Synergistic approach for Aerosol Characterization Studies during BAECC campaign

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- **Introduction and motivation**

  Aerosol lidar techniques provide information about aerosol vertical distribution and optical properties with high resolution in height and time, highly valuable for variability studies and for investigating aerosol modification processes and aerosol-clouds interactions.

  It was demonstrated that the combined use of advanced lidar aerosol vertical profiles, satellite observations and transport models permits the identification and accurate aerosol typing. A methodology for aerosol masking based on profiling/transport model synergistic approach (Mona et al., ACP 2012) was recently applied for providing the 4D distribution of the volcanic particles over Europe during the Iceland volcanic eruption on 2010 (Pappalardo et al., ACP 2013).

  The Planetary Boundary Layer aerosol content typing however is still undefined with this approach because of the complexity of the problem. Local aerosol formation and diffusion processes occurs in this altitude region and cannot be resolved the proposed methodology. The detailed characterization of aerosol at the surface provided during the BEACC campaign could provide the missing information for solving the PBL aerosol characterization puzzle.

- **Scientific objectives**

  The purpose of this project is fostering the full exploitation of advanced lidar aerosol vertical profiles for the aerosol characterization.

  This overall objective is reached through 2 specific sub-objectives:

  a) harmonizing BAECC ARM2 and Polly XT lidar data both in content, format and archiving

  b) exploring possible synergies with aerosol measurements at the ground, satellite observations and transport models.

  The combination of the expertise of the TNA beneficiary on aerosol lidar techniques and applications with the plethora of state-of-the-art measurements collected within the BAECC campaign at SMEAR II infrastructure is suitable for developing a synergistic approach for aerosol monitoring and characterization in the whole atmospheric column. On the other hand, the participation at BAECC of the ARM2 HSRL (High Spectral Resolution Lidar) and of the Kuopio EARLINET lidar is the right occasion for applying the ACTRIS /ARM agreement on data sharing across product holdings signed on November 2012 (http://science.energy.gov/~/media/ber/pdf/CESD_EUworkshop_report.pdf).

  The reported main objectives are addressed through specific activities:

  - using the Single Calculus Chain (SCC) developed in the EARLINET/ACTRIS component by CNR-IMAA for harmonizing analysis, formatting, and archiving of the ARM (US) and Polly XT (EU) advanced lidar systems;
  - acquiring the needed know-how about aerosol measurements and analysis at the surface performed during the campaign
  - developing a synergistic approach for aerosol characterization in the whole atmospheric column

- **Reason for choosing station**

  For this project, the plethora of state-of-the-art measurements collected within the BAECC campaign will be used in conjunction with satellite aerosol observations and transport models. BAECC campaign data collected at SMEAR II infrastructure provides the unique combination of instrumental facilities for developing a synergistic approach for aerosol monitoring and characterization in the whole atmospheric column.

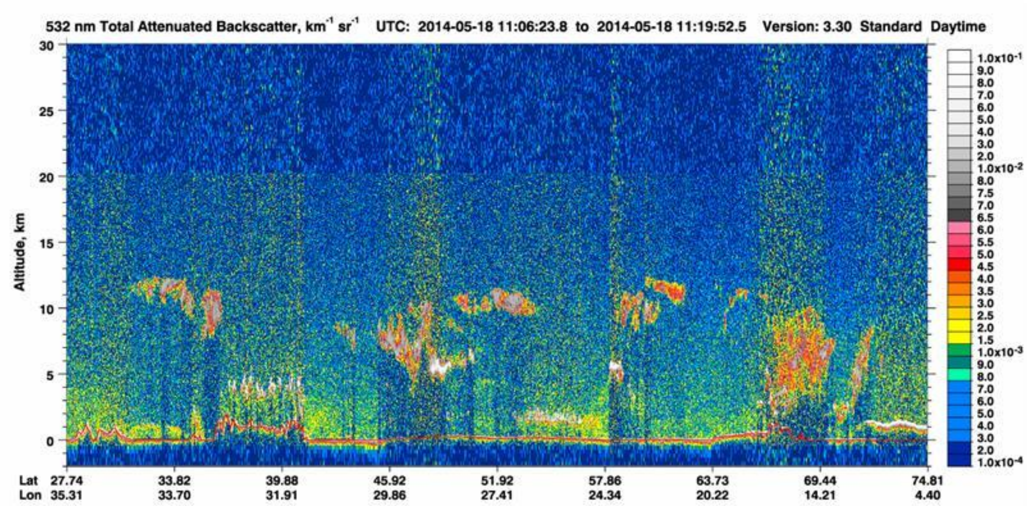
- **Method and experimental set-up**

Activities related to the project started already before the visit to the SMEAR facility.

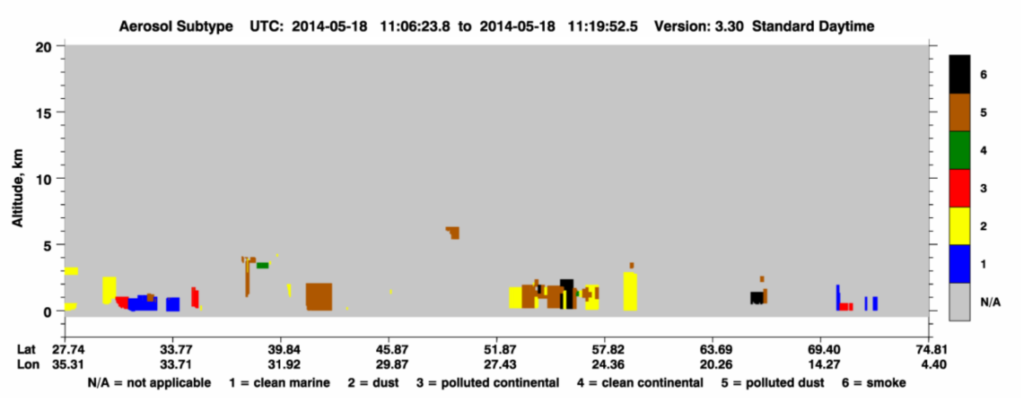
For the purposes of the TNA project, data collected since the beginning of the campaign were browsed through specific web sites: http://www.atm.helsinki.fi/smartSMEAR/ (SMEAR data), http://polly.rsd.tropos.de/lidar/ for the EARLINET lidar data and http://www.archive.arm.gov/discovery/#v/home/s/ for ARM2 facility data.

Looking to these data, cases with free troposphere aerosol layer intruding into the PBL were searched. About 20 days with FT layers were found in the PollyXT dataset. A peculiar case is observed in 18-20 May 2014 period.

In this period a layer above 2 km is observed, and then there seems to be an increase of the PBL top and a potential intrusion of the lofted (potentially transported) aerosol into the PBL. This layer seems also to be depolarizing. The SDS-WAS system reports for this case an arrival of Saharan dust particles over North-Eastern Europe and for some of the models of the SDS-WAS system even over Southern Finland. Also BSC -DREAM, which seems to not predict dust over Finland, reports a faint desert dust layer over Finland. BSC-DREAM forecast of dust vertical profile over Linkoping (Southern Finland) for 18 May, accounts for the arrival of some dust in the 2-4 km which is the altitude range of the FT layer observed at Hyytiala on the same day. The analysis of the Hysplit backtrajectories furthermore confirms this. Satellites also capture this event. CALIPSO sees a layer up to 3km on 18 May night and then on daytime in the Finland area. These layers are depolarizing layers and are identified as polluted dust and a mixture of dust/polluted dust/biomass burning.



CALIPSO attenuated backscatter at 532 nm as observed on 18 May 2014 for the track passing also over Finland.



CALIPSO aerosol mask for observation done on 18 May 2014 for the track passing also over Finland.

Dust events are very rare on Finland; the arrival from the Eastern Africa is even rarer. So that this is an almost rare case for studying dust travelling the Eastern Sahara - Southern Finland path. The potential intrusion into the PBL makes this event more interesting for the possibility of linking vertical profiling measurements to measurements at the ground.

The 18-19 May case has been selected as test case for both Task **a** and **b.**

- **Preliminary results and conclusions**

<u>*Task a*</u>

During the visit to BAECC, PollyXT and HSRL data were made compliant with EARLINET database netCDF format, through the following system-specific steps:

PollyXT

- raw signals have been formatted accordingly to the Single Calculus Chain (SCC hereafter) requirements
- system has been inserted into the EARLINET HoI
- data have been processed through the SCC (pre defined time slots of 1 h)

HSRL

- Data are available through ARM data discovery tool at a resolution of 30 s 30 m
- Data were processed through the online tool developed by Ed Eloranta (time slots of 1h, set by the user)
- data have been converted in the EARLINET format through a devoted MatLab routine

These activities were carried out thanks to the cooperation with respective data originators. In particular, Polly- XT was installed in Hyytiala with another TNA project (Ronny Engelmann), few months before this TNA. For including the system into the SCC acceptable systems, the characterization of the instrument after the installation at BAECC was needed and provided by dr Engelmann. The interpretation of Polly XT data in addition is always discussed with Dr. Engelmann. For the use, analysis and formatting of ARM-2 data, the presence of AMR2 scientists on-site was not needed. On-line discussion and exchange of information was sufficient for this.

These data are the base for the study of aerosol intrusion into the local PBL. HSRL provides aerosol extinction, backscatter and linear depolarization at 532 nm.

PollyXT is able to provide aerosol extinction at 355 and 532 nm, aerosol backscatter at 355, 532 and 1064 nm and the linear depolarization at 532. In daytime conditions, however PollyXT performances are limited because of the low SNR of the Raman channels. In these conditions, only backscatter profiles retrieved by Klett/iterative method are obtained (i.e. with an assumption regarding the aerosol extinction-to-backscatter ratio, a quantity related to the aerosol type, dimension and hygroscopicity).

Harmonization of the HSRL and PollyXT data is not limited only to format issue. The aerosol optical properties has obtained by the 2 instruments have been compared, post-processed in a similar way and finally combined for gaining a better insight of the aerosol properties.

Task b

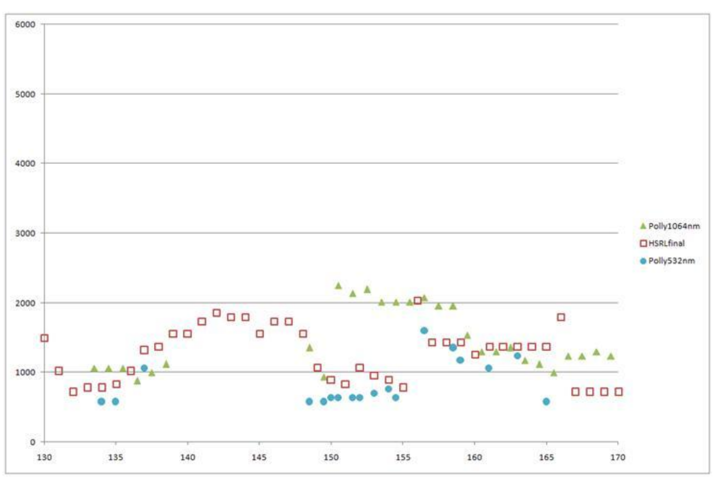
PBL retrieval

EARLINET data typically reports information about the top of BL+residual layer in the netCDF file in an appropriate Variable. This quantity is particularly important for climatological studies (Matthias et al., JGR, 2004; Mona et al., in prep. 2015), but it is also really interesting for air quality purposes and case studies (e.g. Mona et al., ACP 2012).

An operative definition agreed within EARLINET reports that the top of BL+residual layer (PBL herein after) corresponds to the first significant minimum starting from the ground in the aerosol backscatter derivative. The evaluation of the PBL top is not yet implemented within the SCC and it is leave to the responsibility of each PI. At network level, a working group is dealing with this issue and potential links to meteorological communities. On the other side, operational techniques and different derivative thresholds are under testing for the PBL evaluation.

An algorithm for PBL retrieval in line with EARLINET definition and standard has been applied to both HSRL and PollyXT data.

The aerosol backscatter profile at 532nm obtained through the elastic/Raman technique could reduce this PollyXT limitation. This is because the elastic/Raman technique makes use of a ratio of the two signals, and almost completely correct for the geometrical factor of the overlap. A very good agreement is found for the PollyXT and HSRL values in this configuration.

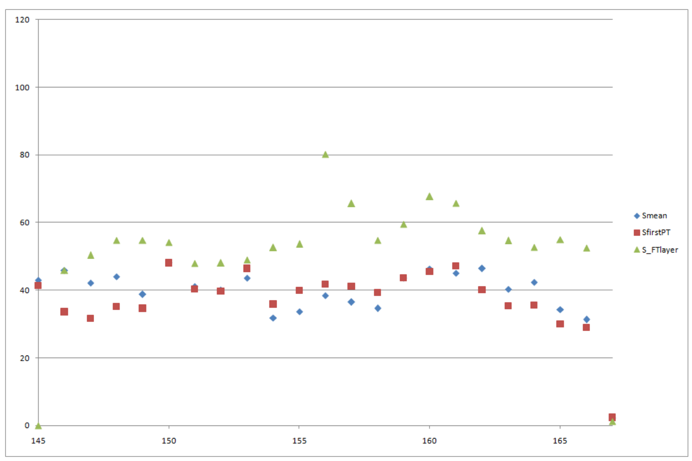


PBL top height as calculated starting from HSRL and PollyXT profiles.

Typical diurnal evolution of the PBL is detected by HSRL the on May 18.
On 19 May, the PBL evolution is disturbed by the presence of a close by layer just above the PBL. Around 02 UT on 19 May, the PBL top had a sudden increase from 0.9 km to 2 km. The 1064nm identification (not reliable as PBL, but indicative of a structure) shows the presence of a layer extending above the PBL up to 2 km (see also RCS figure reported above).

Optical properties in the PBL

The PBL IB and AOD suddenly increase during the intrusion into the PBL. These quantities depend on backscatter and extinction on the aerosol content and on aerosol properties, with backscatter more sensitive than extinction on aerosol size and extinction on absorbing properties. Ratios of extensive properties as backscatter and extinction coefficient are instead independent on the aerosol load and can provide directly information about the aerosol properties and are more important for aerosol typing. An important quantity is the lidar ratio S: the ratio between aerosol extinction and backscatter at the same wavelength.



lidar ratio S at 532 nm for pbl (blue diamond), first point of the profile from the ground (red square) and FT layer (green triangle).

Before the intrusion, the mean lidar ratio at 532 nm (HSRL) is almost constant inside the pbl as demonstrated by the good agreement between first point in the profile and pbl averaged values. Values are around 35 sr in 154-158 time period and increase to about 40-50 sr right after the intrusion. These values are in agreement with values typically observed for desert dust particles over continental Europe (e.g. Papayannis et al., 2008). Then the S temporal behavior follows the one of FT layer with a slight decrease.

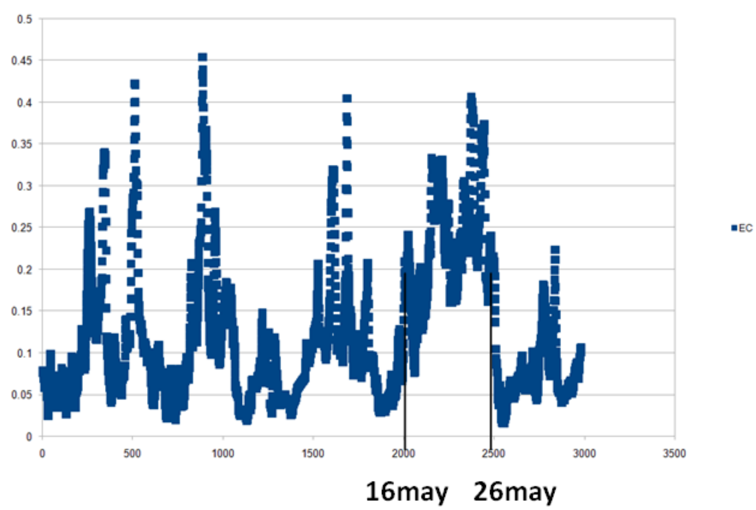
Observation at the surface

The second objective of SACS is the integration of lidar and at ground measurements. Lacking of knowledge about in situ data, the PI improved the situation during her staying in Hyytiala through a constant and fruitful discussion with hosting people. She visited the facility through a detailed tour. She discussed technical and scientific details about ACSM measurements and analysis, really important for the comprehension of physical meaning of these measurements. Relevant papers were provided to dr Mona for adding valuable information about this topic. All this knowledge will foster on one hand the integration with vertical profiles measurements acquired at BAECC and on the other hand the adding of ACSM instrument and capability at the PI's institute.

In this respect, ACSM data are of particular interest, first of all because ACSM provides continuously and automatically information about principal aerosol components concentrations, relevant information for the aerosol typing. Moreover, ACSM operation and use has been strongly improved within the ACTRIS infrastructure, even through a dedicated intercomparison campaign. As a result, ACSM distribution around Europe is increasing. The integration of this instrument data with lidar data can be regarded as a big step in the long way of remote sensing-at ground aerosol measurements integration.

ACSM provides the concentration in microg/m3 of 5 aerosol components: Organic Aerosol, SO4, NH4, CHCl and NO3. Information about EC Elemental Carbon are obtained by Ethalometer. Aerosol distribution is provided by DMPS Differential Mobility Particle Sizer. Impactor DEKATI PM-10 data provided typically daily PM10 concentration, but the comparison with these data is complex because just 1 sample from 16 to 19 May was collected.

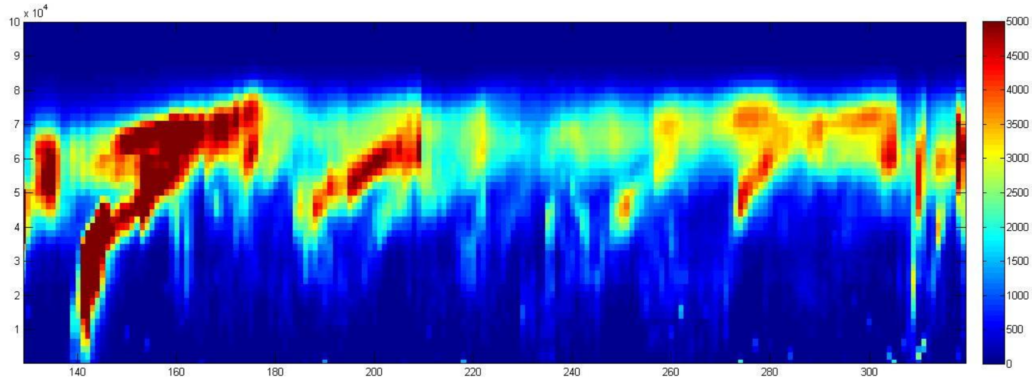
A big increase in the EC is observed in the April- May time series. For April-June period, the EC content is on average 0.12 mg/m3. There is some increment in the temporal behavior up to about 0.14 mg/m3. A big unusual increase up to 0.22 mg/m3 is observed for the 16-26 May period. This period corresponds to the Saharan dust arrival over Finland.



EC concentration in mg/m3 as a function of the sample.
Sample 1 is recorded on 1April, 02:19. each sample roughly corresponds to 30 sec.

An increase of Organic, So4, NO3 and EC components was found on 18-19 May just right after the intrusion time identified by lidar data analysis started at 05 UT May. It is important to comment here that a 2km high PBL was identified for lidar at that hour, so certainly mixing and deposition time is expected. This links the increase of these 4 components to the arrival of free troposphere and long-rage transported aerosols into the PBL. Further analysis on particles volatility properties could further improve the knowledge about this case and support the Saharan dust origin of observed particles.

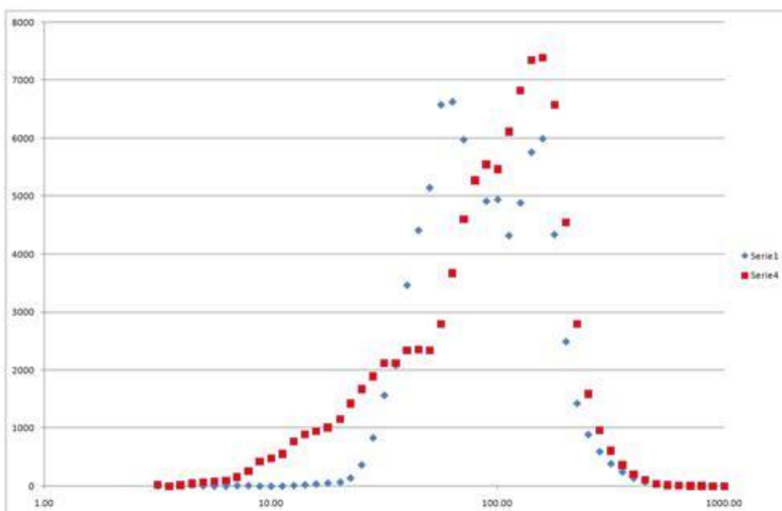
For the same event, a typical banana is observed in the mobility particle size distribution (DMPS measurements). In particular a decrease in the mean diameter is observed at the intrusion time followed by an increase of the particles dimension.



concentration of particles (color code) as a function of the time for different particle mobility diameter (y-axis) as measured by DPMS

Around 00UT, 18 May (140 hours since May 12, 16UT), a new mode of very small particles appears, and the 100 nm mode accounts for more particles. Passing the time, the two modes become equivalent in concentration and the middle diameter range is more populated. This could indicate aggregation processes. Then there is a shift towards diameter close to 100 nm.

Toward the end of this new particles formation process, the dust layer intrusion in the PBL occurred (started at hour 157th). The particle distributions just before and after the intrusion starting show that the 50 nm mode is significant reduced incrementing the 100 nm mode relevance (still aggregation processes). The arrival of particles in the 5-30 nm range is also observed.



Particle distributions just before (blue) and after (red) the intrusion starting.

This temporal behavior of the size distribution of particles collected at ground level could give a better insight of the modification in optical properties as observed by lidar. In this respect it is important to underline that both DMPS is limited to 1μm as maximum particle diameter. The case study under investigation is related to Saharan dust intrusion into the Hyytiala PBL, and Saharan dust particles typically account for large particles (1- 50 μm). However recent studies show that also Pm1 measurements are affected by the presence of Saharan dust particles (e.g. Boselli et al., 2012, Chen et al., ACP 2011), with also smaller mode around 0.1 μm (e.g. Chen et al., 2011).

- **Outcome and future studies**

Lidar-based inversion code will be used for investigating the content of larger particle and the impact of Saharan dust on observed particle distribution at the ground. Microphysical inversion with Veselovskii code using a combination of HSRL and Polly optical properties dataset would give information about size distribution into the FT aerosol layer identified into lidar observations and in the PBL after the lofted layer intrusion. In addition, on long-term, the main results of the TNA project will be the provision of ARM2 and Polly XT lidar data in a standardized way and the definition of start-up strategy for in situ-remote sensing aerosol observations synergistic approach .

- **References**

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