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# Retrieval of aerosol backscatter, extinction, and lidar ratio from Raman lidar with optimal estimation

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Retrieval of  
backscatter and  
extinction from  
Raman lidar

A. C. Povey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

## Abstract

Optimal estimation retrieval is a form of non-linear regression which determines the most probable circumstances that produced a given observation, weighted against any prior knowledge of the system. This paper applies the technique to the estimation of aerosol backscatter and extinction (or lidar ratio) from two-channel Raman lidar observations. It produces results from simulated and real data consistent with existing Raman lidar analyses and additionally returns a more rigorous estimate of its uncertainties while automatically selecting an appropriate resolution without the imposition of artificial constraints. Backscatter is retrieved at the instrument's native resolution with an uncertainty between 2 and 20 %. Extinction is less well constrained, retrieved at a resolution of 0.1–1 km depending on the quality of the data. The uncertainty in extinction is > 15 %, in part due to the consideration of short one-minute integrations, but is comparable to fair estimates of the error when using the standard Raman lidar technique.

The retrieval is then applied to several hours of observation on 19 April 2010 of ash from the Eyjafjallajökull eruption. A highly depolarizing ash layer is found with a lidar ratio of 20–30 sr, much lower values than observed by previous studies. This potentially indicates a growth of the particles after 12–24 h within the planetary boundary layer. A lower concentration of ash within a residual layer exhibited a backscatter of  $10 \text{ Mm}^{-1} \text{ sr}^{-1}$  and lidar ratio of 40 sr.

## 1 Introduction

Aerosols impact the Earth's radiation budget both directly, by reflecting solar radiation back into space (Haywood and Shine, 1995), and indirectly, by altering the properties and distribution of clouds (Lohmann and Feichter, 2005) or reacting with other species (Colbeck, 1998). The lack of knowledge about the global distribution and composition of aerosols is currently the single greatest source of uncertainty in estimates of net

## Retrieval of backscatter and extinction from Raman lidar

A. C. Povey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

radiative forcing and therefore is a factor in the ability to predict the impacts of climate change (IPCC, 2007).

Lidar (light detection and ranging) is an active remote sensing technique for observing the distribution of molecules and particles in the atmosphere as a function of height by means of the light they backscatter from a laser beam. The name intentionally emulates radar as both techniques use the time-of-flight of a pulsed source to deduce the distance to the scatterer (Fugii and Fukuchi, 2005). Despite its exceptionally high spatial and temporal resolution, lidar is not as widely applied as other techniques in the study of aerosol. With the launch of a space-based lidar (Vaughan et al., 2004) and the development of networks across Northern America (Welton et al., 2001), Europe (Pappalardo et al., 2005), and Asia (Sugimoto et al., 2008), there is an increasing volume of under-used lidar data.

The energy observed by a lidar is a function of the extinction and backscattering coefficients – the cross-section per unit volume to either attenuate the beam or to scatter light directly back towards the instrument. These coefficients are functions of the microphysical properties of the aerosol present, such as refractive index and size distribution. Deriving such properties directly is possible, but the problem is very poorly constrained. Its solution either requires a greater number of measurements, such as a multi-wavelength system (Müller et al., 1999), or further assumptions about the scatterers. These complexities are disregarded here in favour of the better-constrained estimation of extinction and backscatter.

Optimal estimation retrieval is a form of non-linear regression which determines the most probable circumstances that produced a given observation, weighted against any prior knowledge of the system. For several decades, it has been successfully applied to the analysis of satellite (e.g. Marks and Rodgers, 1993; Li et al., 2008; Watts et al., 2011), radar (Grant et al., 2004), and ground-based radiometer observations (e.g. Guldner and Spankuch, 2001) but has not seen substantial use within the lidar community. This paper applies the technique to the estimation of aerosol extinction and backscatter from two-channel Raman lidar observations. The retrieval processes

**Retrieval of backscatter and extinction from Raman lidar**

A. C. Povey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion





that expectation is described by the a priori covariance  $\mathbf{S}_a$ . The quantity  $-2\ln P(\mathbf{x}|\mathbf{y})$  is hereafter referred to as the cost as it measures the goodness of fit for a solution. Good models should have a cost approximately equal to the number of measurements. Hence, the cost will herein be quoted normalised by the length of  $\mathbf{y}$ .

5 It can be shown that the iteration,

$$\mathbf{x}_{i+1} = \mathbf{x}_i + [(1 + \Gamma_i)\mathbf{S}_a^{-1} + \mathbf{K}_i^T \mathbf{S}_e^{-1} \mathbf{K}_i]^{-1} \{ \mathbf{K}_i^T \mathbf{S}_e^{-1} [\mathbf{y} - F(\mathbf{x}_i, \mathbf{b})] - \mathbf{S}_a^{-1} (\mathbf{x}_i - \mathbf{x}_a) \}, \quad (3)$$

converges to the most probable state  $\hat{\mathbf{x}}$ , where the Jacobean  $\mathbf{K}_i = \nabla_{\mathbf{x}_i} F(\mathbf{x}_i, \mathbf{b})$  and  $\Gamma_i$  is a scaling constant. General practice, outlined in Fig. 1, is that if the cost increases after an iteration,  $\Gamma_i$  is increased by a factor of ten. Otherwise, it is reduced by a factor of two. Evaluation ceases after:

- The cost function decreases by much less than the number of measurements;
- The cost decreased and the change in the state is much less than the predicted error,

$$(\mathbf{x}_{i+1} - \mathbf{x}_i)_j \ll \sqrt{\mathbf{S}_{jj}} \quad \forall j,$$

15 where the error covariance matrix of the solution  $\mathbf{S} = (\mathbf{K}_i^T \mathbf{S}_e^{-1} \mathbf{K}_i + \mathbf{S}_a^{-1})^{-1}$ .

- The step in state space,

$$\{ \mathbf{K}_i^T \mathbf{S}_e^{-1} [\mathbf{y} - F(\mathbf{x}_i, \mathbf{b})] - \mathbf{S}_a^{-1} (\mathbf{x}_i - \mathbf{x}_a) \} (\mathbf{x}_{i+1} - \mathbf{x}_i)^T,$$

is much less than the length of the state vector;

- 30 iterations, which is considered a failure to converge.

20 The averaging kernel,

$$\mathbf{A} = \hat{\mathbf{S}} \hat{\mathbf{K}}^T \mathbf{S}_e^{-1} \hat{\mathbf{K}}, \quad (4)$$

Retrieval of  
backscatter and  
extinction from  
Raman lidar

A. C. Povey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



describes the extent to which the true and a priori states each contribute to the solution as it can be shown that,

$$\hat{\mathbf{x}} = \mathbf{A}\mathbf{x} + (\mathbf{I} - \mathbf{A})\mathbf{x}_a + \hat{\mathbf{S}}\hat{\mathbf{K}}^T\mathbf{S}_e^{-1}\mathbf{e}, \quad (5)$$

where a hat indicates the value after convergence. An ideal retrieval would have a kernel equal to the identity. In practice, the rows of  $\mathbf{A}$  will be peaked functions showing how the information in one retrieved bin is derived from an average of the true values around it. The width of that peak is therefore a measure of the resolution of the retrieval.

## 2.2 Existing lidar analyses

The energy observed from a height  $R$  is expressed by the lidar equation (Measures, 1992),

$$E(R, \lambda) = \frac{E_L C(R, \lambda)}{R^2} \bar{\beta}(R, \lambda) \exp \left[ - \int_0^R \bar{\alpha}(R', \lambda_L) + \bar{\alpha}(R', \lambda) dR' \right], \quad (6)$$

where  $\bar{\beta}(R, \lambda)$  is the coefficient for incident laser light, wavelength  $\lambda_L$ , to be backscattered at a wavelength  $\lambda$ ;  $\bar{\alpha}(R, \lambda)$  is the extinction coefficient;  $E_L$  is the energy of the laser pulse; and  $C(R)$ , known as the overlap function, describes the alignment and efficiency of the detection system. As both the extinction and backscatter are unknown, a single profile presents an underconstrained measurement.

The atmosphere is assumed to contain only two components such that,

$$\bar{\beta} = \beta + \beta^{(m)} \quad (7)$$

$$\bar{\alpha} = \alpha + \alpha^{(m)}, \quad (8)$$

where  $\beta$ ,  $\alpha$  denote backscattering and extinction by aerosols and  $\beta^{(m)}$ ,  $\alpha^{(m)}$  denote scattering by molecules, which is well-modelled by Rayleigh scattering.

## Retrieval of backscatter and extinction from Raman lidar

A. C. Povey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



## Retrieval of backscatter and extinction from Raman lidar

A. C. Povey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

The dominant return for any lidar is the elastic profile (where  $\lambda = \lambda_L$ ), from which the backscatter is commonly derived by a technique known as onion peeling or the Fernald–Klett method (Klett, 1981; Fernald, 1984). A Raman lidar monitors a second channel containing the Raman scattering from a single species in the atmosphere, such that  $\bar{\beta}$  becomes a known function of number density. Ansmann et al. (1992) outlined a means to invert Eq. (6) in such circumstances to derive the extinction and backscatter separately.

A few applications of non-linear regression to lidar have been published. A retrieval of ice water path and effective radius in cirrus clouds from coincident, space-borne lidar and radar measurements was developed in Delanoe and Hogan (2008), though its results were found to be highly dependant on the microphysical assumptions. Pounder et al. (2012) derived high-quality extinction retrievals from three simultaneous observations with different fields of view using a linearised model of the lidar equation that included multiple scattering while applying Twomey–Tikhonov smoothing rather than an a priori. Marchant et al. (2010) presented an original, if limited, linearised scheme that decomposed scattering over a basis of precomputed aerosols. This was expanded to a retrieval of effective radius in multiwavelength studies via a Kalman filter in Marchant et al. (2012).

A related method known as regularisation has also been applied to Raman lidar. The introduction of Veselovskii et al. (2002) provides a good review of early attempts and the methodology. Shcherbakov (2007) and Pornsawad et al. (2012) demonstrated that such methods return solely positive extinction and can produce more accurate products than the Ansmann method but require artificial smoothing within the retrieval, which generates significant errors where there are significant gradients. Evaluation also requires a set of basis functions to be defined, artificially imposing a structure onto the system.

It is preferable to impose the basis for smoothing solutions through an a priori covariance matrix derived from actual data and the physical processes driving the system, as facilitated by optimal estimation retrieval. The impact of these assumptions can be





axis with even 33 m spacing will be used. Provided the grid size is smaller than the features to be resolved, its choice only impacts the computational cost.

Neglecting multiple scattering, the number of photons observed from range bin  $R_i$  will be,

$$E_i^{(L)} = E_L \frac{C_i^{(L)}}{R_i^2} \left[ \frac{\sigma_R^{(L)}}{\beta} N_i + \text{spline}_{r \rightarrow R_i}[\tilde{\beta}] \right] \exp \left[ -2 \left( \sigma_R^{(L)} \mathcal{N}_i + \text{spline}_{r \rightarrow R_i}[\tilde{\chi}] \right) \right] + E_B^{(L)} \quad (10)$$

$$E_i^{(ra)} = E_L \frac{C_i^{(ra)}}{R_i^2} N_i \exp \left[ - \left( \sigma_R^{(L)} + \sigma_R^{(ra)} \right) \mathcal{N}_i - \left( 1 + \frac{\lambda_L}{\lambda_{ra}} \right) \text{spline}_{r \rightarrow R_i}[\tilde{\chi}] \right] + E_B^{(ra)}, \quad (11)$$

where  $\mathcal{N}_i = \int_0^{R_i} N(R') dR'$ ; a tilde represents a variable on the retrieved grid  $r$ ; the aerosol optical thickness  $\chi = \int_0^R \alpha(\lambda_L, R') dR'$ ;  $\beta$  and  $\chi$  are evaluated at  $\lambda_L$ , though this dependence is dropped for brevity; and  $E_B$  is the background count rate which is estimated from observations as  $R \rightarrow \infty$ . The calibration function  $C(R)$  is assumed known and is input into as a parameter.

The optical thickness is evaluated by the trapezium rule,

$$\tilde{\chi}_j = \tilde{\alpha}_0 r_0 + \frac{1}{2} \sum_{k=1}^j [\tilde{\alpha}_k + \tilde{\alpha}_{k-1}] [r_k - r_{k-1}]. \quad (12)$$

Note that the extinction is assumed constant through the first bin, such that it acts as a boundary term rather than a physically meaningful value. This avoids various difficulties with observation very near the instrument.

Extinction and backscatter are both functions of  $N$  and so will be correlated. This should be identified within  $\mathbf{S}_a$  but is not be easily estimated. Further, the use of correlated variables will emphasise degenerate states of the forward model, which can slow the retrieval's convergence. This can be averted by retrieving the lidar ratio instead, which is independent of  $N$ ,

Retrieval of backscatter and extinction from Raman lidar

A. C. Povey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Retrieval of backscatter and extinction from Raman lidar**

A. C. Povey et al.

Title Page	
Abstract	Introduction
Conclusions	References
Tables	Figures
◀	▶
◀	▶
Back	Close
Full Screen / Esc	
Printer-friendly Version	
Interactive Discussion	

$$\tilde{\chi}_j = \tilde{\beta}_0 \tilde{B}_0 r_0 + \frac{1}{2} \sum_{k=1}^j [\tilde{\beta}_k \tilde{B}_k + \tilde{\beta}_{k-1} \tilde{B}_{k-1}] [r_k - r_{k-1}]. \tag{13}$$

All elements of  $\mathbf{x}$  should be positive or the retrieval will explore unrealistic models. For the retrieval of  $\beta$  and  $\alpha$ , this will be prevented by setting all negative values to zero after evaluating Eq. (3). For the retrieval of  $\beta$  and  $B$ , it was found preferable to instead retrieve  $\ln \beta$  while enforcing a lower limit of unity on  $B$ .

The measurement and state vectors are then,

$$\mathbf{y} = \begin{pmatrix} \varphi_0^{(L)} \\ \varphi_1^{(L)} \\ \vdots \\ \varphi_{m-1}^{(L)} \\ \varphi_0^{(ra)} \\ \varphi_1^{(ra)} \\ \vdots \\ \varphi_{m-1}^{(ra)} \end{pmatrix} \quad \text{and} \quad \mathbf{x} = \begin{pmatrix} \tilde{\beta}_0 \\ \tilde{\beta}_1 \\ \vdots \\ \tilde{\beta}_{n-1} \\ \tilde{\alpha}_0 \\ \tilde{\alpha}_1 \\ \vdots \\ \tilde{\alpha}_{n-1} \end{pmatrix} \parallel \begin{pmatrix} \ln \tilde{\beta}_0 \\ \ln \tilde{\beta}_1 \\ \vdots \\ \ln \tilde{\beta}_{n-1} \\ \tilde{B}_0 \\ \tilde{B}_1 \\ \vdots \\ \tilde{B}_{n-1} \end{pmatrix}.$$

The first guess for  $\mathbf{x}$  in the iteration Eq. (3) is taken as  $\beta = 10^{-5} \text{ Mm}^{-1} \text{ sr}^{-1}$  and  $B = 58 \text{ sr}$ . These values were chosen as they tend to reduce the number of iterations and their value does not affect the final result (if the retrieval converges).

One final note must be made of the treatment of measurement error (which is assumed uncorrelated),  $\mathbf{S}_e$ . The observed photon counts should be Poisson distributed, such that their variance is equal to their mean. This is widely used to justify approximating the variance of a lidar measurement with the measurement itself. This is not strictly valid as the measurement is only a single sample of a distribution. However, a lidar



sums profiles over several seconds or minutes of laser shots during data collection, giving no further measure of their variance.

The optimal estimation scheme requires an unbiased estimate of the variance. Using the measurement itself causes the retrieval to favour observations that coincidentally suffer large, positive noise as they are then presumed to be more precise. This effect is most pronounced at low signal levels and introduces a high bias into the retrieval. To alleviate this, the variance will be estimated from the application of a five-bin, sliding-window average to the data. The impact of this will not be explored in detail, though preliminary studies found that even minimal smoothing of the variance vastly reduced biases.

For further details, derivations, and justification of the forward model, please consult Chapt. 2 of Povey (2013).

## 2.4 A priori

Arguably the most important component of an optimal estimation scheme is its a priori. Ideally, the a priori would not greatly affect the retrieval, but in practice it constrains which states are deemed to be both physically possible and likely. In this problem, solutions should be reasonably smooth as aerosols are often well-mixed through the planetary boundary layer (PBL), but gradients shouldn't be completely excluded as layering does occur.

The exact composition and optical properties of aerosol are highly variable and climatologies (from which an a priori would be derived in most applications) rarely exist. Some generalised descriptions of representative aerosol types have been explored in the literature. The OPAC model (Optical Properties of Aerosol and Cloud, Hess et al., 1998) defines size distributions, refractive indices, and number densities for a variety of cloud and aerosol particles. These provide the necessary inputs for Mie codes (Grainger et al., 2004) to calculate the extinction and backscatter. Combinations of these based on expected and observed compositions then produce characteristic aerosol mixtures, such as marine or urban.

### Retrieval of backscatter and extinction from Raman lidar

A. C. Povey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



## Retrieval of backscatter and extinction from Raman lidar

A. C. Povey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



For the data to be considered, the continental type should be appropriate – comprising soot with soluble and insoluble aerosols. An ensemble of scattering properties was constructed by randomising the abundance of these components, using the OPAC model values as the mean of a Gaussian distribution with width estimated by 10 % of that mean (the exact value assumed was found to be unimportant). Aspherical particles produce an effectively identical distribution. The resulting distributions of  $\beta$ ,  $\alpha$ , and  $B$  are shown in Fig. 2. The a priori is based on qualitative fits to these, shown in blue when retrieving linearly and in red for a logarithmic retrieval. Though the logarithmic retrievals appear to give a better fit to the distributions, the retrieval of  $\ln\beta$  and  $\ln\alpha$  was found to be overly constrained. Though the lidar ratio is theoretically a better description of the state, its distribution is not symmetric and so not necessarily well-suited to optimal estimation. A relatively broad a priori distribution has been selected to compensate. These distributions demonstrate an approximately 95 % correlation between  $\beta$  and  $\alpha$ , which is included in  $\mathbf{S}_a$  for the linear retrieval. Though its exact value appears to be unimportant, it would be desirable to obtain a more rigorous estimate.

The OPAC model states that the density of non-dust aerosols decreases exponentially with a scale height of 2 km. The prescribed values will therefore decrease similarly in  $\mathbf{x}_a$ . Further, there will almost certainly be some vertical correlation of the measurements due to vertical mixing. The simple model of a Markov process proposed by (2.83) of Rodgers (2000) shall be used with correlations decaying exponentially with separation,

$$(\mathbf{S}_a)_{ij} = \sqrt{(\mathbf{S}_a)_{ii}(\mathbf{S}_a)_{jj}} \exp\left(-\frac{|r_i - r_j|}{H}\right), \quad (14)$$

where  $H$  is a scale height.

The scale height can be estimated by investigating the covariance of some measure of aerosol scattering. A convenient option is backscatter sondes (NDACC, 2000), which measure the light backscattered from a xenon flashlamp approximately every 30 m during a balloon ascent (Rosen and Kjome, 1991; Rosen et al., 2000). Profiles over ten

## Retrieval of backscatter and extinction from Raman lidar

A. C. Povey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

years of observations at three sites have been used in Fig. 3 to produce a correlation matrix of backscatter ratio with height. Its rows decay roughly exponentially with height, which when fitted to Eq. (14) give  $H = 1\text{--}2$  km in the free troposphere.

This will not necessarily apply within the PBL, which is only weakly coupled to the free troposphere (Oke, 1987). Several studies of the vertical distribution of aerosol within the PBL have been performed with tethered balloons, though the data could not be readily accessed. These generally find that aerosol concentrations are constant with height (Figs. 10–12, 4, and 2 of Ewell et al., 1989; Greenberg et al., 2009; Ferrero et al., 2010, respectively), but occasionally observe fine structure (Fig. 6 of Ferrero et al., 2011). Further, a myriad of literature covers observations of aerosol layers tens to hundreds of metres thick (e.g. Di Girolamo et al., 1999; Dacre et al., 2011) or variations within lofted aerosol features (e.g. Althausen et al., 2000; Huang et al., 2010).

The a priori covariance matrix should represent both the general tendency for aerosols to be well-mixed throughout the troposphere and the fine scale structure that occasionally occurs. The average position of the top of the PBL should be expressed by a significant decrease in correlation between areas above and below it. At the moment, there is insufficient information to quantify these effects with any degree of certainty. As such, a conservative estimate of  $H = 100$  m is used, which will not make the best use of the available information but does not overconstrain the solution.

### 3 Simulations

Simulated data can be easily produced with the forward model, using the NOAA (1976) standard atmosphere. The PBL extinction profile is modelled by an error function (Steyn et al., 1999) multiplied by an exponential decay above the PBL. Aerosol and cloud layers are modelled by Gaussian peaks (Biavati, G., personal communication, 2011). An analytic model of the RACHEL instrument is used to generate the calibration function and detector nonlinearity (see Povey et al., 2012). Once simulated, Poisson noise is added to the profiles.

### 3.1 Sensitivity

The retrieval from six simulated profiles by both proposed configurations is shown in Fig. 4. The two configurations give equivalent results and successfully retrieve the simulated profile in cases (a–d). Cases (e) and (f) return large costs, such that it is obvious they have failed. In (e), an incorrect nonlinear correction causes an underestimation where the observed profile has maximal energy. The observation of a cloud in (f) is reasonable within the PBL but fails above that. The large scattering within the cloud is outside of the range prescribed by the a priori and, though it obtains a decent fit to the visible region of the cloud, vertical correlations cause incorrect retrieval beneath it. Successfully fitting cloud and aerosol observations simultaneously requires a more detailed forward model and a priori.

The lidar ratio profiles indicate that there is a decrease in the information content of the measurement above the PBL, where scattering (and therefore the magnitude of the return) is lower. The two configurations react differently to this. The lidar ratio configuration returns a smooth  $B$  profile that tends towards its a priori value above the PBL, as would be expected, while the extinction configuration gives a much noisier profile, indicating it is less constrained by the a priori.

A further six simulations containing small-scale fluctuations are presented in Fig. 5. The two configurations behave as before, with the lidar ratio mode returning a smoother profile but losing sensitivity above the PBL. The “layers” of cases (g) and (h) are correctly positioned by both modes, if slightly underestimated in magnitude. In case (i), the layers are not resolved (see Fig. 6) as the features occupied only one retrieval bin. Doubling the resolution gives equivalent performance to cases (g) and (h) but with slightly increased noise and significantly increased processing time. Cases (k) and (l) are more difficult retrievals as they present lower SNR. They are still consistent with the true profile but with greater errors.

Both configurations give a respectable fit to the extinction profile, though they do increasingly underestimate the magnitude of peaks as they become narrower. This de-

## Retrieval of backscatter and extinction from Raman lidar

A. C. Povey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



extinction retrieval is better, though it underestimates the error in the PBL. Both fail to appreciate the error caused by the improper dead time correction.

Those figures also compare the retrieval to the Ansmann method. For a fair comparison, the derivative is averaged over 300 m to be equivalent to the effective resolution of the retrieval. They are in good agreement in the PBL and the retrievals exhibit a lesser spread and error than the Ansmann solutions in the free troposphere. The Fernald–Klett method gives equivalent answers when given the correct lidar ratio.

### 3.2.2 Parameter error

In real retrievals, there will be some error in the model parameters  $\mathbf{b}$ . This additional uncertainty can be included in the retrieval by extending the measurement uncertainty to cover all sources of error,

$$\boldsymbol{\epsilon}_y = \boldsymbol{\epsilon} + \mathbf{K}_b(\mathbf{b} - \hat{\mathbf{b}}) + \Delta\mathbf{f}, \quad (15)$$

where  $\mathbf{K}_b = \partial F / \partial \mathbf{b}$ ;  $\hat{\mathbf{b}}$  is the best estimate of the true parameters  $\mathbf{b}$ ; and the last term describes any inability of the forward model to describe the true state.

Concentrating on only the parameter error for now, this can be implemented by replacing all occurrences of  $\mathbf{S}_e$  with,

$$\mathbf{S}_y = \mathbf{S}_e + \mathbf{K}_b \mathbf{S}_b \mathbf{K}_b^T. \quad (16)$$

This significantly increases the computing cost of the retrieval, as  $\mathbf{S}_y$  must now be inverted in each iteration. A reasonable approximation is to only re-evaluate  $\mathbf{S}_y$  after the last iteration. The full calculation is considered here.

The Ångström coefficient can vary quite significantly but is commonly accepted to lie in the range 0.6–1.4, such that an error of 0.4 is reasonable (Klett, 1985). Radiosondes measure pressure and temperature at a given height with an accuracy of 0.5 hPa, 2 K, and 60 m, from which an error in number density and  $\chi^{(m)}$  of 0.5 % is expected (Kitchen,

## Retrieval of backscatter and extinction from Raman lidar

A. C. Povey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion







so  $r_0 = 100$  m was chosen to concentrate these effects within a region where parameter errors will be large regardless.

Forward model error is defined in Sect. 3.2.3 of Rodgers (2000) as,

$$\mathbf{G}_y[\mathbf{f}(\mathbf{x}, \mathbf{b}, \mathbf{b}') - F(\mathbf{x}, \mathbf{b})], \quad (17)$$

where  $\mathbf{G}_y = \partial \hat{\mathbf{x}} / \partial \mathbf{y}$ , the sensitivity of the retrieved state to the measurement, and  $\mathbf{f}$  is the exact, true profile including any processes the forward model  $F$  may not describe. This systematic error is generally difficult to estimate as, if  $\mathbf{f}$  were known, it would most likely be used as the forward model instead.

There are some processes that are clearly not included within the current forward model. Multiple scattering has been neglected as it is mostly important for lidars with a wide footprint, such as space-based system, or for observations within clouds, where this algorithm is already known to perform poorly for other reasons. Though appropriate numerical models of multiple scattering exist (Eloranta, 1998), this is left as an area for future work if retrievals within clouds are desired.

There is a small difference between the bin-averaged backscatter that is sampled and the true backscatter defined by Mie theory, for which the error can be evaluated with Eq. (17). It is greatest in the entrainment layer (or at any other sharp gradient), being at most 1 % of the total error.

The models of the calibration function and detector nonlinearity are idealised versions of the truth. A rough estimate of these contributions can be produced by considering alternative models, such as Donovan et al. (1993). For case (e), these are over 100 times larger than the other errors, again demonstrating that great care must be taken with the dead time correction. Measurements in the linear observation regime are negligibly affected by the choice of nonlinear correction.

None of these errors describe the discrepancies shown in the free troposphere in Fig. 9 as that is dominated by the a priori uncertainty. In regions where the data are the dominant contribution to the retrieval (i.e. where the area of the averaging kernel is near unity), increasing the a priori variance does not affect the retrieved profiles.

## Retrieval of backscatter and extinction from Raman lidar

A. C. Povey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion







## Retrieval of backscatter and extinction from Raman lidar

A. C. Povey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

increasingly from reality. As the retrieval has no knowledge of that, it retrieves smaller backscatter to compensate. The Ansmann method is not affected as it considers a ratio of channels. The difference between the Ansmann solution and the retrieval is effectively a constant factor of the failure in the calibration, with the results otherwise being consistent. For example, both methods observe larger  $\beta$  in updrafts than downdrafts (where vertical wind was observed by a Doppler lidar).

The retrievals are consistent in their estimates of the lidar ratio and are no worse than the Ansmann method, which is greatly affected by overlap when estimating  $\alpha$ . The aerosol layer near 1 km at 10:00 GMT gives a lidar ratio of around 30 sr. This is a residual layer lying above a developing mixed layer where lidar ratios are larger (around 50 sr). The low lidar ratio indicates large, likely spherical, particles which are reasonable for an aged residual layer. The results are better in the evening, observing a peak  $B$  of 70 sr over a background of 50 sr, indicating the appearance of smaller particles. By this time, convective mixing has collapsed into a persistent updraft so the increase in depolarization ratio could indicate that newer, non-spherical particles are being lofted from the surface or are advected over the site. Advected aerosol is more likely considering the brevity of the peak.

Figure 15 compares the retrieved  $\chi_{\infty}$  during that day to AERONET measurements (Woodhouse and Agnew, 2010–2011). Their agreement is reasonable if not impressive. The retrieval tends to return larger  $\chi$  than observed by AERONET, though it also contains substantial variability that the latter does not. This is likely due to the inaccurate measurement of laser energy, though this is under investigation. A calibration performed at 10:00 GMT was used throughout this day and that is the only AERONET measurement that was in any way input into the retrieval – the remainder are independent. Regardless, the retrieved values are equivalent to those given by the Ansmann algorithm, indicating that the retrieval is correct for the parameters it has been given. It is the calibration of the system, not the method of retrieval, producing the poor comparison.

### 4.3 Eyjafjallajökull ash

The eruptions of the Eyjafjallajökull volcano in Southern Iceland during April and May of 2010 produced the single most significant volcanic ash event over Northern Europe in the age of aviation. The closure of airspace cancelled around 100 000 flights, inconveniencing millions of travellers across the globe and resulting in massive losses for airlines and related industries. Owing to the density of personnel and instrumentation within the reach of this plume, it has become one of the most studied atmospheric events in history. The introduction of Johnson et al. (2012) provides a reasonable overview of the literature published to date and more will certainly be published over the years to come.

The CUV was operated, in addition to routine measurements, on 19 April to observe ash within the boundary layer. The optimal estimation retrieval (in extinction mode) was applied to these observations, shown in Fig. 16. Plot (a) presents the depolarization ratio observed by the EZ lidar while plots (c–d) show the retrieved backscatter and lidar ratio with errors outlined in plots (e–f). Ash particles have a large depolarization due to their asphericity and these measurements indicate the presence of a 400 m thick ash layer within the PBL, though the observed value of 0.1 is smaller than that expected for mineral dusts (0.35–0.37, Ansmann et al., 2011). It exhibits a low backscatter ( $< 10 \text{ Mm}^{-1} \text{ sr}^{-1}$ ) and lidar ratio (20–30 sr) compared to the remainder of the scene. These are well outside the range of 50–82 sr reported in the literature for similar ash in the free troposphere (Ansmann et al., 2010; Marengo and Hogan, 2011; Hervo et al., 2012). Combined with a smaller depolarization ratio, the retrieval indicates that the properties of the ash have changed significantly after 12–24 h within the PBL. The decreased  $B$  implies a growth of the particles, though this occurs without the influence of water as Raman observations indicate that the humidity is half its ambient value.

A mixed layer forms beneath the aerosol (see the vertical velocity in plot b). There,  $B = 50\text{--}80$  sr with minimal depolarization, which is broadly consistent with urban aerosols (Müller et al., 2007). Backscatter is fairly homogeneous throughout the

## Retrieval of backscatter and extinction from Raman lidar

A. C. Povey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

layer except during a period of updrafts around 13:00 GMT when  $\beta$  decreases from 10 to  $6 \text{ Mm}^{-1} \text{ sr}^{-1}$ . The absence of a similar change elsewhere in the PBL gives some confidence that this is a real variation rather than a calibration artefact.

A more weakly depolarizing aerosol resides in a poorly-mixed residual layer above the ash layer. It persists until 14:00 GMT when they mix. Backscatter and lidar ratios are large at the top of this layer and decrease with height. This could be simple stratification within a poorly-mixed layer or smaller particles may have concentrated at the top of the layer whilst larger particles have begun to settle, though it does not seem that sufficient time has passed to produce so large a gradient.

Finally, a thin layer of aerosol is present above the PBL (labelled in plot c). The EZ lidar did not resolve this, so no measure of the depolarization is available. Expressing lidar ratios of 40–60 sr with low  $\beta$ , the layer is consistent with aerosol typically observed at CFARR and there is no reason to label it as ash.

Figure 18 presents the distribution of retrieved extinction and backscatter for all points with a depolarization ratio measured to better than 100%. Lines of constant  $B$  are added for reference. Points likely to contain ash are shown in the left plot by filtering for depolarizations greater than 0.03. The residual layer appears as a concentration of points around  $\beta = 10 \text{ Mm}^{-1} \text{ sr}^{-1}$  and  $B \simeq 40 \text{ sr}$ . The mixed layer appears in the right plot as a more continuous distribution between  $B = 40$  and 60 sr. The failure of the retrieval near the surface is evident in a vertical line of points at  $\beta = 8 \text{ Mm}^{-1} \text{ sr}^{-1}$ . In the free troposphere,  $\beta < 1 \text{ Mm}^{-1} \text{ sr}^{-1}$ , where poorly constrained retrievals produce a broad distribution in both plots. There are very few observations of the thin ash layer, but their presence is evident in observations near  $B \simeq 20 \text{ sr}$  in the left plot not expressed on the right.

## Retrieval of backscatter and extinction from Raman lidar

A. C. Povey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



## 5 Conclusions

An optimal estimation retrieval scheme for aerosol scattering properties from Raman lidar observations was proposed, using the lidar equations as a simple forward model. The a priori state and covariance matrix were based on the properties of aerosol outlined in the OPAC model. Scattering was assumed to be vertically correlated between bins, decaying exponentially over a scale height of 100 m. This is smaller than observed by balloon-borne measurements but ensures that the PBL and free troposphere are not coupled.

The state of the atmosphere can be described at each height by the aerosol backscatter and either the extinction or lidar ratio. These possibilities were assessed by considering their ability to process simulated data. The lidar ratio configuration was found to lose sensitivity in the free troposphere, relying excessively upon its a priori assumptions, as shown by the disappearance of the averaging kernel. If extinction is retrieved instead, it and backscatter should be retrieved linearly with a correlation assumed between them (95 % here, though more investigation of this value is necessary). This configuration maintains sensitivity throughout the profile.

In the analysis of simulated and real data, the proposed retrieval is consistent with existing analyses. In addition, it returns a more rigorous estimate of the uncertainties. Backscatter was always retrieved at the finest resolution allowed (mostly 33 m, but this remains true at the instrumental limit  $< 10$  m) and with an uncertainty between 2 % in the most ideal circumstances and 20 % in the least. Extinction and the lidar ratio are less well constrained, expressing resolutions of 300–500 m in simulations and 0.1–1 km with real data. Importantly, these are different from the scale of vertical correlations assumed a priori and increase as SNR decreases. The retrieval has selected the most suitable resolution independently, unlike the smoothing filters used in most studies. The uncertainty in extinction retrieved from real data is relatively large ( $> 15\%$ ) but that is in part due to the short time scales evaluated (one minute in Figs. 14 and 16). The integration time can be increased to reduce these errors to any desired level (at least

## Retrieval of backscatter and extinction from Raman lidar

A. C. Povey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion





## Retrieval of backscatter and extinction from Raman lidar

A. C. Povey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

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## Retrieval of backscatter and extinction from Raman lidar

A. C. Povey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

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## Retrieval of backscatter and extinction from Raman lidar

A. C. Povey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

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## Retrieval of backscatter and extinction from Raman lidar

A. C. Povey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

- Sugimoto, N., Matsui, I., Shimizu, A., Nishizawa, T., Hara, Y., Chenbo, X., Uno, I., Yumimoto, K., Zifa, W., and Soon-Chang, Y.: Lidar network observations of tropospheric aerosols, *Proc. SPIE*, 7153, 71530A, doi:10.1117/12.806540, 2008. 9299
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## Retrieval of backscatter and extinction from Raman lidar

A. C. Povey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Table 1.** A priori values and uncertainties, as shown in Fig. 2.

		$\beta$	$\alpha$	$B$
Linear	Mean	$4 \times 10^{-6}$	$2 \times 10^{-4}$	58
	SD	$3 \times 10^{-6}$	$2 \times 10^{-4}$	7
Log	Mean	-12.4	-8.2	-
	SD	0.4	0.5	-



## Retrieval of backscatter and extinction from Raman lidar

A. C. Povey et al.

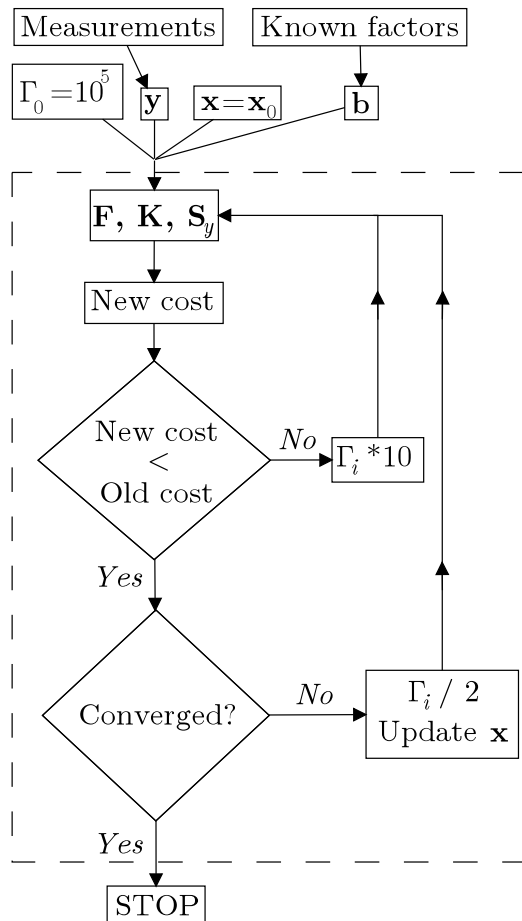


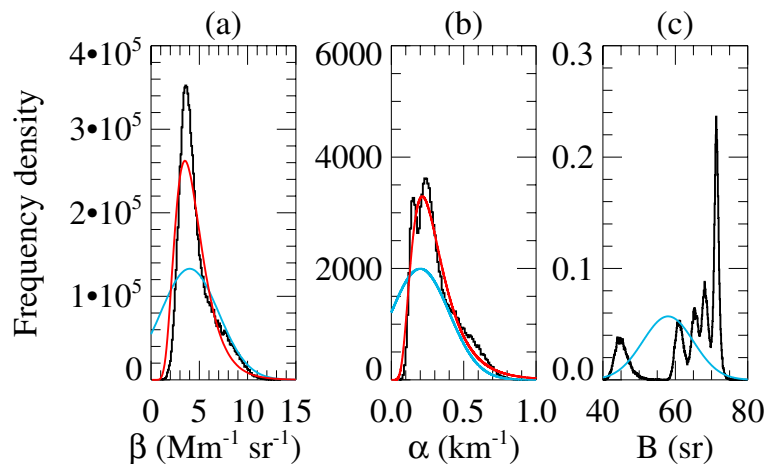
Fig. 1. Schematic of the optimal estimation retrieval algorithm.

Title Page	
Abstract	Introduction
Conclusions	References
Tables	Figures
◀	▶
◀	▶
Back	Close
Full Screen / Esc	
Printer-friendly Version	
Interactive Discussion	



## Retrieval of backscatter and extinction from Raman lidar

A. C. Povey et al.



**Fig. 2.** A priori aerosol distributions of **(a)** backscatter, **(b)** extinction, and **(c)** lidar ratio. The blue curve represents the linear retrieval of  $x$  and the red  $\ln x$ . Fit values summarised in Table 1.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

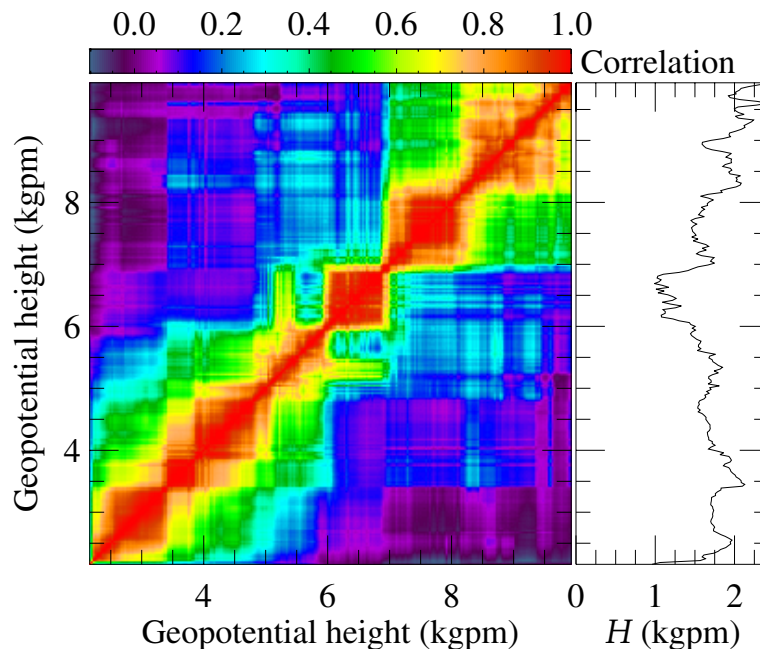
Full Screen / Esc

Printer-friendly Version

Interactive Discussion

**Retrieval of  
backscatter and  
extinction from  
Raman lidar**

A. C. Povey et al.



**Fig. 3.** The observed vertical correlation of backscatter. Left – Autocorrelation of backscatter with height, derived from 198 backscattersonde profiles collected between 1989 and 2000 at Laramie, WY, USA; Lauder, New Zealand; and Thule, Greenland. Box-like features are produced by layers of unusually large aerosol concentration during a single launch. Right – Least-squares fit of Eq. (14) to each row of that matrix for  $H$ .

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

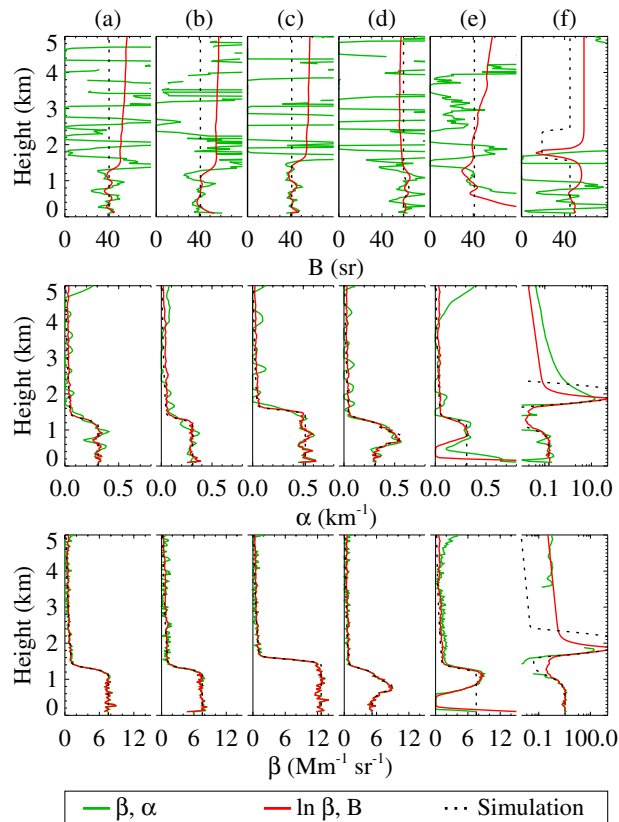
Full Screen / Esc

Printer-friendly Version

Interactive Discussion

## Retrieval of backscatter and extinction from Raman lidar

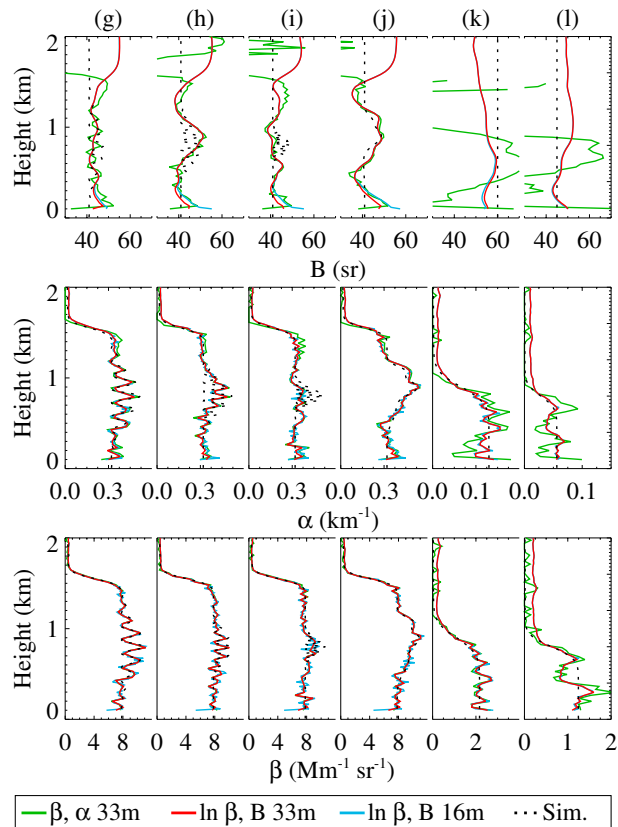
A. C. Povey et al.



**Fig. 4.** Performance of the retrieval with simulated data for the linear (green) and logarithmic (red) retrieval modes. **(a)** An idealised, well-mixed PBL with  $\chi = 0.50$ . **(b)** As **(a)**, but observed at 20% of the previous laser energy. **(c)** Similar to **(a)**, but with  $\chi = 0.89$ . **(d)** As **(a)**, but with a larger  $B$  and the addition of an aerosol layer at 800 m. **(e)** As **(a)**, but applying an incorrect nonlinear correction. **(f)** Observation of a cloud, shown on a log scale.

## Retrieval of backscatter and extinction from Raman lidar

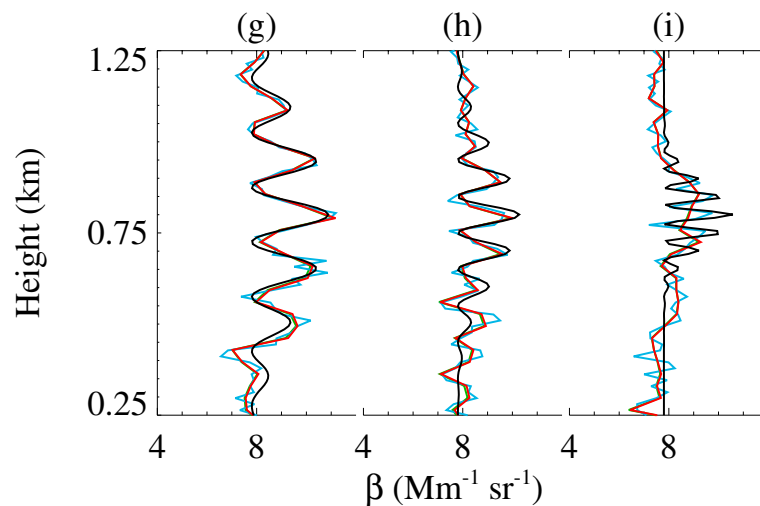
A. C. Povey et al.



**Fig. 5.** As Fig. 4 but highlighting the sensitivity to fine-scale fluctuations. The blue curve shows the logarithmic retrieval at twice the previous resolution. **(g)** As **(a)**, but with the addition of sinusoidal “aerosol layers” of width 300 m. **(h)** As **(g)**, but width 200 m. **(i)** As **(g)**, but width 100 m. **(j)** As **(a)**, but including three overlapping layers. **(k)** As **(a)**, but with  $\chi = 0.12$ . **(l)** As **(a)**, but with  $\chi = 0.04$ .

**Retrieval of  
backscatter and  
extinction from  
Raman lidar**

A. C. Povey et al.



**Fig. 6.** A closer examination of cases (g–i) of Fig. 5, highlighting how the coarse retrievals fail to capture the smallest features and that all resolutions tend to smooth the magnitude of the peaks.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

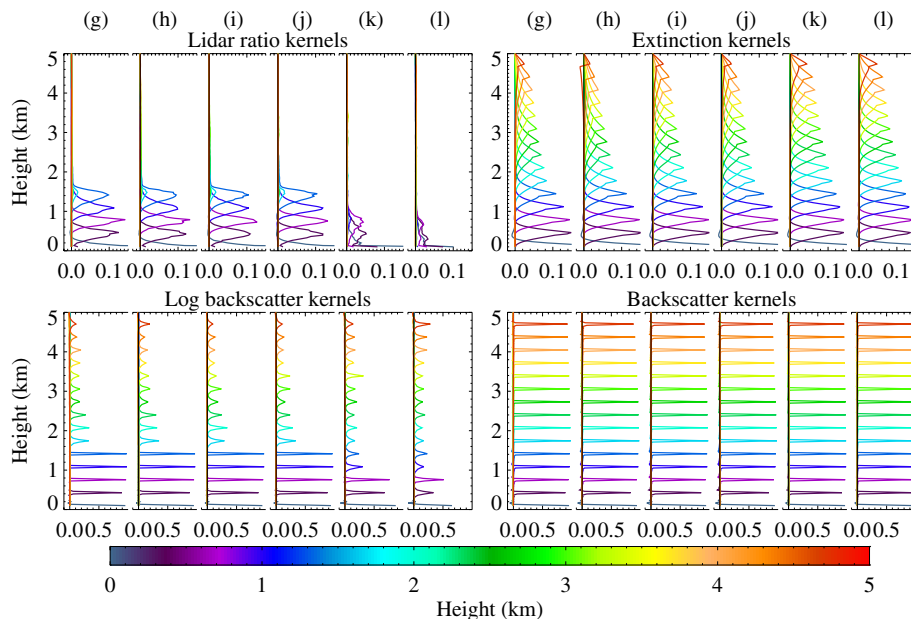
Full Screen / Esc

Printer-friendly Version

Interactive Discussion

## Retrieval of backscatter and extinction from Raman lidar

A. C. Povey et al.



**Fig. 7.** Selected rows of the averaging kernels for Fig. 5, denoting the relative contribution of the true state at each height (y-axis) to the value retrieved at a height denoted by the colour. Above colour bar: Lidar ratio configuration. Below: Extinction configuration.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

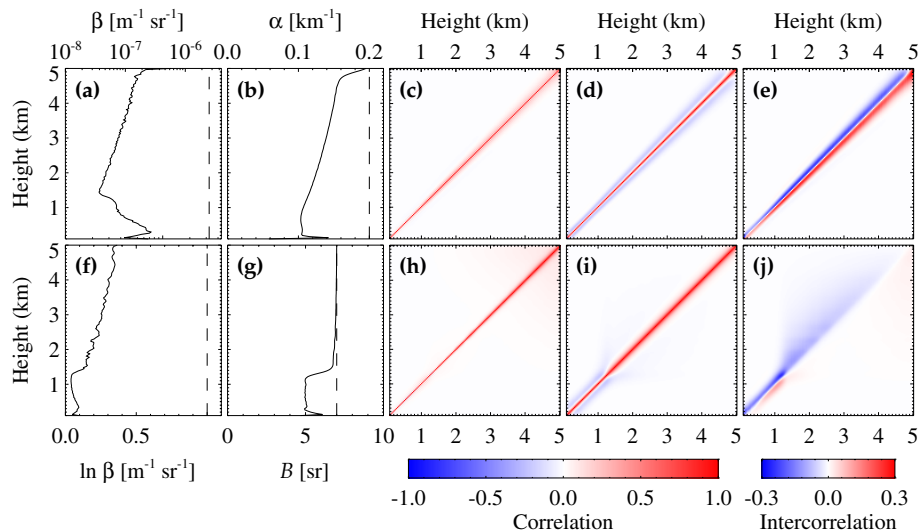
Full Screen / Esc

Printer-friendly Version

Interactive Discussion

## Retrieval of backscatter and extinction from Raman lidar

A. C. Povey et al.



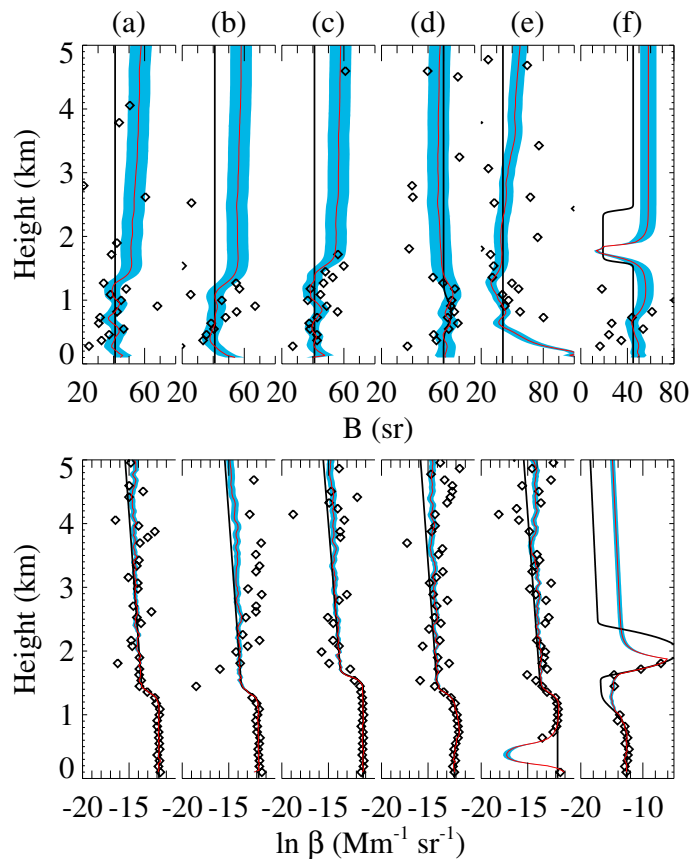
**Fig. 8.** Errors for the retrieval of case **(a)** by the linear and log modes (top and bottom rows, respectively). **(a)** Backscatter error. **(b)** Extinction error. **(c)** Backscatter autocorrelation. **(d)** Extinction autocorrelation. **(e)**  $\beta$  vs.  $\alpha$  intercorrelation. **(f)** Log backscatter error. **(g)** Lidar ratio error. **(h)** Log backscatter autocorrelation. **(i)** Lidar ratio autocorrelation. **(j)**  $\ln \beta$  vs.  $B$  intercorrelation.

[Title Page](#)
[Abstract](#)
[Introduction](#)
[Conclusions](#)
[References](#)
[Tables](#)
[Figures](#)
[◀](#)
[▶](#)
[◀](#)
[▶](#)
[Back](#)
[Close](#)
[Full Screen / Esc](#)
[Printer-friendly Version](#)
[Interactive Discussion](#)



Retrieval of  
backscatter and  
extinction from  
Raman lidar

A. C. Povey et al.



**Fig. 9.** Retrieval from the lidar ratio configuration (red) showing its error (blue) compared to that of the Ansmann method (diamonds) and the simulated profile (black).

Retrieval of  
backscatter and  
extinction from  
Raman lidar

A. C. Povey et al.

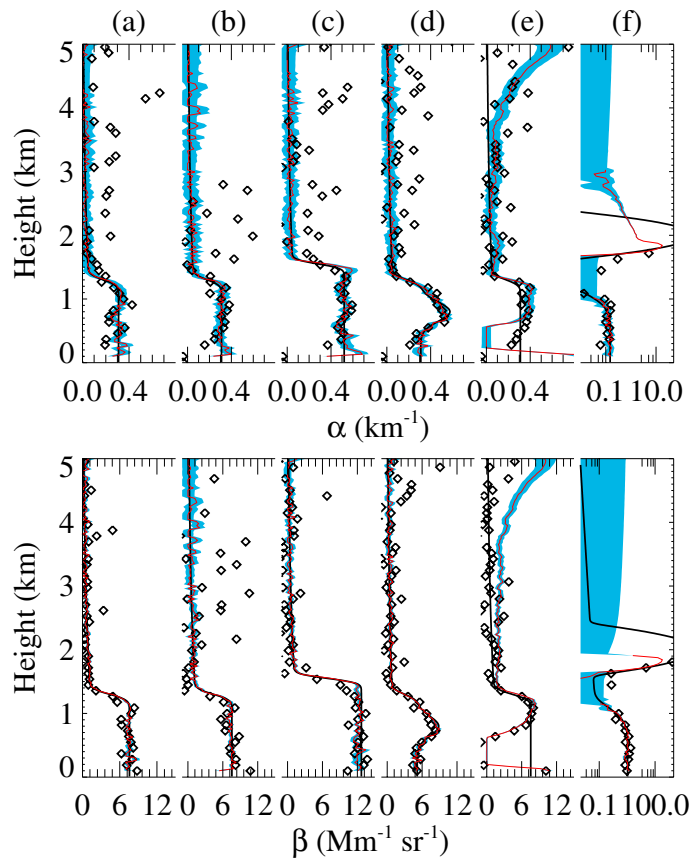
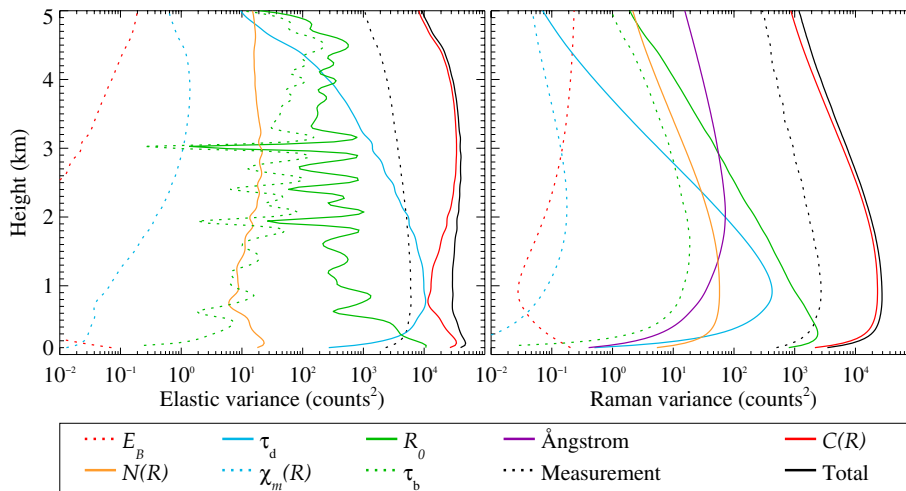


Fig. 10. As Fig. 9 but for the extinction configuration.

## Retrieval of backscatter and extinction from Raman lidar

A. C. Povey et al.



**Fig. 11.** The contributions of model parameters to the total retrieved variance.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

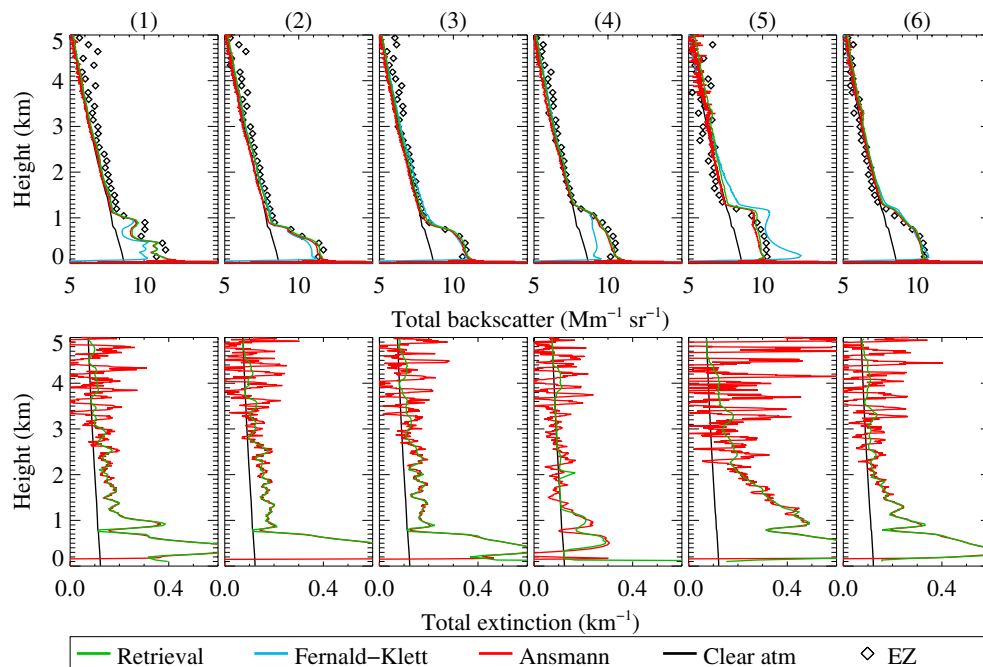
Full Screen / Esc

Printer-friendly Version

Interactive Discussion

## Retrieval of backscatter and extinction from Raman lidar

A. C. Povey et al.

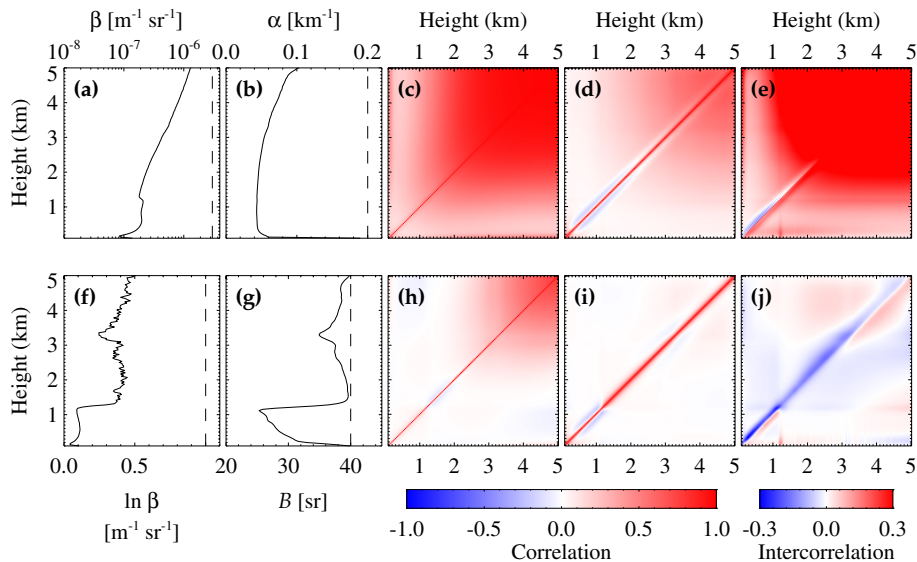


**Fig. 12.** Various estimates of total backscatter (top) and two-way extinction (bottom) for six analogue profiles observed by the CUV during March 2010. The attenuated backscatter coefficient  $\bar{\beta} \exp[-2(\chi + \chi^{(m)})]$  reported by the EZ lidar is shown in diamonds for comparison to the total backscatter. The scattering that would be observed from a clear atmosphere is shown in black, highlighting negative  $\alpha$  returned by the Ansmann technique.

[Title Page](#)
[Abstract](#)
[Introduction](#)
[Conclusions](#)
[References](#)
[Tables](#)
[Figures](#)
[◀](#)
[▶](#)
[◀](#)
[▶](#)
[Back](#)
[Close](#)
[Full Screen / Esc](#)
[Printer-friendly Version](#)
[Interactive Discussion](#)

## Retrieval of backscatter and extinction from Raman lidar

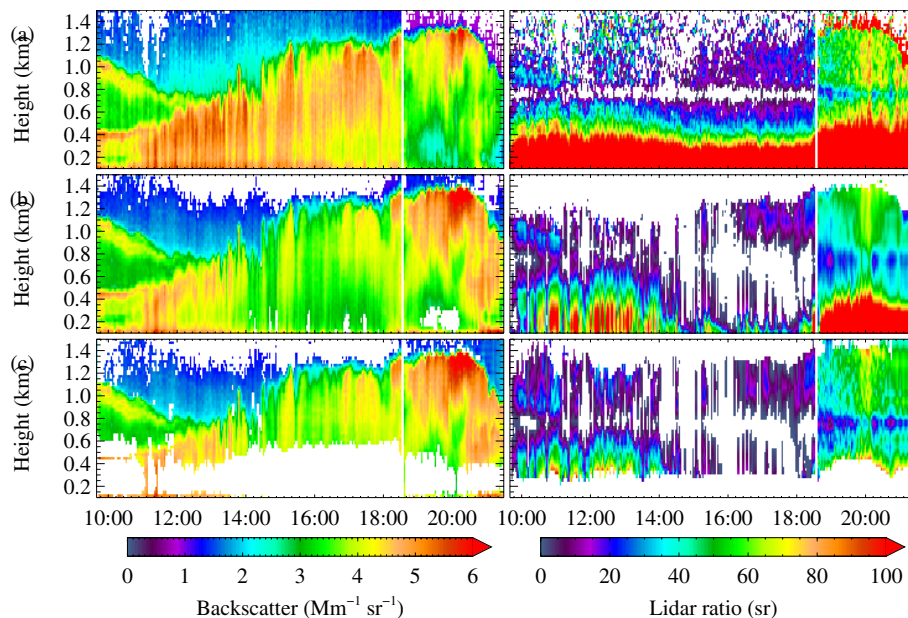
A. C. Povey et al.



**Fig. 13.** As Fig. 8, but for real data from case (1) of Fig. 12. The large correlations are a function of parameter error in  $E_B$ , which affects all levels equally and becomes dominant as the SNR decreases.

## Retrieval of backscatter and extinction from Raman lidar

A. C. Povey et al.

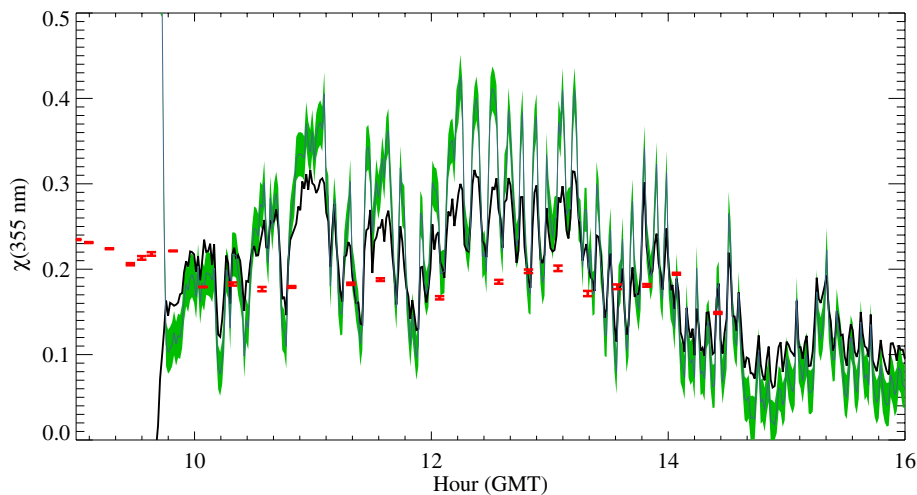


**Fig. 14.** The backscatter (left) and lidar ratio (right) retrieved from photon counting CUV observations on 2 March 2010. Results with an error greater than 20 % of  $\beta$  or 30 sr have been removed. **(a)** The Ansmann method after averaging the data over 30 m to give a similar resolution to the retrieval. **(b)** Retrieval of  $\beta$  and  $\alpha$ . **(c)** Retrieval of  $\ln\beta$  and  $B$ . The feature at 800 m in all lidar ratios is due to an inaccurate estimate of  $N$ .

[Title Page](#)
[Abstract](#)
[Introduction](#)
[Conclusions](#)
[References](#)
[Tables](#)
[Figures](#)
[⏪](#)
[⏩](#)
[◀](#)
[▶](#)
[Back](#)
[Close](#)
[Full Screen / Esc](#)
[Printer-friendly Version](#)
[Interactive Discussion](#)

**Retrieval of  
backscatter and  
extinction from  
Raman lidar**

A. C. Povey et al.



**Fig. 15.** Retrieved aerosol optical thickness at 355 nm (grey/green) compared to that observed by AERONET at level 2.0 (red) for 2 March 2010. The analogue Ansmann solution integrated between 0.25 and 2 km is shown in black.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

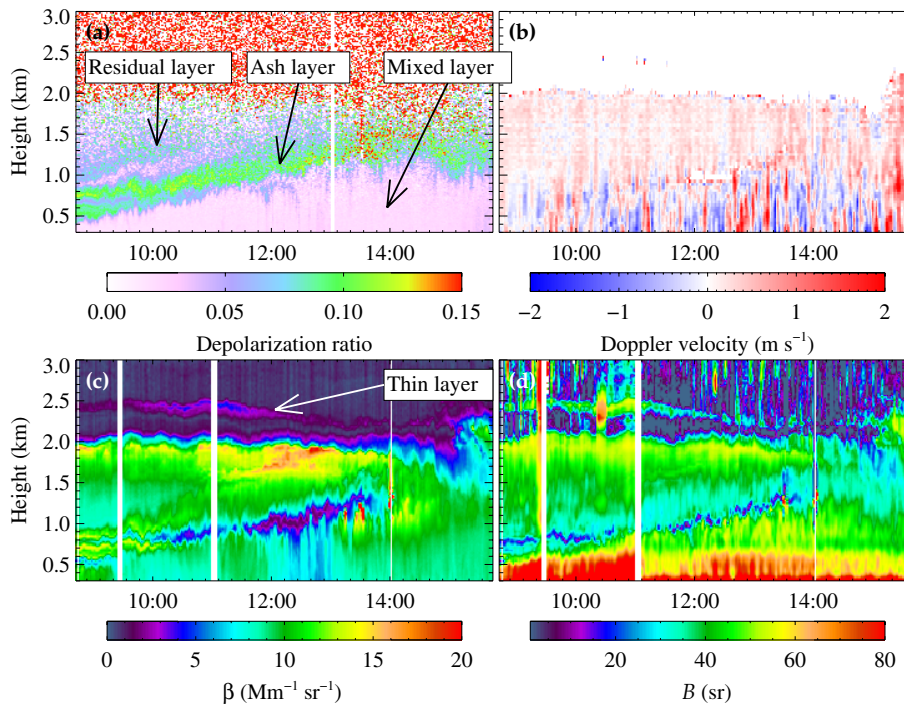
Full Screen / Esc

Printer-friendly Version

Interactive Discussion

## Retrieval of backscatter and extinction from Raman lidar

A. C. Povey et al.



**Fig. 16.** Observations of the Eyjafjallajökull ash plume at CFARR on 19 April 2010. **(a)** Depolarization ratio observed by the EZ lidar. Values above 1.9 km are dominated by noise. **(b)** Vertical velocity observed by a Halo Doppler lidar. **(c)** Backscatter retrieved (in the linear mode) from CUV measurements. **(d)** Lidar ratio retrieved from same.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Retrieval of  
backscatter and  
extinction from  
Raman lidar**

A. C. Povey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

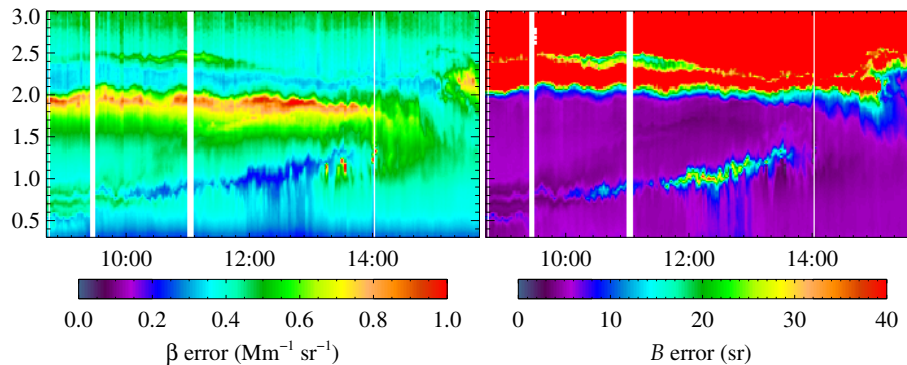
Back

Close

Full Screen / Esc

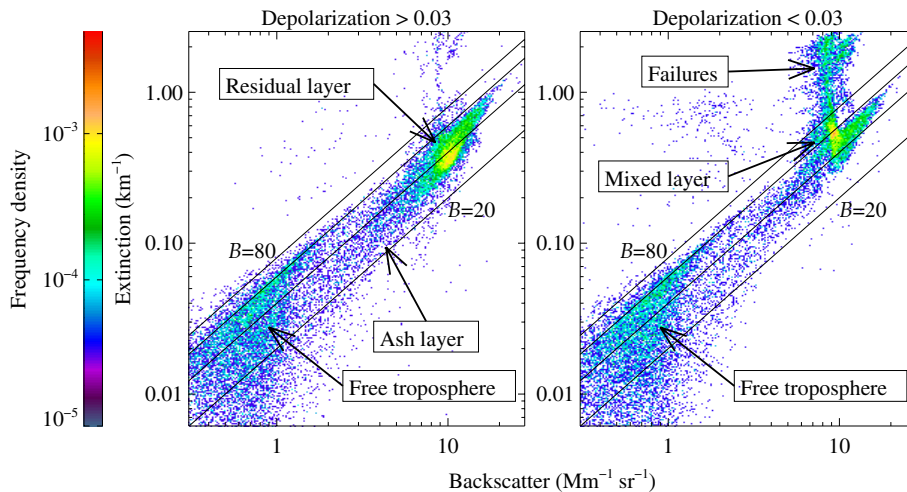
Printer-friendly Version

Interactive Discussion

**Fig. 17.** Retrieved errors for plots (c–d) of Fig. 16.

## Retrieval of backscatter and extinction from Raman lidar

A. C. Povey et al.



**Fig. 18.** Distribution of extinction and backscatter for 19 April 2010. Lines delineate lidar ratios of 20, 40, 60, and 80 sr. Left: Points observed to have a depolarization ratio  $> 0.03$ , which likely contain some quantity of ash. Right: The remaining points, corresponding to typical PBL aerosols and failed retrievals near the surface.