration values are observed (of less than 2.5μg/m^3) because snow keeps aerosols captured at the soil surface. Concurrently, in winter, air masses arrive at the Observatory mostly from the Pacific Ocean, presenting a lower aerosol content (see Sec. 4).

3.2 Elemental analysis using PIXE and SEM/EDX

The PIXE technique [6] is performed on PM2.5 and PM2.5-10 samples of aerosols collected at the Auger Observatory with the A-G 240, to analyze their elemental composition (from S up), at the TANDAR Laboratory accelerator facility of the Comision Nacional de Energia Atomica, Argentina. The targets were irradiated with heavy ions ^{16} O (7+ charge state) and the induced X-rays, characteristic of the elemental composition of the samples, were measured using an EG&G Ortec Si(Li) detector (sensitive area of 80 mm^2, 12.5μm Be window), with a resolution of 220 eV at 5.9 keV (Ka Mn line). More details about the experiment can be found elsewhere [7]. The X-ray spectra were analyzed with the WinQxas 1.40 computer code developed by IAEA. A PIXE analysis performed on 19 samples of each fraction corresponding to June-August 2008 showed that S, Cl, K, Ca, Ti, Mn and Fe, represent 25% and 13% of the PM2.5 and PM2.5-10 total mass, respectively. The rest of the mass is due to elements with low atomic number Z (not detected with our X-ray setup). S dominates in the fine fraction and Ca in the coarse fraction [7]. Elemental composition was also studied using a Scanning Electron Microscope (Philips SEM 515) with an Energy Dispersive X-ray system (EDAX Falcon PV 8200), provided with a Si(Li)-Be window detector. With this SEM/EDX arrangement, the detection of elements of Z higher than 11 (Na) is possible, complementing the PIXE results. Semiquantitative standardless analysis with ZAF factors for matrix correction was used for composition calculations. Si, Al, Ca, Mg and Fe, the typical mineral soil elements, are the major components, indicating that the aerosols present at the Auger Observatory consist mostly of suspended mineral dust from the soil of the Andean region. SEM observations indicate that the mass not detected by PIXE corresponds to

Si and Al (aluminosilicates)[7].

3.3 Morphological (shape and size) analysis using SEM images

SEM micrographs of the sampled aerosols, like the one shown in Fig. 1, are analyzed in morphology with software developed by using the ImageJ application (http://rsb.info.nih.gov/ij/). An analysis performed over 23 SEM images of June-August 2008 gave the result shown in Fig. 2 for the relative frequency of appearance of the different aerosols diameters, in the represented intervals (of 0.5μm, except the first one that ranges from 0.4 to 0.5μm, 0.4μm being the minimum aerosol size taken into account in the analysis, which corresponds to the pore size of the filters). Most of the analyzed aerosols (64.9%) were in the range 0.5-1μm. The shapes of the aerosols have been investigated through an approximation of circularity (defined as $4\pi\times$ Area / Perimeter^2) applied to the plane SEM images of the particles. It ranges between 0 and 1 (the latter for perfect circles). The analysis showed that 75% of the analyzed PM10 particles have circularity bigger than 0.5.

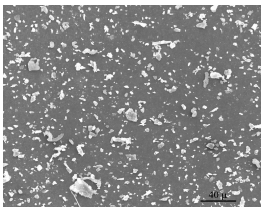

Figure 1: PM2.5-10 sample of 14 August 2008. Mass concentration: 13.6 μg/m^3.

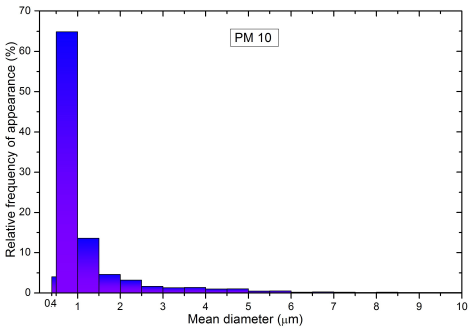

Figure 2: Relative frequency for different aerosol diameters. Period: June-August 2008.

3.4 Concentration and size distribution analysis using Grimm 1.109 data

Concentration measurements were obtained with Grimm 1.109 in Coihueco every 5 minutes, for different size channels, for June - August 2011. Mean mass concentrations for this period, for each size channel, normalized by the width of the size range, are shown in Fig. 3. The area under the histogram is the total mean concentration for June - August 2011. Table 1 gives the concentration values for some size ranges. PM2.5 and PM2.5-10 represent 21.2 % and 78.8 % of PM10. The particulate matter with aerodynamic diameter d>10μm contributes only 5.6% of the total concentration. The aerosol size range from 0.5 to 1μm represents only 2.2% of the total concentration (Table 1).

PMtotal	9.0	100%
PM0.5 (d≤0.5μm)	0.7	7.8%
PM>0.5 (d>0.5μm)	8.3	92.2%
PM2.5 (d≤2.5μm)	1.8	20.0%
PM10 (d≤10μm)	8.5	94.4%
PM2.5-10 (2.5<d≤10μm)	6.7	74.4%
PM>10 (d>10μm)	0.5	5.6%
PM0.5-1.0 (0.5<d≤1μm)	0.2	2.2%

Table 1: Concentrations values (in μg/m^3) for different aerosol size ranges (inμm) and percent contribution of these size ranges in the total aerosol concentration.

By contrast, the size analysis performed on SEM images of filters collected with A-G 240 during June-August 2008 showed that the great majority of the analyzed aerosols, 64.9 %, have sizes in that range (Fig. 2). Comparing both results for the same season of the year -even if they are for different years- it is evident that while the aerosols of the 0.5-1μm range are the most abundant, their contribution to total mass concentration has little significance, due to their light mass. Instead, concentrations measured by Grimm show a peak in the 4-5μm range (of 1.7μg/m^3, representing 18.9% of the total mass concentration) while atmospheric particles are less abundant at this size range according to SEM image analysis.

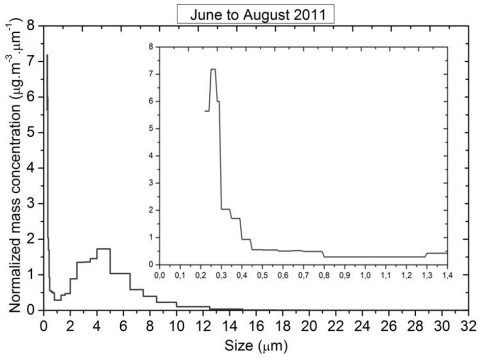

Figure 3: Left: Mean aerosol concentration for different size intervals, normalized by the width of the interval, obtained using the Grimm 1.109, June-August 2011. Right: idem with the smaller diameter range expanded.

3.5 Mean size analysis using FRAM data

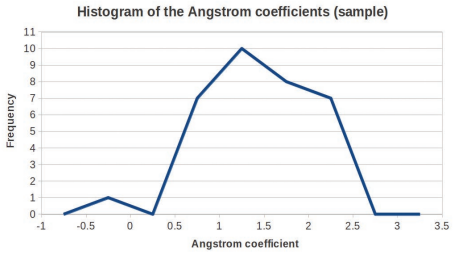

Figure 4: Histogram of FRAM measurements of the Ångström coefficient

FRAM measures the difference of the observed magnitude m_{OBS} and catalogue magnitude m_{CAT} of given stars within a selected standard field. Using the known airmass AM for the object, the total extinction coefficient κ can be easily derived. The result is transformed into total optical depth τ: $\kappa = (m_{OBS} - m_{CAT})/AM$; $\tau = (\sqrt[5]{100}/e)\kappa = 0.924\ \kappa$. The aerosol optical depth τ_A is obtained after subtraction of the computed molecular Rayleigh part. Since the absolute calibration of the FRAM telescope might be time dependent, the system is calibrated each night using observations of several different AM (within a few hours). Then, the observed dependence of κ is fitted on the AM independently of the telescope calibration constants, obtaining a precise result on κ. The Ångström coefficient γ is then obtained from the resulting aerosol optical depths for individual standard stars within one field, fitting the values in different wavelength filters. The Ångström coefficient varies from 0 to 4. Typical values: 0 (coarse particles, e.g. desert dust), 2 (fine particles, e.g. ash or automobile exhaust), 4 (molecules). This method of γ determination using CCD photometry of Landolt fields has been used at FRAM only since December 2012. From the available measurements (December 2012 - March 2013) we have constructed the histogram for γ (Fig. 4). The mean value of γ is 1.1 ± 0.7, indicating that both coarse and fine particles are present at the Observatory, in agreement with conclusions in other parts of this work.

4 Sources and evolution study using trajectories of air masses, HYSPLIT

HYSPLIT is an air-modeling program to calculate air mass displacements from one region to another [8]. It was used to analyze forward and backward trajectories of air masses to infer the sources and evolution of aerosols present at the Auger Observatory. 48 hour backward trajectories were evaluated every hour, throughout the year, for 2008-2010.

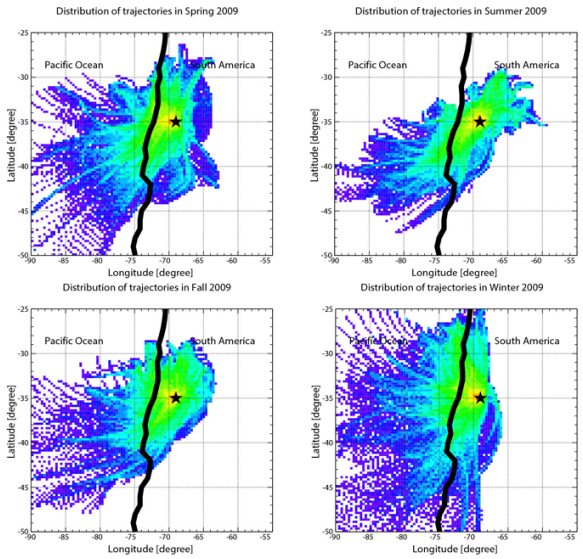

Figure 5: Seasonal analysis of 48 hours HYSPLIT backward trajectories evaluated every hour, arriving at the Auger Observatory at 500 m AGL.

A backward analysis performed at a start altitude of 500 m AGL showed that air masses originate mainly over the Pacific Ocean during the clear nights and travel principally through continental areas during the previous 48 h for hazy nights. Clear and hazy nights were identified using aerosol optical depth values at 3.5 km AGL, at Los Morados FD site, obtained from the CLF data during the mentioned period. Clear and hazy nights correspond to aerosol optical depths up to 0.01, for the former, and from 0.1 up, for the latter [9]. Elemental results previously described (Sec. 3.2), which indicated that most of the aerosols are soil suspended particles of the Andean region, explain the lower aerosol optical depth when the air masses have traveled mainly over the ocean during the previous 48 h. From the different monitors of the Observatory it is known that the presence of aerosols is lower in winter than in the rest of the year. A seasonal analysis performed with HYSPLIT during 2009 shows that in winter the backward trajectories of air masses spend more time over the ocean than in the other seasons (Fig.5).

5 Conclusions and Future Plans

The characterization of aerosols collected at the Auger Observatory is giving interesting information about their morphology and composition, thanks to the application of advanced analysis techniques used for the first time in a cosmic ray observatory. The results obtained from direct sampling and analysis complement information supplied by other aerosols monitors at the Observatory, which are evaluating the effect of these particles in fluorescence light attenuation. The results agree qualitatively with available FRAM data that estimate the mean size of the local aerosols, and can be combined with studies of air masses trajectories to infer the sources and evolution of these particles. This detailed aerosol characterization surpasses its application in cosmic rays showers reconstructions, being of major interest in other fields of study. A collaborative project, being designed with atmospheric scientists, is expected to give valuable information about the atmosphere at the southernmost latitudes of the globe.

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