Deliverable D12.6: Final report on the comparison and suitability of the different techniques to measure the aerosol particle fluxes at the ACTRIS-2 sites

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One of the main aims of the joint research activity in WP12 was to combine tower-based in-situ and ground-based remote sensing observations of aerosol particle fluxes to understand the horizontal scales over which each observation type is representative, and provide recommendations for measuring aerosol fluxes at an ecosystem level within a European infrastructure network. The methods for tower-based particle flux observations have been better established than those for remote sensing. In the latter, vertical profiles of particle fluxes in planetary boundary layer (PBL) can be determined from co-located Doppler and aerosol lidars by combining the turbulent vertical-wind component derived from the first instrument with aerosol variance and microphysical properties obtained from the second. During ACTRIS-2, such co-located measurements have been carried out in terms of field campaigns at Pallas (FMI, Finland) and Košetice (Czech Republic), in addition to Hyytiälä (Finland), Cabauw (the Netherlands), and AGORA (Spain), where the requirement for horizontal homogeneity for flux measurements and the possibility to compare with in-situ measurements are met. Alternatively, a method using only the Doppler lidar in its fast-response mode together with an aerosol lidar to provide mean statistics of the aerosols is examined.

This document describes comparisons so far made between the different techniques and provides recommendations for such measurements to be conducted within the European infrastructure ACTRIS.

Measurement campaigns

The team from Univ. of Granada carried out several campaigns in order to measure and compare in-situ and remote sensing particle fluxes. During these campaigns, measurements were gathered using similar setups. The Eddy Covariance (EC) setup consisted of a sonic anemometer (81000 RM-Young) for measuring 3D wind speed and direction and a condensation particle counter (TSI3776) for measuring particle number concentrations in the range 2.5 nm - 3 µm. Both instruments measured with a sampling frequency of 10 Hz. Remote sensing setup consisted of aerosol and wind profiles measured by Aerosol and Doppler Lidar instruments for retrieving vertical aerosol flux profiles. Elastic Lidar instrument was the multiwavelength Raman lidar MULHACEN with detection at 355, 387, 408, 532, 607 and 1064 nm (Raman channels only at night-time) with a temporal and spatial resolution of 2 s and 7.5 m, respectively. Doppler Lidar is a (Halo Photonics) model Stream Line operates at 1.5µm with a temporal and spatial resolution of 2s and 30 m, respectively. The campaigns have been

1. AMAPOLA (Jaén, Spain, 18-29 Apr 2016): AMAPOLA campaign was held in a rural area in an extended highly uniform olive grove site. Halo Streamline Doppler Lidar and the aerosol lidar VELETA were separated by an approximate distance of 2 m, and they were measuring with ~0.5Hz. Simultaneously, tower-based particle flux measurements were conducted in a 10 m height tower around 5 m apart from the remote sensing instruments.
2. AGORA: continuous measurements of tower-based particle flux measurements were conducted at Granada from June 2016 to May 2018 in a 50 m height tower around 250 m apart from the remote sensing instruments. The anemometer and the CPC inlet were located on top of the tower. The CPC was located 10 m below, in a shelter equipped with a Peltier cooling system. Intensive measurement campaigns (apart of routine measurements) of remote sensing instrumentation were conducted (SLOPE I, May-Aug 2016; SLOPE II, Jun-Aug 2017).
3. POLIMOS (Rzecin site, Poland, June-September 2018): This measurement campaign was performed in an undisturbed peatland with a floating carpet of mosses in western Poland. Similar setup was used, and EC was around 450 m apart from the remote sensing instruments.

In Hyytiälä (Finland), concurrent lidar and tower-based fluxes were measured during the BAECC (Biogenic Aerosols—Effects on Clouds and Climate) campaign in spring 2014. During the campaign, co-located Halo Streamline Doppler Lidar and PollyXT aerosol lidar measurements were conducted at the same time as continuous tower-based aerosol particle fluxes.

In Košetice (Czech Republic), a particle flux measurement campaign using both in-situ and remote sensing techniques was conducted from 15 Aug to 15 Sep 2017 using co-located Halo Streamline Doppler lidar and PollyXT multi-wavelength lidar together with EC measurements made on the tower at a height of 80 meters above ground. The setup comprised a Gill ultrasonic anemometer and CPC (TSI3775) measuring with a frequency 1 Hz. In addition, an SMPS and APS measurement were set up at the tower platform at 230 m. PollyXT was modified especially for this campaign in a way that the original photon-counting near-range detector was replaced by an analogue detector which gave the opportunity to measure lidar backscatter profiles with a high temporal resolution of 5 s. An example of the derived particle backscatter coefficient is given in Fig. 1. On the afternoon of 23 Aug the PBL extended up to 1.3 km height. From 12-16 UTC the convective mixing can be observed, especially at the entrainment region between the PBL and the free troposphere. Such data are available for the entire period of the campaign. PollyXT was operational from 14 Aug – 29 Oct 2017. However, it was found that the aerosol optical depth and the particle mass concentration was generally low during the campaign.

The Doppler lidar was installed directly at the base of the tower from 16 August – 20 September. Fig. 2 (bottom) shows the measurement of the vertical wind fluctuations. It can be seen that the convective mixing in the PBL started around 8 UTC and calmed down around 16 UTC, in accordance with the PollyXT measurements. Apart from the vertical-wind measurements the Doppler lidar was partly operated in scanning mode (white stripes of Figure 3) in order to derive the horizontal wind and the scales of turbulence as well.

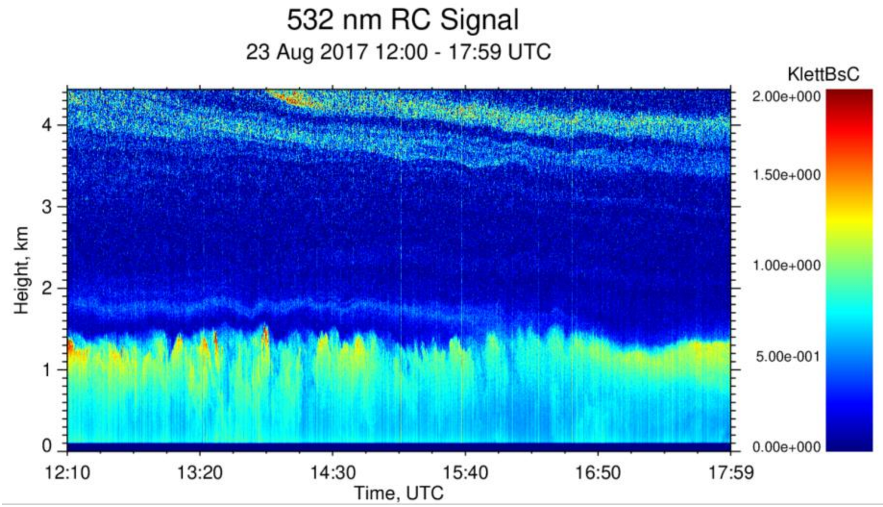

Figure 1: Particle backscatter coefficient at 532 nm derived from the newly installed analogue detector with the Klett-method for Polly XT operating at Košetice on 23 Aug 2017. The temporal and spatial resolution is 5 s and 7.5 m respectively.

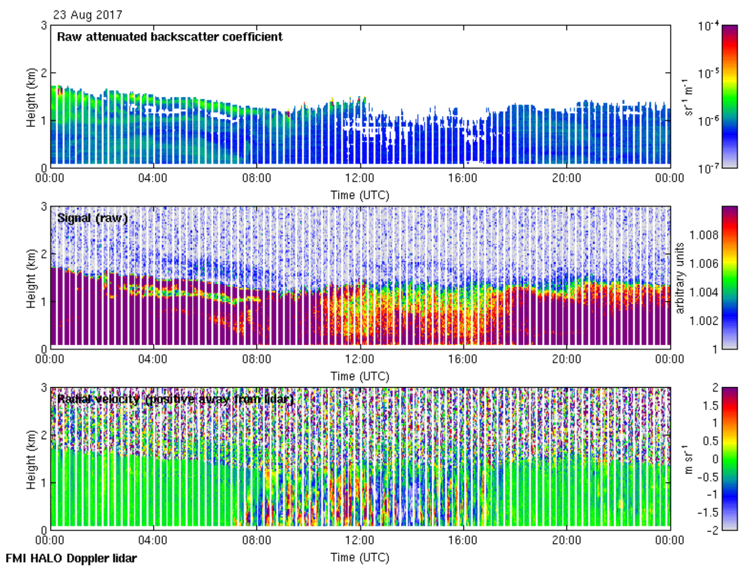

Figure 2: Measurement of the attenuated backscatter at 1.5 µm, signal, and vertical velocity observed from the Doppler lidar at Košetice on 23 Aug 2017.

PollyXT measured 3β+2α+2δ multi-wavelength optical products in addition to the high-resolution fluctuations of the backscatter coefficient at 532 nm (Fig. 3). The lidar ratios of 49-50 sr, an Angström exponent >1.5, and the low depolarization ratios <4% indicate the presence of small, non-absorbing continental background aerosols. The optical depth of the PBL as estimated from the integrated extinction at 532 nm was as low as 0.07. For such aerosols a typical conversion factor of 10-15 (μg/m³) / ($Mm^{-1}sr^{-1}$) can be assumed for the calculation of the particle mass from the aerosol backscatter coefficient at 532 nm.

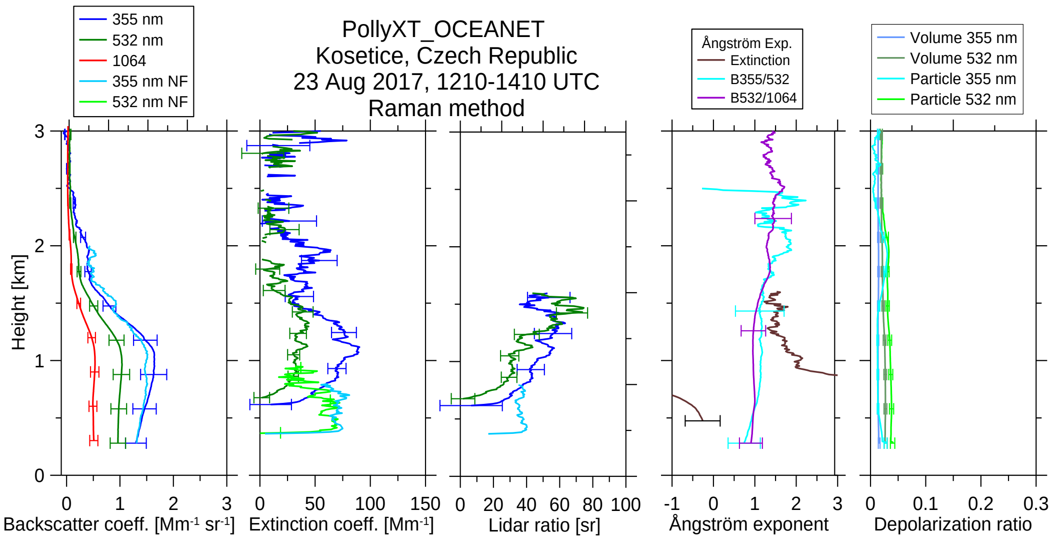

Figure 3: Multiwavelength optical products derived from PollyXT on 23 Aug 2017.

Co-located vertical flux measurements were conducted during the CINDI-2 campaign in Cabauw, the Netherlands, in Autumn 2016. Although the Zephir 300 instrument provided wind information, unfortunately this continuous wave lidar was not able to provide wind measurements at sufficient frequency for flux measurements. Thus, comparison of the techniques is not possible at this site.

At Pallas (Finland), simultaneous particle flux measurements using both *in-situ* and remote sensing techniques were conducted at the Kenttärova station during late autumn of 2016 as part of the PACE (*Pallas Cloud Experiment*) campaign. For boundary layer fluxes, a Halo Streamline Doppler Lidar and PollyXT multi-wavelength Raman aerosol lidar were operated during the campaign. Low total particle number concentrations and problems with data stream synchronization has proven to be challenging with the present setup. At this Artic site, the challenge in both in-situ and remote-sensing derived particle fluxes is the clean air causing low counts obtained with the in-situ instrumentation and very weak lidar signals. Since the measurement uncertainty for both Doppler velocity and attenuated backscatter is directly related to the signal-to-noise ratio (SNR), this results in large measurement uncertainties. Usually, this issue would be solved by using long integration times, but this would not be appropriate for measuring fluxes. This was furthermore complicated by the shallow boundary layer during the campaign causing issues in the minimum range that each instrument can obtain reliable data from. There is usually no useful Doppler lidar signal outside the boundary layer, so the challenge is to make aerosol lidar measurements within a shallow boundary layer that may be less than 300 m deep.

Results

Granada

The analysis was started from the SLOPE II campaign data using only Doppler lidar and in-situ measurements. Particle number fluxes from the in-situ measurements, were calculated using EddyPro software. Maximum covariance technique was used for the correction of the lag time between the time series of vertical wind and particle number concentration registered. Two trigonometric rotations were applied to the wind components to cancel out the mean vertical wind velocity. Linear de-trending filter and de-spiking were then applied to the concentration and temperature signals. After this processing, the aerosol fluxes were calculated over 30 min periods._In order to compare these fluxes with the remote-sensing derived fluxes, as a starting point only Doppler lidar data were used. After some instrumental corrections, attenuated backscatter coefficient (β_{att}) profiles with the same frequency as vertical velocity were obtained and the covariances of these properties were calculated. An example of the temporal evolution of such profiles is shown in Fig. 4 for 14th Jun 2017.

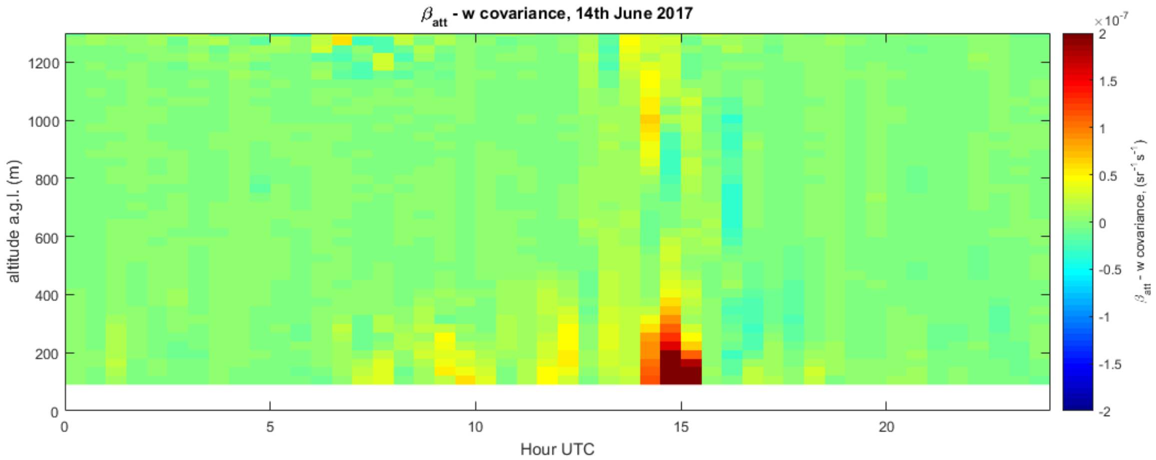

Figure 4: β_{att}-w covariance profiles obtained with Doppler lidar during SLOPE on 14th Jun 2017.

In Fig. 5, the time series of β_{att}-*w* covariance (named as β_{att}-flux) from lidar at the first available altitude (105 m) for the same day is compared with the time series of particle concentration flux from in-situ (at 50 m). Figure 6 shows the complete time series and Fig. 7 correlation between the two fluxes for June 2017. Some clear dependence can be detected but more detailed statistical analysis (e.g. linear de-trending or de-spiking) must be included on remote sensing technique, and more filters must be applied in order to compare only data that are suitable for flux calculations (until now, all Doppler lidar data are used). The slope of the linear fit is $(4.3 \pm 0.9) \cdot 10^{-17}$ m^2 sr^{-1}. The challenges of the comparison include

- Different magnitudes are calculated: particle concentration (in-situ) vs particle backscatter coefficient (remote sensing).
- Only a preliminary statistical approach with remote sensing measurements was done (more statistical corrections have to be applied in order to remove data that are not suitable for flux calculations).
- Different measurement frequencies: 10 Hz (in-situ) vs 0.5 Hz (remote sensing).
- Different altitudes were compared, although they are as close as possible and likely in the constant flux layer where the fluxes should only vary for 10 % in maximum.

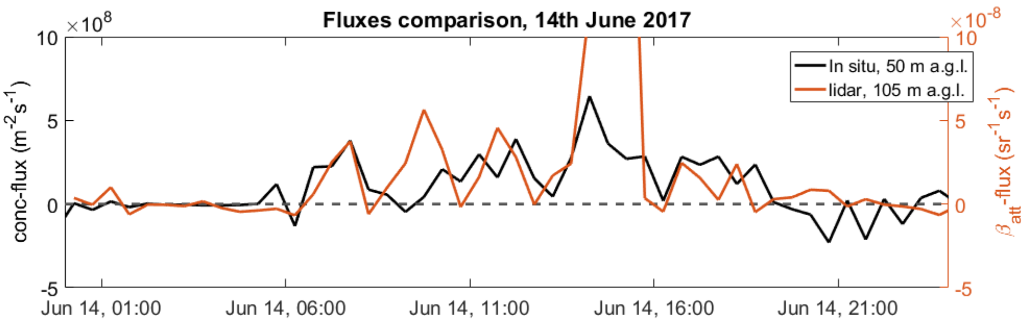

Figure 5: β_{att}-w covariance time series obtained with Doppler lidar for 14th June 2017, and particle concentration flux time series from in-situ measurements.

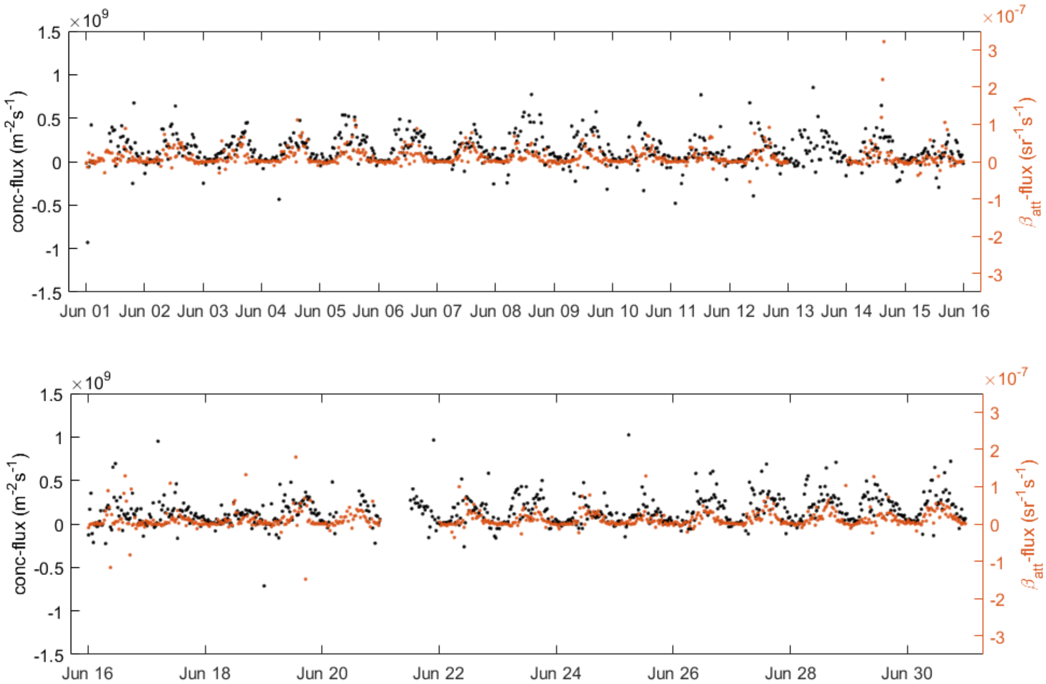

Figure 6: β_{att}-w covariance time series obtained with Doppler lidar for Jun 2017, and particle concentration flux time series from in-situ measurements.

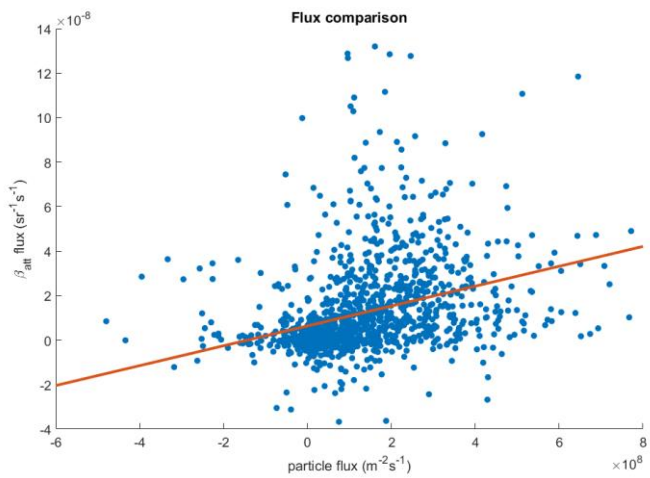

Figure 7: Scatter plot of β_{att}-w covariance obtained with Doppler lidar for Jun 2017, versus particle concentration flux from in-situ measurements.

The mean daily evolution of in-situ and remote-sensing derived particle fluxes and for the different vertical velocity moments (i.e., variance, skewness and kurtosis) using all data from June 2017 are shown in Fig. 8. The diurnal evolution of the particle fluxes follow each other well despite the challenges listed above. The surface particle flux (in-situ) has slightly wider shape which could be due to several reasons including smaller measured particles, or smaller turbulent length scales close to the surface. There are more variability between the vertical velocity moments between the two methods which can originate for example from the different measurement frequencies.

The next steps are deeper statistical analysis of Doppler lidar signal, in order to apply more filters and corrections to select data that are suitable for flux calculations and including aerosol lidar + Doppler lidar data at different wavelengths to the analysis

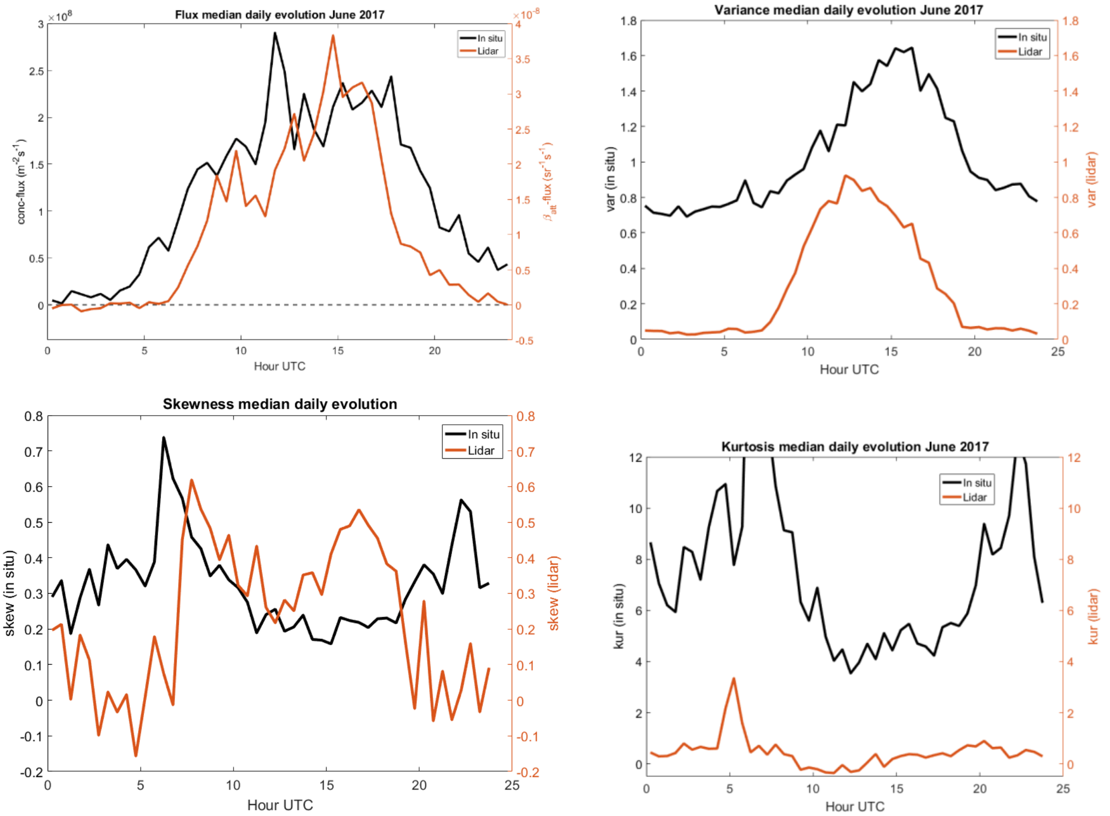

Figure 8: Scatter plot of β_{att}-w covariance obtained with Doppler lidar for June 2017, versus particle concentration flux from in-situ measurements. In situ data correspond to measurements at 60 m over the surface while for the lidar data we are using the Doppler lidar bin at 105 m over the station, ~130 m over the surface.

Hyytiälä

The first step in the comparisons of the remote-sensing and tower-based techniques has been to compare the Doppler lidar and EC derived turbulence data and dissipation rate. EC data from five different levels was post-processed using approaches similar as used within ICOS.

For the Doppler lidar, the velocity fluctuations were calculated from SNR (O'Connor et al. 2010) and post-processed to correct for the background (Manninen et al. 2016) (Fig. 9). Figure 10 shows the behavior of dissipation rate on 13 July 2016. One can see good correspondence in the dissipation rate with both techniques and the onset and decease of turbulence occurs in the same way.

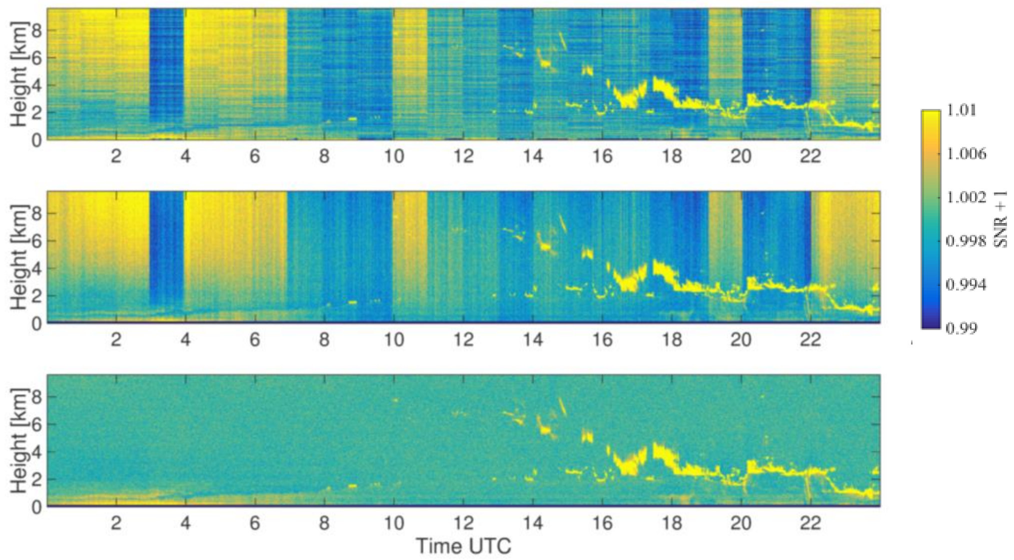

Figure 9. Signal to noise ratio (SNR) from Doppler lidar in Hyytiälä on 13 Jul 2016.

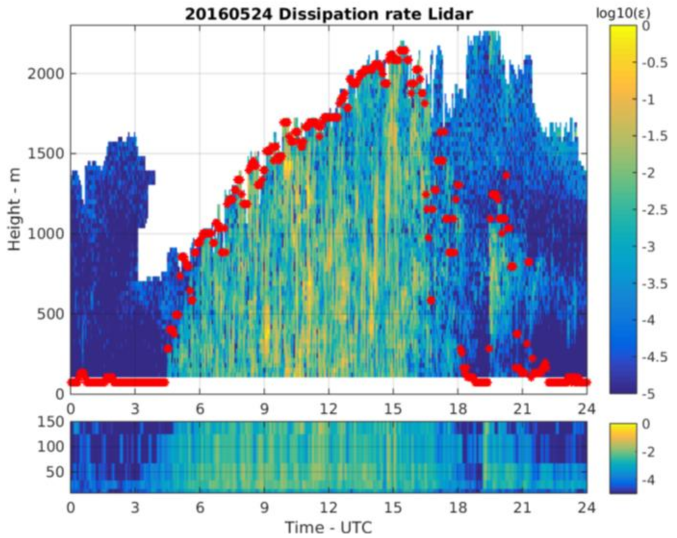

Figure 10. Time-height plots of dissipation rate derived from Doppler lidar (above) and tower-based measurements (below) at Hyytiälä on 13 Jul 2016.

Now that we know that the turbulence characteristics compare well, the next step is to extend the analysis to particle fluxes derived from the β_{att}-w covariance using the Doppler lidar in its fast-response mode. During the initial intercomparison campaign the β_{att}-w covariance was calculated using the Doppler lidar data and an example shown in Fig. 11. It is clear that the clean air environment at Hyytiälä is challenging for the Doppler lidar, and although velocity information appears reliable, the signal (and hence attenuated backscatter) is weak with the Doppler lidar unable to reliably reach the top of the boundary layer.

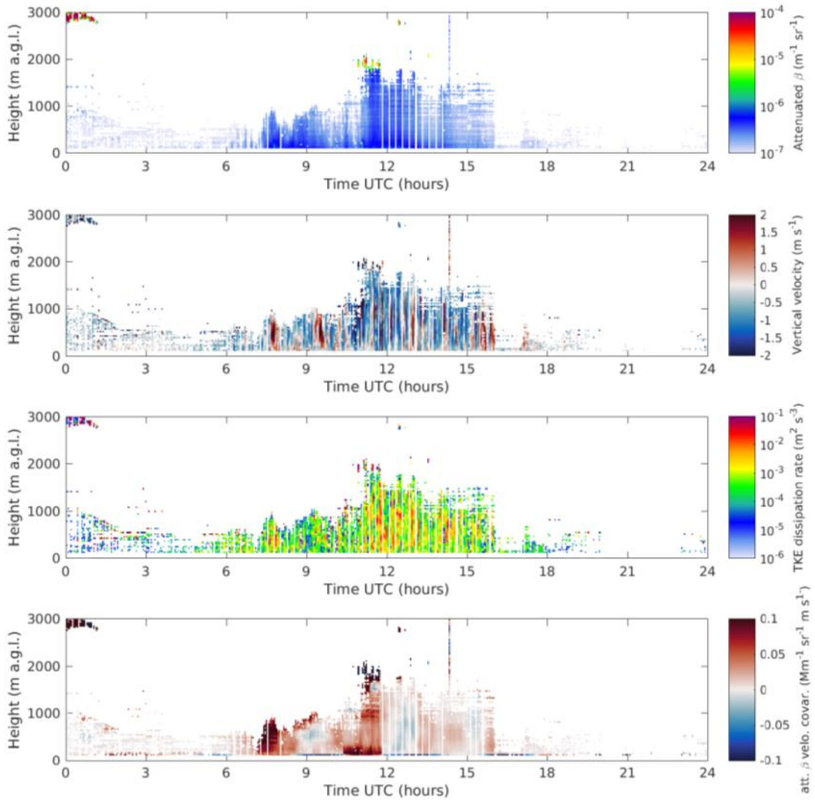

Figure 11. Time-height plots of attenuated backscatter, velocity, dissipation rate and βatt-w covariance at Hyytiälä on 22nd May 2014.

Unfortunately, the particular Halo Streamline instrument operating at this time was not recording the background files necessary for full Halo Doppler lidar post-processing as described in Vakkari et al. (2018). Calculation of dissipation rate and β_{att}-*w* covariance is only possible during certain periods, and the uncertainty in β_{att}-*w* covariance particularly is rather high.

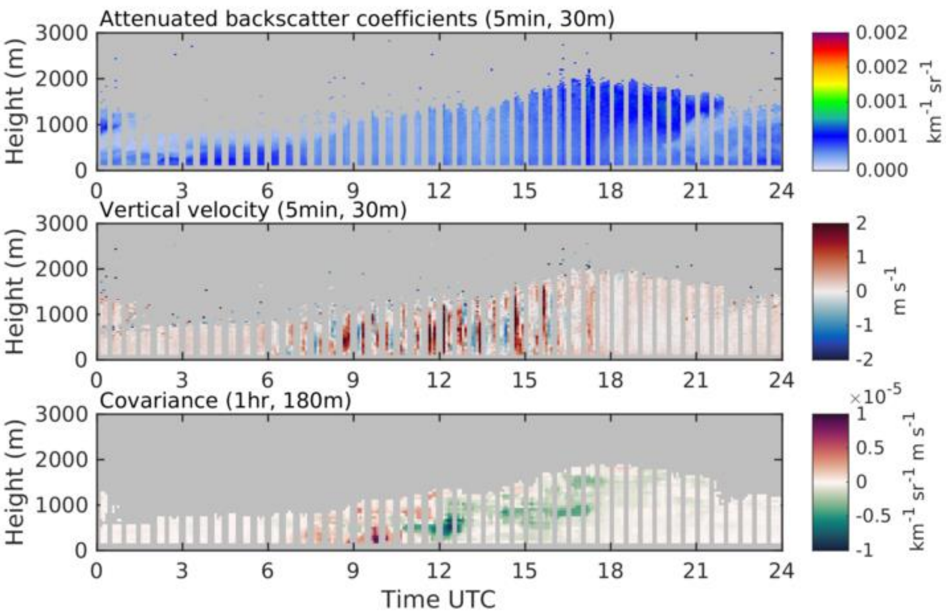

Figure 12. Time-height plots of attenuated backscatter, velocity, and βatt-w covariance at Hyytiälä on 7th May 2016.

Figure 12 shows an example from a later date at Hyytiälä when the background Doppler lidar files were also stored; the surface appears to act as a source during the morning and then as a sink during midday and afternoon. However, in general at Hyytiälä, the clean air still poses a challenge even when the full post-processing can be performed.

Kosetice

In order to derive the aerosol mass flux from lidar in the next step the covariance from the data of both lidars was calculated. For this process both datasets had to be resampled to the same time and height grids (it was also found by the correlation analysis that the time stamps of the Halo Doppler lidar were out by approximately a minute because the system was not connected to a time server). Figure 13 shows such a prepared dataset. The arrows show how vertical motions actually transported cleaner air parcels from the surface towards higher altitudes and more polluted air downwards. This observation overall points towards a downward turbulent flux during this period.

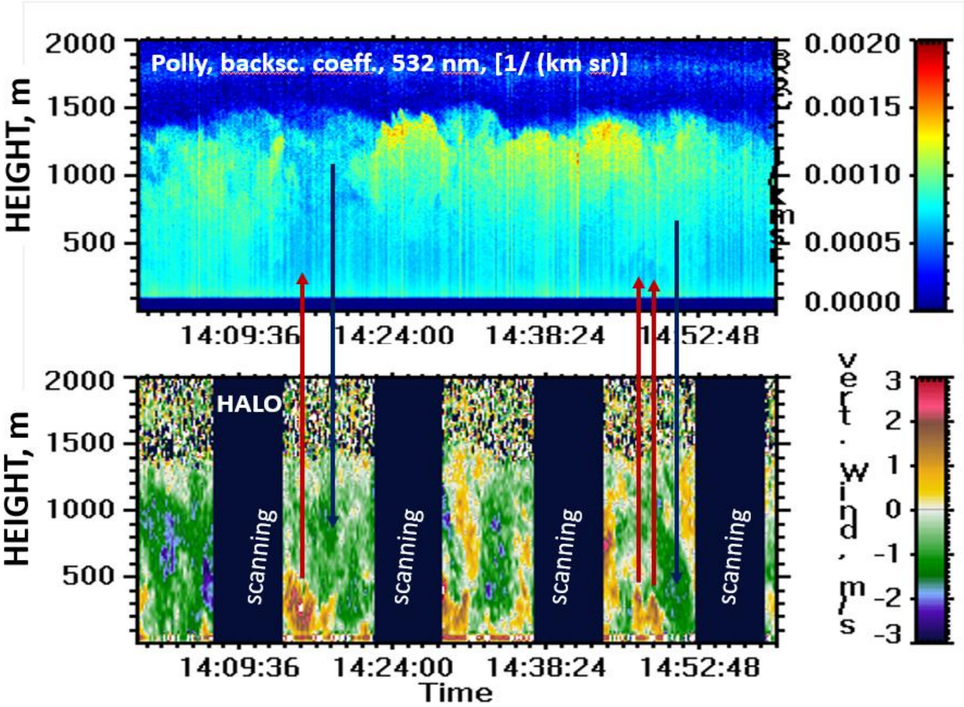

Figure 13: Synchronized dataset of PollyXT data (top) and HALO Doppler lidar (bottom).

In the last step the "backscatter-flux" profiles were calculated and converted to mass flux profiles with the assumption that the particle population remained the same during the entire period. Figure 14 shows the resulting profiles for three different time periods. Between 13-15 UTC an almost linear flux profile as indicated by the line between the surface flux and the entrainment flux can be seen (apart from surface values which might be erroneous because the two lidars were separated by a distance in the scale of the typical eddy size at low altitudes). The linear line indicates the negative surface flux which was mentioned in the previous paragraph. Later during the day and with the decaying activity of the convection the turbulent flux in the boundary layer decreased as well.

At the same time the EC derived surface particle fluxes were small and directed upward on the first two time slots and downward in 16:30-18:00. The EC fluxes are so small that they are close to the detection limit of the measurement setup and thus comparisons between the different techniques is challenging at this site.

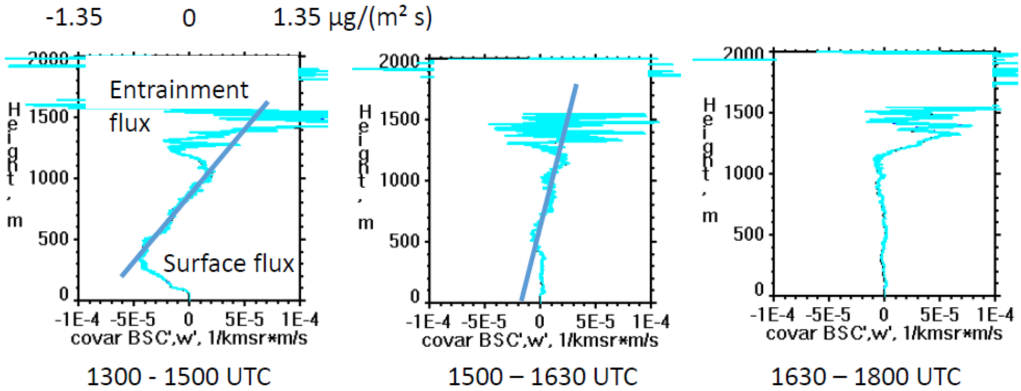

Figure 14. Profiles of the vertical aerosol mass flux from lidar measurements in Košetice on 23 August 2018 for three different periods. The top scale shows the estimated conversion from the "backscatter-flux" to the more meaningful particle mass flux.

The following remarks can be given: The fact that PollyXT is tilted by 5° and because of the horizontal displacements of both instruments they did not necessarily always observe the same air parcels. Hence there might be the effect that the smallest turbulent eddies are missed. However, the bigger ones with scales of the order of the actual measurement height are the main contribution to the turbulent flux at greater altitudes. Therefore, in the region of the entrainment between the PBL and the free troposphere were usually the largest flux values are measured the errors should be smaller than at the surface were most of the fluxes are generated on eddy sizes of 50 m or less. Thus care should be taken when looking at the flux profiles from remote-sensing closer to the ground <200-500 m for this campaign.
Unfortunately, the aerosol optical depth during the campaign was often very low, so that even the Doppler lidar was not always able (especially during the convective time of the day) to fully resolve the vertical winds in the entire PBL; the centre panel in Fig. 2 shows the signal falling below 1.01 (equivalent to -20 dB).

Conclusions

The aim of Task 12.3 was to compare and provide recommendations for aerosol particle fluxes obtained with remote-sensing and in-situ techniques. For this purpose, measurement campaign using co-located Doppler and aerosol lidars to provide aerosol particle fluxes in the planetary boundary layer simultaneously with tower-based aerosol particle fluxes above different ecosystems.

Both remote-sensing and in-situ techniques for measuring aerosol particle fluxes find clean environments very challenging, at least with the available technologies used in ACTRIS-2. Thus, if such measurements are planned in clean environments, such as arctic areas, particular attention should be paid to the range and sensitivity of the instruments. In general, the measurement campaigns and work performed in WP12 have shown how both in-situ and remote-sensing observations need to be carefully designed; understanding the instrument performance is vital for obtaining successful aerosol particle fluxes in the boundary layer.

With tower-based observations one needs to make sure that the measurement location meets the criteria of the method employed. The measurement setup must be installed at an altitude sufficient enough for the effects of the surface to be considered 'blended' and the surface appears as homogeneous. In addition to this, for example in Hyytiälä, particle flux observations were started in new location, but the more detailed analysis has shown that this location is not suitable for aerosol particle fluxes due to the effects of the measurement tower itself. The type of particle counter used becomes particularly important in

more clean environments whereas in environments with large particle fluxes such as in urban areas, the counter used can be more robust. As an example, in Kosetice, direct comparisons between the in-situ and remote-sensing techniques is challenging as the surface particle fluxes are relatively small; more detailed analysis and filtering of the data needs to be made before any final conclusions on the comparability of the two techniques can be made.
However, as has been shown during these campaigns, realistic ecosystem-level surface particle flux observations and deposition velocities above different environments can be obtained. There is a manuscript in preparation where recommendations for the instrumentation and post-processing will be given if such measurements will be continuously done within ACTRIS.

So far, only preliminary analysis on remote-sensing derived aerosol particle fluxes have been performed, but based on these, the derived fluxes seem to correspond well with the tower-based aerosol particle fluxes. This is despite the challenges encountered, and the clear differences between the methods employed. Table 1 summarises the experiences in using Doppler and/or aerosol lidars to calculate particle fluxes in the BL. There is an advantage in using Doppler lidar only β-w covariances since these are obtained from the same volume at the same time, however much more effort must be made to characterise the β-value itself and provide the β-to-mass conversion appropriate for the Doppler lidar wavelength. There are additional uncertainties associated with the β-value obtained from Doppler lidar, compared to β_{part} obtained from aerosol lidars, and the Doppler lidar signals are often weaker than those obtained from aerosol lidars, again increasing the measurement uncertainty.

Table 1.Pros and cons of using Doppler or aerosol lidar

	Only Doppler	Doppler + Aerosol
Pros	Lower altitude retrieved (105 m a.g.l.) Implicit synchronization	β_{part} (without att) Possibility of approx. microphysics
Cons	Attenuation to be considered Focus correction must be applied	Overlap altitude ~800m a.g.l. Manual synchronization

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