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Profiles of particulate matter size distributions using a balloon-borne lightweight aerosol spectrometer in the planetary boundary layer

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Abstract

A novel method for making cost-effective and reliable size-segregated (15 classes in the range 0.3–20 μm) vertical profiles of particulate matter up to 1000 m AGL in the planetary boundary layer is described and evaluated. The performance of a commercial miniature particle mass spectrometer is tested against standard gravimetric and optical instruments. The instrument was deployed on a 5 m^3 kytoon at two sites in the Lower Fraser Valley, British Columbia during the summer of 2001. Results point to significant differences between daytime and nighttime profiles as well as evidence that size distributions in elevated layers have signatures that reflect recirculation of daytime aerosols. Comparisons with lidar observations show good agreement with vertical profiles and indicate that the technique may provide an important aid to interpretation of lidar data.

Keywords: PM_{10}; Vertical profile; Air pollution; Mass spectrometer; Mass distribution; Lidar

1. Introduction

To date, most particulate matter observations have been made at ground level (Morawska, 1999) and in broad size ranges (generally $PM_{2.5}$, PM_{10} or total suspended particulate (TSP)). However, air pollution modelling and management requires an understanding of pollutant distributions in three-dimensions, especially within the lower troposphere and in particular the planetary boundary layer (PBL, generally <2 km above ground). Attempts to measure the vertical size-segregated distribution of particulate matter in the PBL have been constrained by the size and weight of instrumentation (difficult to deploy routinely on balloons). Successful measurements have been made from aircraft but at considerable expense, while attempts to establish vertical profiles with tethered balloon platforms have been limited to low altitudes ($\sim$300 m) and very large balloons supporting gravimetric samplers for $PM_{2.5}$ (Clark, 2000). Remote sensing techniques such as Raman lidar (Li et al., 2000) hold considerable promise for the examination of particle size variations in the lower troposphere but should be supported by in-situ observations. Balloon borne meteorological sensors have been used in the past as a complement to lidar measurements of aerosol distributions in the PBL (e.g. Hooper and Eloranta, 1986; Kolev et al., 2000). However, until recently fast response sensors capable of providing particle size distributions have been unsuitable for balloon deployment because of power and weight considerations.

The objective of this study is to describe and validate a novel approach to establish size segregated vertical profiles of particulate matter in the PBL using a balloon borne lightweight particle spectrometer. Measurements

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are made in tandem with lidar observations in order to both validate the new technique and to illustrate the utility of combined measurements. Potential applications of this technique include:

- Development of algorithms to provide optical closure from lidar and aid in interpretation of high resolution lidar imagery.
- Elucidation of processes affecting the production, transport, and distribution of airborne particulate matter. Specifically, examination of the diurnal evolution of mixing processes affecting fine particulate matter and the signature of aged polluted layer structures.
- Provision of observations for the development and validation of chemical/meteorological models used to test pollutant abatement strategies.

Results are presented from two days of observation in the Lower Fraser Valley (LFV), British Columbia as part of "Pacific 2001" field campaign in order to demonstrate the efficacy of this technique.

2. Air pollution meteorology of the LFV

The LFV (Fig. 1) has a rapidly growing population of 2.0 million mostly located in Greater Vancouver at the northwestern edge of the valley. Eastward, the valley becomes increasingly rural although several urban centres exist (most notably Abbotsford and Chilliwack). The valley floor is nearly flat, at an elevation of no more than a few hundred metres above sea level, while valley walls to the north rise to 2000 m above sea level within 10 km of the floor. The air pollution meteorology of the region is strongly influenced by local circulations including the land–sea breeze and valley/slope winds (McKendry et al., 1997; Steyn and McKendry, 1988). These circulations promote the development of pollutant layers during anticyclonic conditions together with a complex pattern of pollutant transport (McKendry and Lundgren, 2000).

While the meteorology, chemistry and distribution of gaseous photochemical pollution in the LFV are reasonably well known, particulate pollution in the region is less well understood. McKendry (2000) shows that PM_{10} concentrations in the LFV are relatively low when compared to larger urban regions such as the Los Angeles basin, other Canadian centres, Philadelphia, Birmingham (UK) or a range of urban or rural sites across Europe. Valley wide variations in annual mean concentrations are also relatively low and suggest that LFV PM_{10} concentrations are primarily modulated by meteorological conditions and within-valley sources. Meteorological conditions responsible for elevated particulate matter concentrations in the LFV identified by McKendry (2000) include:

(a) Short summertime periods (1–3 h duration) of reduced dispersion (light or calm winds and the development of a stable nocturnal boundary layer). Hourly concentrations may exceed 200 $\mu g\,m^{-3}$ but appear to be localised in extent.
(b) Summertime, anticyclonic conditions. Under these conditions, when photochemical pollutant concentrations (e.g. O_3) are also observed to build up, particulate concentrations may increase across the entire LFV to daytime values of 50–75 $\mu g\,m^{-3}$.
(c) Occasional wintertime "gap wind" events. These are limited to the eastern end of the LFV and are responsible for extended periods (multi-day) of elevated particulate concentrations (~ 100 $\mu g\,m^{-3}$) associated with the re-suspension of natural coarse-mode surface materials by strong winds.

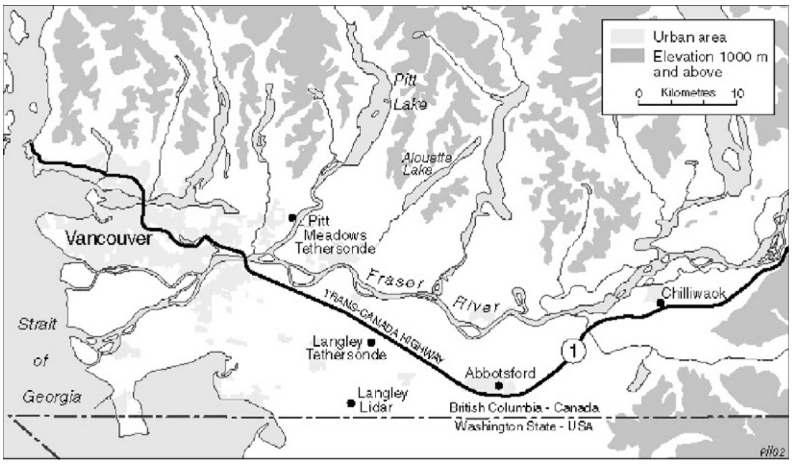

Fig. 1. The lower Fraser Valley, British Columbia.

Primary emissions of PM_{10} in the LFV are dominated by transportation sources (including road dust). Due to its isolated west coast setting and predominantly south-easterly surface winds, the LFV is seldom influenced by long-range transport of particulate matter.

The field study described herein was conducted during summer of 2001 at Langley and Pitt Meadows (Fig. 1). Observations at Langley were taken as part of the Pacific 2001 Field Study—an inter-agency collaborative study of PM_{10} and ozone in the LFV. Pitt Meadows is influenced by Vancouver's urban plume which may extend a considerable distance northward up the Pitt Lake tributary valley during summertime anticyclonic conditions (McKendry et al., 1997). At nighttime, drainage winds exiting the Pitt Valley have been shown to contain aged pollutants originating from the Vancouver side of the LFV (Banta et al., 1997). The nearest highway, and thus major local source, is situated approximately 1 km to the south of the site.

3. Methods

3.1. Aerosol spectrometer

Attempts at size segregated vertical profiling of the boundary layer using tethered balloon have in the past been hampered by weight and power constraints associated with available optically based instruments. The recent emergence of commercially available, miniature, battery-powered aerosol spectrometers has rendered such an approach feasible. The instrument chosen for the present study is the GRIMM *1.108* "*Dustcheck*" (GRIMM Labortechnik, 1996). With weight 2.4 kg (including rechargeable battery) and dimensions $24 \times 12 \times 6\,cm^3$ this instrument is within the payload capacity of a $5\,m^{-3}$ kytoon.

Ambient air is drawn into the instrument by a mass flow controlled pump and passed through a light beam produced by a laser diode. Scattering induced by particles of various sizes is measured by a photo-diode detector, amplified, and finally, binned to give the distribution of particulate matter in 15 different size classes from 0.30 to 20 μm. These data, in the form of particle counts, may be converted to a volume distribution (based on the particulate matter diameter) or a mass distribution ($\mu g\,m^{-3}$). In calculating the latter, particulate density information is required. Generally, this information is not available and so a uniform density is assumed. Consequently, calibration of the instrument is essential in order to accurately represent the particulate matter mass distribution at a given site. This may be achieved by comparing the GRIMM mass distribution with data collected using other measurement principles (e.g. gravimetric techniques).

3.2. Calibration and validation

Primary concern in deployment of the instrument was the variability in wind speeds (and to a lesser extent humidity) encountered during profiling and the impact this may have on the ability of the instrument to capture larger particles. The GRIMM is equipped with a variety of intakes suitable for different wind speeds (ranging from 0.5 to $4\,m\,s^{-1}$). Wind tunnel tests indicated no dependence on the inlet used for wind speeds in the range suitable for deployment of a $5\,m^3$ kytoon (0–$7\,m\,s^{-1}$). Consequently, the $4\,m\,s^{-1}$ inlet was used in all applications in the LFV. The manufacturer suggests that the GRIMM can only adequately dry sampled particles when relative humidity is below 95%. In applications described herein, the instrument was deployed in dry, clear sky conditions when moisture effects were considered negligible.

Instrument calibration is an important consideration as mass concentrations are estimated on the basis of a single assumed particle density. Furthermore, the GRIMM measures only particles with a diameter greater than 0.3 μm. As particle mass distributions typically show a mode at 0.1 μm diameter, it can be assumed that a portion of total mass is excluded from measurements. A comparison of GRIMM measurements with a collocated tapered element oscillating microbalance (TEOM), a standard gravimetric instrument) over a range of summer days shows that the GRIMM consistently underestimates mass concentration (Fig. 2a). However, this bias is not purely systematic. Concentrations for 18 June and 3 July show less deviation. Conditions for those two days were windy hence larger particles likely made up a larger proportion of the mass. GRIMM measurements therefore cannot be systematically adjusted with a single correction factor. However, by considering the mass and density signature for a given site during specific conditions a reasonably robust adjustment can be performed. As there is little variation in sources for a given site, an average density for a given particle size-range can be used to modify the GRIMM output. This would indeed provide a more accurate result than relying on an average density for the entire range. Such density signatures have been reported in Southern California (Chow et al., 1992; Kleeman and Cass, 1998; Watson 1994) and will be available for the LFV as a result of the Pacific 2001 Field Study. Mass signatures at a given site are likely to have greater variation. Specifically, they are likely to vary with day/night and calm/windy situations. These situations, however, provide standard enough mass signatures to allow simple calculation of correction factors suitable for a given condition. Correction factors may be calculated from a gravimetric instrument such as the TEOM or with a Teflon filter incorporated with the GRIMM.

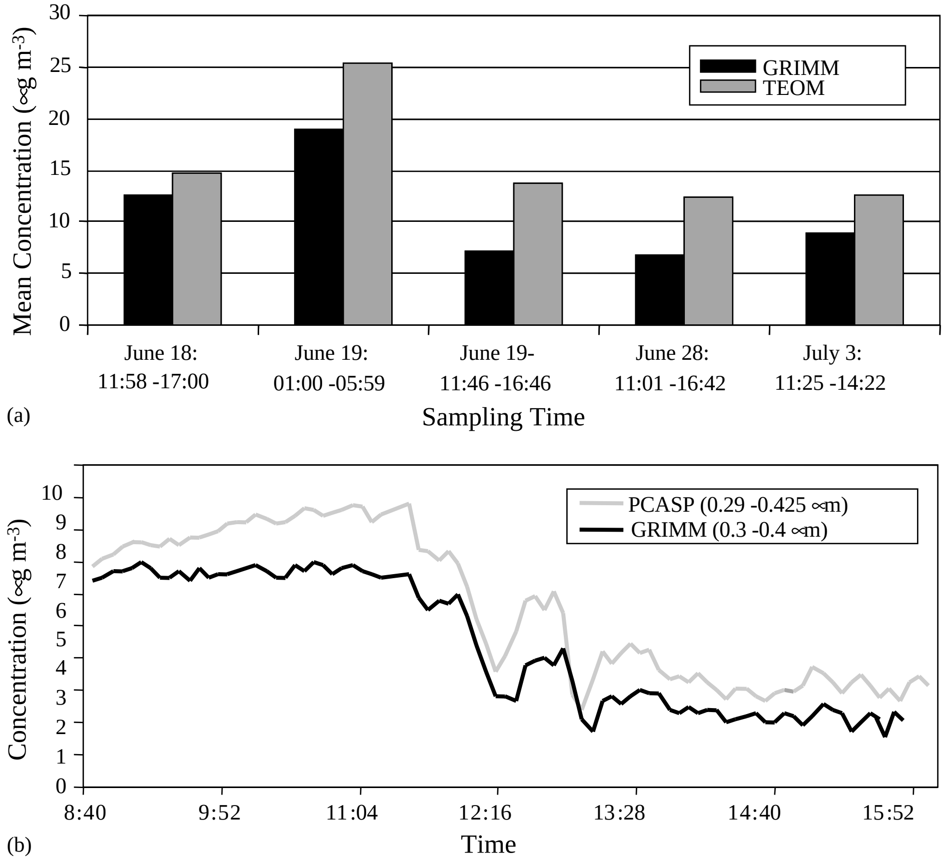

Fig. 2. (a) Gravimetric comparison between the GRIMM and a TEOM. (b) Time series comparison between the GRIMM and a PCASP. PCASP bin range exceeds GRIMM bin range by 0.035 μm.

Fig. 2b compares a time series obtained with the GRIMM to one measured with a Passive Cavity Aerosol Spectrometer Probe (PCASP; a standard optical particle counter that focuses on fine particles $\sim < 3\,\mu m$). The similarity of the two time series suggests that the *GRIMM* is able to accurately resolve temporal variations over short time scales (an important consideration for balloon deployment). The consistent negative bias from the PCASP series most likely results from the GRIMM's narrower size range. Although the instruments were compared for all corresponding bins up to 3 μm, other series indicated that the PCASP seriously underestimated particulate mass as size classes became increasingly coarse. Comparison with gravimetric measurements support this view. However, despite this deficiency, the PCASP comparison serves to confirm that the GRIMM provides reliable size information at a temporal resolution suitable for balloon profiling.

3.3. Tethered balloon

For vertical profiling through the boundary layer, the mini-aerosol-spectrometer was suspended approximately 1 m below a $5\,m^{-3}$ helium filled kytoon. In addition, an Atmospheric Instrumentation Research Inc. (AIR) Tethersonde (TS-3A-SPH) was slung immediately below the spectrometer in order to provide wind speed, wind direction, temperature, humidity and pressure approximately every 10 s during the ascent and descent sequence. Meteorological information was telemetered to a ground station while particulate matter data was recorded on a memory card in the spectrometer as 1 min averages. Timing information enabled the meteorological data and spectrometer data to be merged (most importantly, heights AGL could be established based on the pressure and temperature data). Ascent and descent of the balloon was controlled by an electric winch with a typical ascent/descent sequence reaching 1000 m AGL with a total duration of 70 min. This gave a vertical resolution for the meteorological data of 5 m and 28.5 m for the particulate matter data.

3.4. Lidar

A scanning lidar facility known as Rapid Acquisition SCanning Aerosol Lidar (RASCAL) was used to provide fast elevation scanning profiles of the lower troposphere with a resolution of 3 m along the laser beam axis (Strawbridge et al., 2001). The Langley lidar location is shown in Fig. 1. The technique of lidar (laser radar) provides a unique opportunity to obtain highly vertically resolved information on particulate matter with the added advantage of high temporal resolution. RASCAL data provides a comprehensive optical picture of the atmosphere in three dimensions. This helps to interpret the temporal variation of particulate matter optical properties within a widely inhomogeneous atmosphere. The basic components of a scanning lidar system consist of a laser, beam directing/collection optics and a telescope with a detection package to convert the signal into the appropriate information that can be processed, displayed and saved in real time. The laser source was a Continuum NY61 Nd:YAG laser operating at 1064 nm with a repetition rate of 10 Hz.The scanning optics consisted of a two-mirror design (24″ mirrors) with the first mirror fixed while the second mirror was allowed to rotate in both azimuth and elevation. The collection optics directed the light into a Celestron 14″ Schmidt-Cassegrain telescope with an 8 mrad field-of-view which focused the captured light onto a 3 mm RCA30956E avalanche photodiode (APD) and logarithmic amplifier made by OPTECH Inc. The signal was then sent to a GAGE 12-bit, 100 MHz PCI card where the information was digitized and stored. Although the scanning mirror is capable of scan speeds of 24°/s it was set to 0.2°/s (0.4°/s laser beam speed) at Langley and 0.1°/s at Pitt Meadows. With a 10 Hz laser (recently upgraded to 50 Hz laser) this corresponds to approximately 3 or 6 min for Langley and Pitt Meadows respectively for a typical elevation scanning profile from 3° above the horizon to 70°. The slow scan speed was chosen to improve the solid angle resolution at distances of several kilometers from the source. The lidar image shown here has been truncated to show only the lower portion of the scan for comparison purposes with the kytoon.

4. Results and discussion

4.1. Pitt Meadows

In total 30 profiles (a balloon ascent and descent comprises two profiles) of meteorological variables, 14 of which included the GRIMM, were made to approximately 1000 m AGL at Pitt Meadows in the period 26–27 July. Synoptic conditions were anticyclonic and conducive to the development of photochemical smog. The temporal resolution of profiles measured for 26/27 July was sufficient to interpolate a complete diurnal evolution of size segregated particulate matter. For brevity, only the diurnal evolution of PM_{10} in the vertical is presented in Fig. 3a. Considerable temporal variability in the vertical distribution and mass concentration of particulate matter throughout the day is shown. On the morning of 26 July, the nighttime accumulation of particulate matter below 300 m becomes diluted as the PBL grows. By 12:00 Pacific daylight time (PDT) the vertical profile appears well-mixed as the convective boundary layer grows to ~800 m. For the period from 12:00 to 16:00 PDT wind speeds vary between 1 to 2 m s^{-1} from the south. At approximately 16:00 PST, concentrations are observed to increase near the surface (<100 m AGL) corresponding to an increase in wind speed (4 m s^{-1}). This suggests local emissions, such as crustal material (due to higher wind speed) and exhaust from frequent traffic, not yet mixed by convective activity. An early collapse of the PBL appears to be responsible for this low-level accumulation remaining unmixed. By 1800 PDT, the PBL is already neutral and winds drop abruptly near the surface to near calm conditions. The lack of mechanical and free turbulence allows for this surface accumulation of particulate matter while winds of 6 m s^{-1} above 100 m seem to "clean out" the upper levels.

A stable boundary layer is established at approximately 20:00 PDT and corresponds to an abrupt 180° change in wind direction as well as the onset of a low level jet. The stable boundary layer grows to a maximum of 400 m throughout the night giving a reduced dilution potential. The nighttime increase in PM_{10} concentrations is disrupted by intermittent turbulence between 12:00 and 02:00 PDT. This breakdown is accompanied by the temporary disappearance of the low-level jet.

Although PM_{10} appears well-mixed throughout the PBL in the day, size-segregated profiles show this to be true only for the smaller size classes. Fig. 3b contrasts mass distributions at different altitudes during the day. Clearly, the concentration of particles greater than 2 μm decreases gradually with height. This suggests that particles of this size range tend to settle out by gravity. Particles with diameters of 0.4–2 μm are thoroughly mixed throughout the PBL while those ranging from 0.3 to 0.4 μm show slightly increasing concentration with height. Finer particles are therefore analogous to gaseous pollutants in their physical behaviour. Consequently, smaller particles are likely to have a longer atmospheric residence time and thus are more prone to recirculation. It is possible that the slight increase in concentration of the smallest size class with height may be due an increase in UV radiation with altitude allowing for more gas-to-particle conversion.

The nighttime mass distribution (Fig. 3c) shows significant differences to the daytime case. For all particle size classes, concentrations decrease with height up to 600 m. This may be attributed to the very stable boundary layer limiting the already weak mechanical turbulence (very calm conditions) and causing the

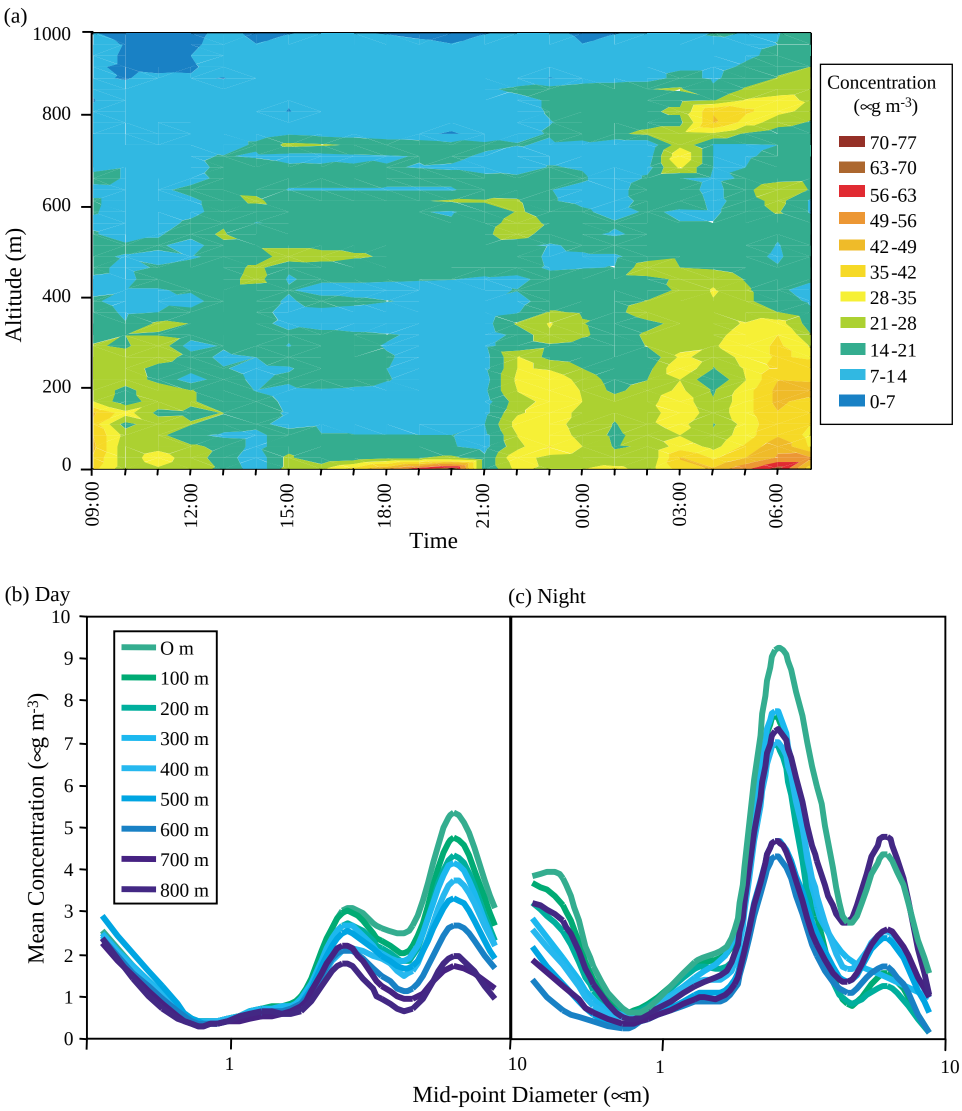

Fig. 3. Pitt Meadows: (a) Diurnal evolution of PM_{10} in the vertical (PDT). (b) Comparison of mass distributions with altitude in a well-mixed convective boundary layer. Distributions are averaged from profiles obtained between 14:45 and 16:00 PDT. Note that the 800 m distribution is slightly above the PBL. (c) Comparison of mass distributions with altitude in a very stable boundary layer. Distributions are averaged from profiles obtained between 03:25 and 04:44 PDT.

stratification of pollutants. At 700 and 800 m, high concentrations from an elevated layer are incorporated in the averaging. Interestingly, the nighttime size distributions have a dominant mode at 2.5 μm rather than the 8 μm mode observed during daytime. This may be the result of the combined effects of diurnal variations in stability and source strengths. For example, light winds at night reduce the entrainment of coarse surface materials. However, their atmospheric concentration may remain similar to daytime values due to reduced dispersion at night. By contrast emissions of fine particles from transportation sources may remain static or even increase into the evening resulting in higher concentrations when combined with reduced dispersion conditions.

Overall, the tri-modal size distribution observed at Pitt Meadows is consistent with signatures reported at similar sites i.e. traffic/urban-influenced sites with semi-rural surroundings. Modes at 0.2 and 2.5/3.0 μm are common for traffic/urban-influenced sites (Hidy, 1975; LeCanut et al., 1996; Morawska et al., 1998, 1999; Shi et al., 2001). A mode appearing at 8.0 μm for a site with

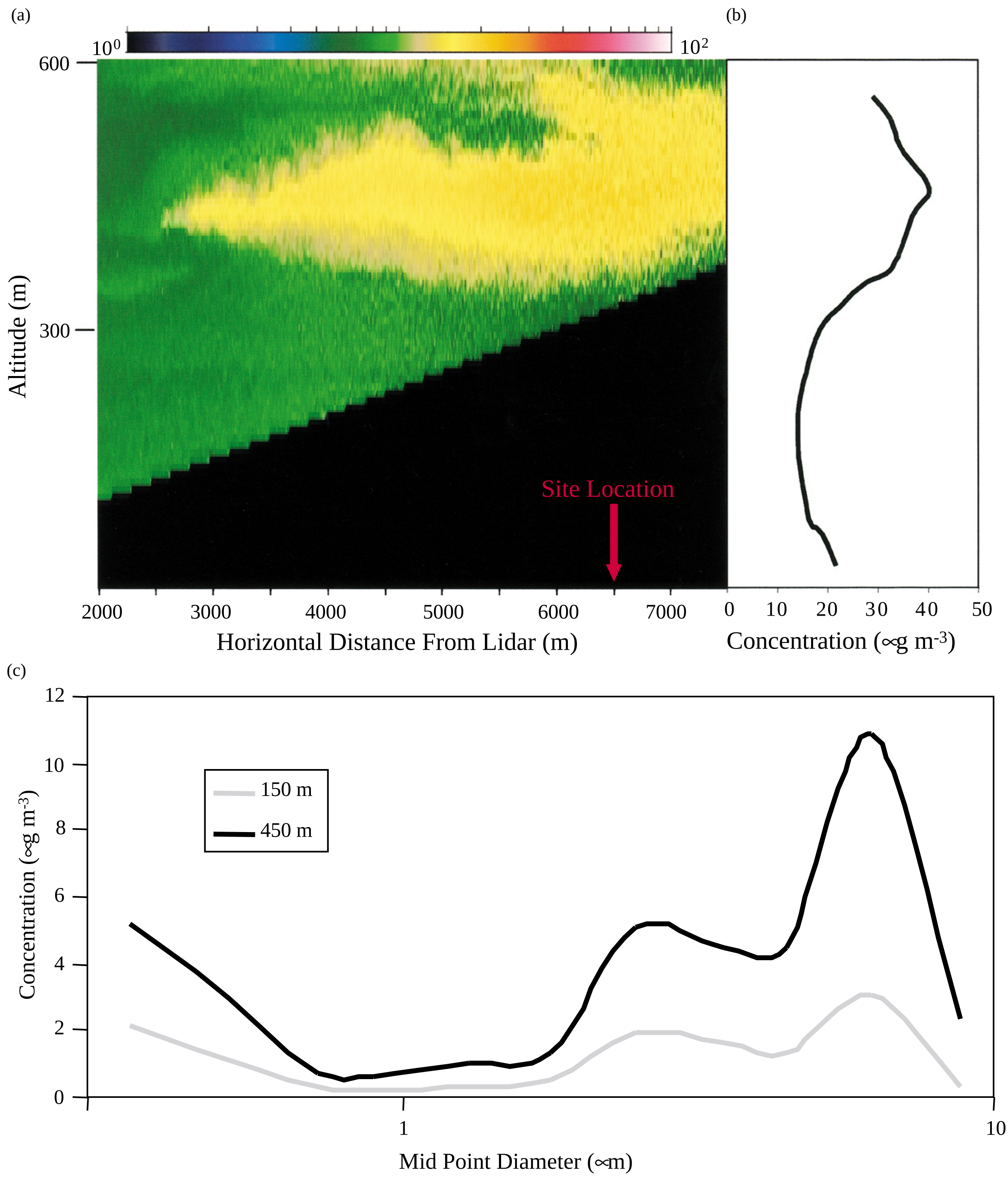

Fig. 4. Langley: (a) RASCAL image from 15 August 2001 at approximately 21:00 PDT showing layer aloft between 400 and 600 m. Note that the black represents the area below the lowest possible elevation scan angle obtainable due to local vegetation near the lidar site. The scanning range is from 0 to 12.3 km with a laser scan speed of 0.4°/s to provide sufficient resolution several kilometres from the instrument. (b) Vertical profile of PM_{10} measured between 20:41 and 20:58 PDT. (c) Comparison of mass signatures in the stable boundary layer and in the aged pollution layer (450 m).

semi-rural surroundings has also been observed by Hidy (1975). Although the signatures presented here are limited to particles with diameters greater than 0.3 μm, it is clear that a mode is present near the 0.3 μm cut-off.

4.2. Langley

Lidar imagery provides a good qualitative assessment of particulate matter pollution the PBL. It is therefore useful for validating in situ measurements, such as those provided by the tethered balloon. Fig. 4b shows a vertical profile obtained with the GRIMM on the evening of 15 August 2001. A significant increase in concentration is evident in a layer extending from 300 to 600 m. Fig. 4a shows the RASCAL image taken at the same time in a vertical slice that extends from the lidar facility through approximately the tethered balloon site 6.5 km to the north. The image clearly confirms the presence of an elevated layer at the same height and location as that detected by the GRIMM. Such elevated layers are well-documented in the complex terrain of the LFV where they have been attributed to residual layer development and daytime "venting" of pollutants along the heated slopes on the northern edge of the valley (McKendry and Lundgren, 2000; McKendry et al., 1997). At night, the development of drainage winds from tributary valleys and the broader land-breeze/valley wind circulation leads to complex advection patterns that transport pollutant layers within the valley (Banta et. al., 1997). The arrival of an elevated layer from the north at Langley, as in this case, is typical of such patterns during anticyclonic summer conditions.

In addition to providing confirmation of the GRIMM observations, the combination of scanning lidar imagery and vertical profiles shown here illustrates how the two methods may complement each other to enhance interpretation of complex three dimensional processes. In this case, the sequence of RASCAL observations showed the arrival of an elevated particulate layer from a northerly direction. Vertical profiles with the GRIMM confirm the presence of the layer (Fig. 4b) and indicate not only its mass concentration, but also show that the layer has a quite different size distribution to that of air in the stable layer below (Fig. 4c). The elevated layer has a signature similar to that of daytime boundary layer air (see Fig. 3b) and suggests that particulate matter in the layer is "aged" and probably originated from the daytime boundary layer. Meteorological profiles support this interpretation (Fig. 5). The profile of potential temperature (Fig. 5a) remains constant with height above 300 m and is indicative of a "residual mixed layer" that persists from the previous afternoon and is underlain by, and is decoupled from, the nocturnal stable boundary layer that develops below. This residual layer also shows enhanced specific humidity (suggestive of a daytime convective mixed layer) and light northwesterly winds in agreement with the RASCAL observations. Clearly, the combination of Lidar imagery, meteorological profiles, and mini-spectrometer profiles provides a powerful tool for determining the origin of elevated pollution layers.

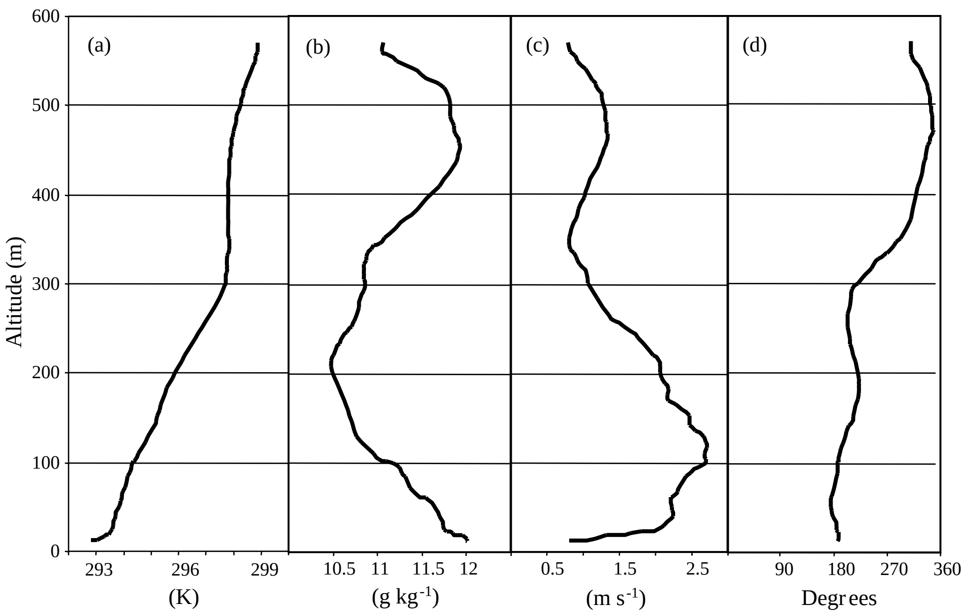

Fig. 5. Langley: Meteorological profiles taken between 20:41 and 20:58 PDT. (a) Potential temperature, (b) Specific humidity, (c) wind speed and (d) wind direction.

5. Conclusions

This study draws on observations from two days in the LFV to demonstrate the novel deployment of a commercial GRIMM mini-aerosol spectrometer on a tethered balloon. Although calibration of the instrument can be non-trivial, deployment of a GRIMM on a kytoon is shown to provide a relatively simple and cost-effective method of obtaining reliable size segregated vertical profiles of particulate matter in the 0.3–20 μm size range.

Observations using this method in the LFV are generally in agreement with previous accounts of the three-dimensional distribution of particulate matter in the boundary layer. Notably, fine material (<2.5 μm in diameter) tends to be well mixed vertically during daytime whereas coarse material tends to be found close to ground and sources due to gravitational settling. However, from the limited observations in the complex terrain of the LFV, it seems that the nighttime case may be more complex. In the stable nocturnal boundary layer all size classes tend to be trapped near ground. However, size distributions characteristic of aged day-time air masses may persist in elevated layers aloft. Further research is required to investigate the role played by such layers and the extent to which they may be mixed to ground.

Clearly, the ability to cost-effectively obtain regular size-segregated vertical profiles of particulate matter in the PBL has important applications. As shown here, the method has considerable potential to enhance the interpretation of lidar imagery. It may also contribute to improving understanding of the processes affecting the production, transport and dispersion of airborne particulate matter. Finally, information provided by this method can enhance the development and validation of models used to test pollution abatement strategies.

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