Aerosol microphysical properties from inversion of tropospheric optical Raman lidar data

M. de Graaf, D. P. Donovan[1] and A. Apituley[2]

[1] *Royal Netherlands Meteorological Institute (KNMI), Wilhelminalaan 10, 3732 GK, De Bilt, The Netherlands (martin.de.graaf@knmi.nl, donovan@knmi.nl)*
[2] *National Institute for Public Health and the Environment (RIVM), Anthonie van Leeuwenhoeklaan 9, 3721 MA, Bilthoven, The Netherlands (arnoud.apituley@rivm.nl)*

ABSTRACT

The inversion of optical Raman lidar data, to retrieve microphysical aerosol properties, is challenging, since solutions depend non-linearly on measurement errors. However, the retrieval of integral properties of the aerosol size distribution, such as total volume or surface area density, depend much less on measurement errors than the size distribution itself, and can be predicted with useful accuracy using a Principle Component Analysis (PCA) based inversion technique.

The PCA kernels are sensitive to complex refractive index, which can be used to account for aerosol type variation in the optical data. This is especially important in the troposphere, where the refractive indices of the ambient aerosols are usually unknown. We show the accuracy that can be expected from this type of analysis.

PCA is a stable method, easy to implement and fast. It is therefore a useful complement to existing inversions. It can also be used as an extra constraint for other, more involved inversion techniques using regularisation and constraints.

1. INTRODUCTION

To obtain accurate detailed information about an aerosol size distribution,a finite number of extinction and backscatter measurements from multi-wavelength Raman backscatter lidars is generally not enough However, various integral properties of the aerosol size distribution such as aerosol total volume, surface area density and aerosol extinction and backscatter at various wavelengths can be predicted with useful accuracy using PCA. In general, PCA allows the prediction of aerosol physical properties without any restrictive assumption about the size distribution. However, the shape and refractive index of the particles in question must still be assumed. PCA has been successfully applied to stratospheric sulfate aerosol lidar measurements to retrieve aerosol effective radius, surface area, and volume profiles [1]. The method is here extended to include various tropospheric aerosol models, including water soluble aerosol, soot, and mineral dust, using the sensitivity of the PCA kernels to varying refractive index. The method is presented and applied to measurements from the ground-based Raman Lidar named 'Caeli'.

2. PRINCIPLE COMPONENT ANALYSIS

The Mie solutions of Maxwell's equations in the far field give the extinction and backscatter of a spherical scatterer at a given height and wavelength:

$$\alpha(\lambda) = \int_0^\infty \pi r^2 Q_{\alpha,\lambda}(r)\frac{dn(r)}{dr}dr, \quad (1)$$

and

$$\beta_\pi(\lambda) = \int_0^\infty \pi r^2 Q_{\beta_\pi,\lambda}(r)\frac{dn(r)}{dr}dr, \quad (2)$$

respectively, where r is the particle radius, $Q_{\alpha,\lambda}(r)$ is the extinction efficiency, $Q_{\beta_\pi,\lambda}(r)$ is the backscatter efficiency, and $dn(r)/dr$ is the aerosol size distribution. Equations (1) and (2) can be rewritten as

$$g_i = \int_0^\infty K_i^V(\lambda, r, m)\frac{dV(r)}{dr}dr, \quad (3)$$

where $dV(r)/dr$ is the aerosol volume size distribution, g_i is either the backscatter or extinction measurement and K_i^V is the appropriate extinction or backscatter volume kernel for wavelength λ_i. Similarly, the problem can be written in terms of the aerosol surface area distribution $dS(r)/dr$ using surface kernels K_i^S:

$$g_i = \int_0^\infty K_i^S(\lambda, r, m)\frac{dS(r)}{dr}dr. \quad (4)$$

Using an appropriate numerical quadrature rule eq. 3 can be discretised and inverted using a standard principle component analysis. The total volume vector then becomes

$$\bar{\mathbf{v}} \simeq \mathbf{K^t U L^{-1} U^t}\bar{\mathbf{g}}, \quad (5)$$

where $\mathbf{K}$ is the discretised volume kernel, $\mathbf{L}$ is a diagonal matrix containing the eigenvalues of a matrix $\mathbf{C}$ (so

© Proceedings of the 8th International Symposium on Tropospheric Profiling, ISBN 978-90-6960-233-2 Delft, The Netherlands, October 2009. Editors, A. Apituley, H.W.J. Russchenberg, W.A.A. Monna

defined that $C_{ij} = \sum_k K_{ik} K_{kj}$) and $\mathbf{U}$ is the matrix containing the corresponding eigenvectors as its columns. Details can be found in [1]. Any other integral property of the aerosol can now be estimated using

$$P \simeq \bar{\mathbf{w}}^{\mathbf{t}} \mathbf{K}^{\mathbf{t}} \mathbf{U} \mathbf{L}^{-1} \mathbf{U}^{\mathbf{t}} \cdot \bar{\mathbf{g}} \tag{6}$$

and weighting functions $\bar{\mathbf{w}}$ (e.g. $w_j = 1$ for volume and $w_j = 3/r_j$ for surface). The solutions can also be expressed in terms of surface kernels and appropriate weighting functions.

2.1. Kernel dependence on Refractive index

In general, the results will depend on the complex refractive index m of the measured aerosol. The kernels of eq. 3 contain information on the refractive index, which can be used in the PCA, see Fig. 1. Kernels for four different types of aerosols are shown, which span a large range in refractive index. The kernels were smoothed by choosing a lognormal aerosol size distribution with a small mode width $\sigma = 1.1$ to reduce the small scale oscillations of the kernels, while retaining the sensitivity of the PCA. Soot has a large real part of refractive index and is highly absorbing, mineral dust aerosol is also, but less, absorbing, while the other aerosol types, water soluble and sea salt at 95% humidity, have refractive indices near that of water. The latter two types have very similar kernels, due to their similar refractive indices, and can't be distinguished. However, Fig. 1 shows clearly that three types of aerosols have very different kernels and this information can be used in the retrieval algorithm.
The refractive index sensitive kernels are used to retrieve integral microphysical properties of tropospheric aerosol, like total volume, irrespective of the refractive index of the ambient aerosol.

2.2. Kernel selection

In order to find the optimum kernel for the retrieval of total volume, the extinction and backscatter coefficients are retrieved first, which can be done using weights $w_j = 3Q_{\alpha,\lambda}(r_j)/4r_j$ for aerosol extinction and $w_j = 3Q_{\beta_\pi,\lambda}(r_j)/4r_j$ for aerosol backscatter, and compared to the input extinction and backscatter coefficients. Doing so using all five wavelengths is trivial, because in that case in eq. 6

$$\bar{\mathbf{w}}^{\mathbf{t}} \mathbf{K}^{\mathbf{t}} \mathbf{U} \mathbf{L}^{-1} \mathbf{U}^{\mathbf{t}} = \mathbf{I}, \tag{7}$$

the identity matrix, simply returning the measurements. The extinction and backscatter coefficients at each wavelength can also be estimated from the solutions at other wavelengths. So, instead of using the kernels at all wavelengths, one is omitted and the extinction and backscatter at that wavelength are estimated using the remaining kernels:

$$\{\bar{\alpha}, \bar{\beta}\}^{\mathbf{t}} = \bar{\mathbf{w}}_{\mathbf{r}}^{\mathbf{t}} \mathbf{K_r}^{\mathbf{t}} \mathbf{U_r} \mathbf{L_r}^{-1} \mathbf{U_r}^{\mathbf{t}} \cdot \bar{\mathbf{g}}_{\mathbf{r}}, \tag{8}$$

where $\bar{\mathbf{g}}_{\mathbf{r}}$ is the input measurement vector reduced with one measurement. The subscript $\mathbf{r}$ indicates the reduction of the corresponding vectors and matrices with one dimension. Then the kernel is selected which has the smallest difference ϵ between the α or β retrieved in this way and the measurement $\bar{\mathbf{g}}$

$$\epsilon = |\bar{\mathbf{g}} - \{\bar{\alpha}, \bar{\beta}\}^{\mathbf{t}}|, \tag{9}$$

Once the optimum kernel is selected this is then used to predict the aerosol physical properties.

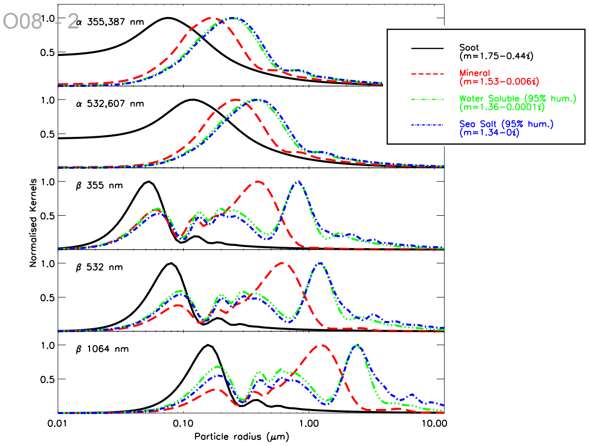

Figure 1. Kernel functions for four different types of aerosols: soot, mineral dust, water soluble at 95% humidity and sea salt at 95% humidity. Optical properties were taken from [2] .

3. RESULTS

The extinction and backscatter coefficients of the aerosol models described above were used as input for the PCA method, to show the kernel sensitivities as a function of particle radius (Fig. 2). The relative volume error ($(V_{\text{true}} - V_{\text{retrieved}})/(V_{\text{true}} + V_{\text{retrieved}})$), which is the total volume retrieved compared to the true volume, is shown as a function of particle radius for all aerosol types. The results from the various kernels are shown by lines, while the optimum retrieval is indicated by symbols. Near perfect retrievals can only be expected for those particle radii where the kernels are distinct enough (see Fig. 1). For example, the top panel shows that good soot aerosol volume retrievals can only be expected for particle radii below about 0.3 μm, while retrievals for mineral dust and water soluble aerosols have the best results for particle radii between about 0.1 and 1.0 μm.

Figure 2 also shows that the selected optimum kernel is based on the same refractive index as the input aerosol model for those particle radii mentioned above. Only water soluble and sea salt aerosols are sometimes interchanged, but those refractive indices are almost equal. Note that the refractive indices used in the input and those used in the retrievals are exactly the same, so Fig. 2 shows the maximum achievable results for the method. However, the results of the volume retrievals and other bulk quantities are not so dependent

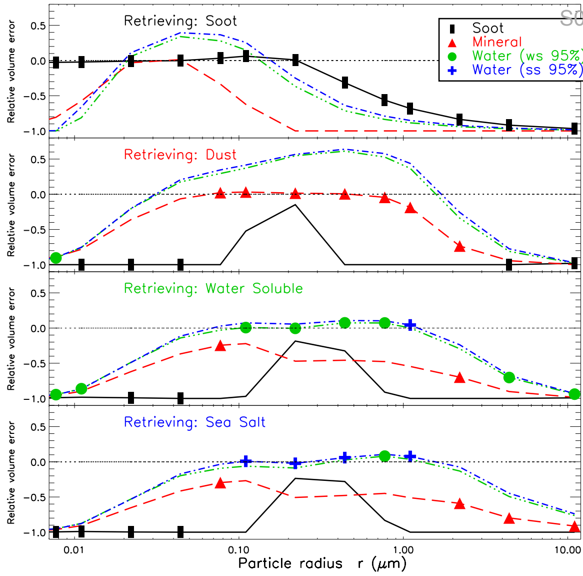

Figure 2. Relative error of total volume retrievals as a function of particle radius, for the four different aerosols types (as input). The coloured lines show the retrievals using the different kernels and the symbols indicate the selected optimal retrieval.

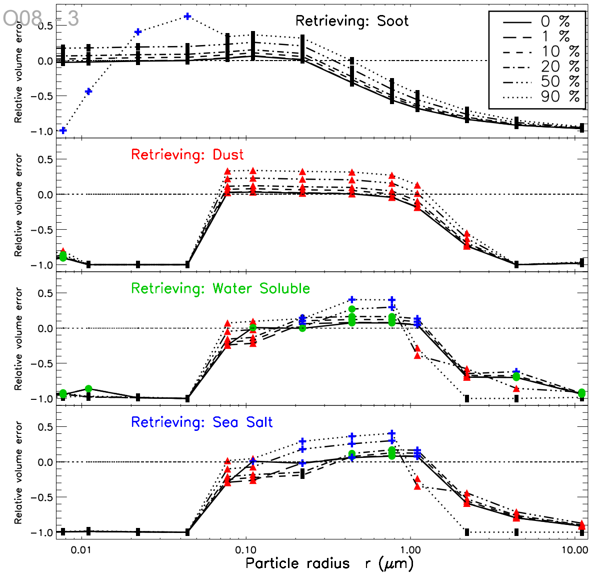

Figure 3. Relative error of total volume retrievals for different levels of error in the input 'signals'. Shown are the selected optimal retrievals as a function particle radius. The solid line (error=0) equals the selected optimal retrievals of Fig. 2.

on the exact refractive index of the aerosols in question, if the aerosol models in the retrieval span a large enough space in refractive index. Increasing the number of aerosol models in the retrieval with different refractive index will increase the accuracy of the retrieved refractive index only marginally, while the retrieval of total volume or other bulk properties are also almost not affected. Outside the particle radius interval where the kernels are sensitive, both the refractive index retrieval and any other retrieval is useless.

3.1. Errors

The results are rather stable for errors in the input signals, i.e. the measured extinction and backscatter measurements. Figure 3 shows the results for total volume retrievals, again as a function of particle radius, for various amounts of error in the input signals. The solid line connects the selected optimum kernels in Fig. 2, i.e. no error on the input signals, while the other curves shows the change of the selected kernels and the relative error in the retrieved total volume for different error levels. The optimum kernel selected in the PCA retrieval becomes unrelated to the refractive index of the input aerosols as the error becomes larger. However, the error of the total volume retrieval is much less affected, and the error in the retrieval is of the same order as the error in the measured signal.

3.2. Application to lidar profiles

The PCA method described above is applied to lidar profiles from Caeli (Cesar water vapor, AErosol and cloud LIdar)[3], a multi-wavelength Raman lidar, built by RIVM in The Netherlands. The data used in this paper were collected in Leipzig, Germany, during an intercomparison campaign in the framework of Earlinet [4].

The data are presented in Fig. 4. The left panel shows the development of the boundary layer in time, which is slowly decaying between 16:00 and 20:00 h on 25 May 2009, from 3.5 to about 3 km in height. The right panel shows the aerosol scattering ratio profiles R (the aerosol backscatter relative to the molecular backscatter $R = (\beta_{\pi,A} + \beta_{\pi,M})/\beta_{\pi,M}$) and extinction coefficient profiles from the same period. The lowest 1 km is not used in the retrieval due to overlap problems between the laser and the receiver. Both the overview and the backscatter and extinction profiles suggest the existence of two separate layers in the boundary layer, from 1 to 2 km altitude, and 2 to 3.5 km altitude.

The backscatter and extinction profiles shown in the right panel of Fig. 4 were used as input for the PCA retrieval (see Fig. 5). The left panel of Fig. 5 shows the chosen aerosol model in the PCA retrieval as a function of height. The next panel shows the result of the retrieval for total volume. The dotted lines show the results for every aerosol model separately, while the thick purple line is the result from the selected models, which is the final result. The total volume is about $3 - 5\,\mu$m/cm^3 over the entire layer, while the selected kernels are mostly soot and sea salt. This might result from a polluted humid aerosol layer, but this has not been verified.

The next panel shows the retrieval of the total surface area. The result using volume kernels is shown in

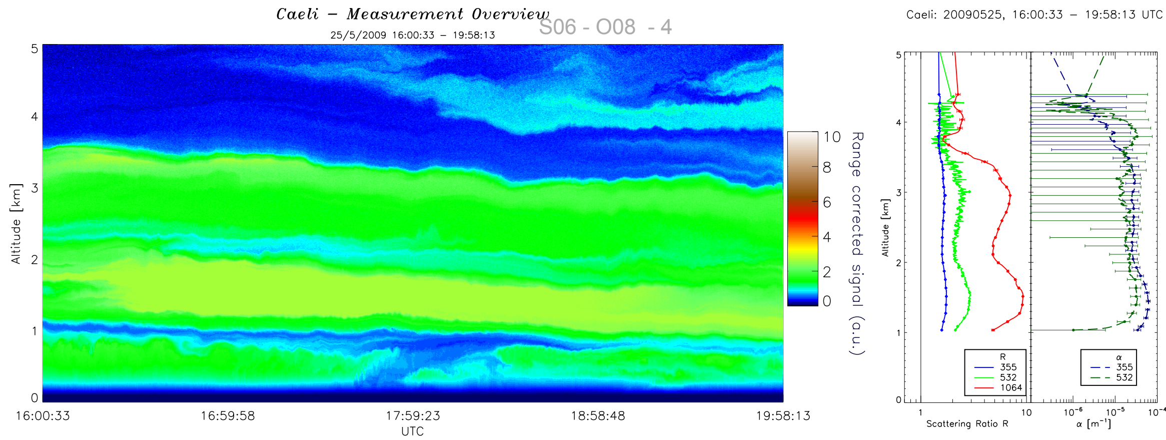

Figure 4. Measurements from the Raman lidar Caeli*, made on 25 March 2009 in Leipzig, from 16:00 to 20.00h. Left panel shows the range corrected signal of the 1064 channel as a function of time in arbitrary units. The right panel shows the time-integrated aerosol scattering ratios R for the three elastic channels and the extinction coefficients for the visible and UV channels for the same period.*

purple, while the result from surface kernels is shown in pink. Clearly, the use of surface kernels produces much more consistent results for the total surface area, as was found before [1]. A vague two-layer structure is visible, with total surface areas ranging from about $20 - 100\,\mu$m/cm^2. The right panel shows the retrieved effective radius, which is a function of the retrieved total volume and the retrieved surface area of the aerosols.

4. CONCLUSIONS

PCA is a very useful method to determine microphysical quantities from lidar optical data. The method can also be used in the troposphere, using the dependence of the kernels on refractive index. Only a few, sufficiently distinct kernels are necessary to achieve useful results of integral microphysical quantities. However, the retrieval of accurate refractive index information using this few kernels is not possible.

ACKNOWLEDGMENTS

This work was financed by the Netherlands Institute for Space Research, project number GO-2005/075.

REFERENCES

[1] D. P. Donovan and A. I. Carswell. Principle component analysis applied to multiwavelength lidar aerosol backscatter and extinction measurements. *Appl. Opt.*, 36(36):9406–9424, 1997.

[2] M. Hess, P. Koepke, and I. Schult. Optical Properties of Aerosols and Clouds: The software package OPAC. *Bull. Am. Met. Soc.*, 79:831–844, 1998.

[3] K.M. Wilson, A. Apituley, and C. Potma. Caeli - a Raman lidar for the diurnal observation of clouds, aerosol and water vapour. *ILRC 24*, 2008.

[4] V. Matthais, et al. Aerosol Lidar Intercomparison in the Framework of the EARLINET Project. 1. Instruments. *Appl. Opt.*, 43:961–976, 2004.

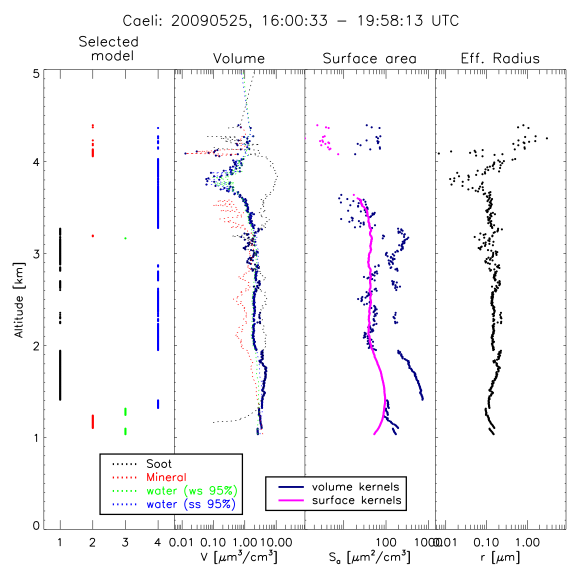

Figure 5. PCA results for the signals shown in Fig.4. The left panel shows the selected aerosol kernels. The next panel shows the total aerosol volume as the thick purple line, while the retrievals from all four different kernel sets are shown as dotted lines in the colors as indicated by the legend. The next panel shows the total aerosol surface area retrieval in purple when volume kernels are used, and in pink the result when surface kernels are used instead. The right panel shows the retrieved particle radius, which is a function of total volume retrieved with volume kernels and total surface area retrieved with surface kernels.