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A review of turbulence in the very stable nocturnal boundary layer and its implications for air quality

J.A. Salmond[1,*] and I.G. McKendry[2]

[1]Division of Environmental Health and Risk Management, Department of Geography, Earth and Environmental Sciences, University of Birmingham, Birmingham B15 2TT, UK
[2]Department of Geography, University of British Columbia, 1984 West Mall, Vancouver, B.C., V6T 1Z2, Canada

Abstract: Turbulence in the very stable nocturnal boundary layer is weak and typically characterized by intermittent bursts of activity. It often exists in isolated layers or pockets generated primarily from localized shear instabilities. As a result, turbulence is rarely in equilibrium with the conditions of the underlying surface. Given the layered structure of the nocturnal boundary layer, the spatial and temporal characteristics of turbulent activity (and resulting vertical mixing) can have a significant affect on local air quality at hourly to diurnal scales. However, while there is a wealth of information concerning turbulent processes operating during daytime conditions, until recently comparatively few studies have focused on the nocturnal case. Nevertheless the three-dimensional distribution of pollutants in the nocturnal boundary layer may have a significant impact on local pollutant budgets at a variety of temporal and spatial scales. This paper reviews recent progress in our understanding of the structure of, and processes operating in, the very stable nocturnal boundary layer. Then, drawing upon case studies from the Lower Fraser Valley, of British Columbia, Canada, it considers the implications of these developments for pollutant transport and surface air quality.

Key words: complex terrain, nocturnal boundary layer, ozone, turbulence.

I Introduction

The mean and turbulent characteristics of the planetary boundary layer play an important role in determining the transportation, storage and dispersion of atmospheric pollutants. The boundary layer is that part of the atmosphere that responds directly to the flows of mass, energy and momentum from the earth's surface, characteristically at timescales of an hour or less (Stull, 1988). Most pollutants are emitted or chemically produced within this layer and its diurnal evolution plays an important role in determining pollutant dispersion pathways and the

*Author for correspondence: E-mail: j.salmond@bham.ac.uk

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chemical properties of atmospheric pollutants (Zaveri *et al.*, 1995).

Stable stratification of the boundary layer develops shortly before sunset, when radiative cooling of the surface layers results in the development of a thermally stable layer (the stable boundary layer (SBL)) close to the earth's surface. With time, this layer becomes decoupled from the upper portions of the boundary layer and a residual layer (RL) remains aloft. The structure of the nocturnal boundary layer (NBL) is primarily determined by complex interactions between the static stability of the atmosphere and those processes (such as wind shear from synoptic or terrain induced flows or low-level jets) that govern mechanical generation of turbulence (Stull, 1988). These processes can operate at a variety of different heights and scales within the boundary layer and their dominance may vary in time and space. As a result, equilibrium or steady-state conditions are rare, and at any given point in time and space conditions may vary considerably with height. Thus the nocturnal boundary layer may range from fully turbulent to intermittently turbulent or even nonturbulent at a variety of heights, temporal scales and spatial locations. This makes it very difficult to observe and predict the transport pathways and diffusion of pollutants in the NBL, particularly in regions of complex terrain (Beyrich, 1994; Bowen *et al.*, 2000).

In contrast to the daytime convective boundary layer, our current understanding of turbulence in the NBL has been slow to develop (Nieuwstadt, 1984a; 1984b; Nappo and Johansson, 1999). This is largely due to the significant challenges associated with studying the NBL from observational, analytical and theoretical perspectives. As a result, individual field studies may be considered 'unrepresentative', or site specific, and are often limited by instrumental errors and uncertainties or trade-offs in experimental design. Nevertheless, in the last 20 years from the classic field experiments of Nieuwstadt (1977–79) in Cabauw, Netherlands (Nieuwstadt, 1984b) to the 1999 Cooperative Atmosphere-Surface Exchange Study (CASES) experiment in Kansas (e.g., Poulos *et al.*, 2002) a number of authors have tried to address this research gap and have yielded some important insights into the structure of, and processes operating in, the NBL.

Similarly, despite an 'explosion' in air quality monitoring and research worldwide (Derwent *et al.*, 1995) emphasis in much of the resulting literature concerning air pollution has been placed on daytime, surface-based scenarios. Under these conditions, concentrations of photochemical pollutants are typically well mixed and theory relating to turbulent transport in the convective boundary layer robust. However, the vertical and horizontal transport processes operating at night have been afforded relatively less attention. While surface measurements of pollutants have limitations (from a three-dimensional perspective) even for the case of fully developed turbulence, for the case of the layered NBL it becomes very difficult to make inferences from surface data about the nature and characteristics of processes operating aloft (Berkowitz *et al.*, 2000). Nevertheless, recent research suggests that processes operating in three dimensions throughout the stable NBL may have a significant impact on near-surface pollutant concentrations (Neu, 1995).

Given the importance of nocturnal turbulent processes to an understanding of the three-dimensional distribution of pollutants throughout the diurnal cycle, the primary objectives of this review are to:

- review recent developments in our understanding of the causes and characteristics of turbulence in the very stable NBL;
- identify the challenges associated with the measurement and analysis of weak, intermittent turbulence;
- describe the structure and vertical distribution of pollutants in a very stable NBL;
- relate recent advances in our understanding of nocturnal turbulent transport to diurnal patterns of pollutant concentration

using a range of examples from the Lower Fraser Valley (LFV) of British Columbia.

This review may be considered a companion to a previous report on processes affecting tropospheric layering of pollutants in urbanized, complex terrain (McKendry and Lundgren, 2000).

II The nocturnal stable boundary layer

In order to facilitate data analysis, modelling and the comparison of experimental results between different sites, the NBL is typically classified into two types or regimes: (i) weakly stable and (ii) very stable (Mahrt *et al.*, 1998). This is based on the turbulent characteristics of the surface layers and the two categories are described in Table 1. Although the definitions of 'very stable' and 'weakly stable' lack consistency between studies, Mahrt *et al.* (1998) identify prototype definitions of the stability regimes which aid classification of any given set of observed conditions.

The very stable regime develops primarily under anti-cyclonic conditions (when concentrations of pollutants can be expected to be highest) and is associated with light and variable winds and only intermittent turbulence in stable surface layers. Examples of the very stable NBL are given in many of the papers resulting from the 1999 CASES experiment in Kansas (Poulos *et al.*, 2002). By comparison, the weakly stable NBL develops under a more disturbed regime resulting in a continuously (if weakly) turbulent stable layer at the surface. This is typified by Nieuwstadt's (1984b) continuously turbulent NBL (Cabauw, Netherlands). In this paper, due to emphasis on pollutant transport during episodic periods of poor air quality (typically associated with anti-cyclonic conditions) we focus our discussion on the very stable case.

1 Depth of the stable layer

Many questions remain regarding the basic structure of the NBL. This is due in part to its variability in time and space, and in part due to limited measurements of turbulent fluxes with height (Mahrt *et al.*, 1979).Observations and model simulations point towards a layered structure of the NBL with a stable layer close to the surface and residual layer aloft (Derbyshire, 1994). Cooling in the RL predominantly results from advection or clear air radiative flux divergence. Thus the RL has

Table 1 Characteristics of different stability regimes in the nocturnal boundary layer (Mahrt *et al.*, 1998)

Characteristics	Weakly stable/continuously turbulent	Very stable/intermittently turbulent regime
Wind	Light to moderate, strong shear	Light and variable (>2 m s^{-1})
Cloud cover	Clouds frequently present	Clear skies
Radiative cooling	Weak	Strong, particularly close to the surface
Turbulence	Continuous in time and with height throughout the stable layer; consistently stronger and well established	Weak, spatial and temporal intermittent and can be isolated in layers above the surface; may be strongest at the top of the inversion layer
Structure of NBL	Textbook NBL; well-marked transition between stable layer and residual layer	Poorly defined structure; difficult to identify top of the stable boundary layer
Similarity theory	Applied with confidence	Inappropriate for the very stable case; routinely fails

similar characteristics to the mixed layer of the day before but is no longer continuously turbulent. Together, the SBL and RL form the NBL.

Given that the SBL and RL are essentially decoupled the depth of the SBL plays a significant role in determining the vertical distribution of atmospheric pollutants in the very stable NBL. However, although a number of different thermal, dynamical and turbulent criteria have been put forward to identify the top of the SBL and distinguish objectively the cut-off between the two layers (Beyrich and Weill, 1993), it is much more difficult to determine the top than for the convective boundary layer and to date no single definition has been accepted (Mahrt *et al.*, 1979; Beyrich and Weill, 1993; Beyrich, 1994).

Winds are typically light and variable near the surface in the SBL (Mahrt *et al.*, 1998) and primarily driven by local topographic flows, buoyancy, friction and entrainment processes. A wind speed maximum, which may develop into a nocturnal low-level jet (LLJ), is usually present at the top of the SBL. In the RL synoptic and mesoscale forcings such as advection or subsidence are important determinants of wind speed and direction. Discrete layers or bands of different velocities are common within the RL, and they may display considerable 'azimuthal meandering' or horizontal motion (Kurzeja *et al.*, 1991). Localized wind circulation systems may also develop which are isolated from the surface layers and which may be reconnected to the surface only with onset of convective turbulence in the morning (Bader *et al.*, 1987).

2 Characteristics of turbulence

Turbulence in the very stable NBL is typically intermittent, comprising of isolated clusters of eddies and rarely in equilibrium with surface conditions. It is often described as 'upside down' turbulence compared to the convective daytime case. This is because it is primarily driven by mechanical shear aloft, which then propagates downwards towards the surface where eddies are constrained by thermal stratification (Mahrt and Vickers, 2002). As a result turbulence can exist in isolated layers or pockets that may not penetrate to the surface. Such turbulence is highly localized in time and space, and is characterized by abrupt changes in variance (often by as much as an order of magnitude) within a short period of time (Coulter, 1990; Howell and Sun, 1999).

Theories of turbulence and wave structure in the NBL are poorly developed. Similarity theory and other stationary models currently used to interpret and predict the behaviour of the boundary layer cannot be easily applied to the very stable case (Finnigan *et al.*, 1984; Rogers *et al.*, 1995). Wyngaard (1973) introduced the concept of z-less scaling to scale turbulence when stratification is sufficiently strong that the turbulence is not primarily driven by or responding to surface characteristics. This local similarity theory applies well to the continuously turbulent nocturnal boundary layer (Nieuwstadt, 1984a; Holtslag and Nieuwstadt, 1986) but it is difficult to apply to the very stable case.

Near the surface in the very stable NBL a significant proportion of the vertical transport of heat, moisture and pollutants occurs in intermittent bursts (Mahrt, 1999; Howell and Sun, 1999; Poulos *et al.*, 2002). These bursts, associated with intermittent turbulence, occur sporadically in time and space (Coulter and Doran, 2002) and may occur several times during the course of the night (Weber and Kurzeja, 1991).

3 Causes of turbulence in the very stable NBL

Turbulence in the very stable NBL is typically mechanically generated as a result of shear associated with changes in wind velocity with height (Mahrt *et al.*, 1998). Various mechanisms/phenomena have been associated with initiation of turbulence.

- *Shear* created near the ground as a result of friction acting on the ambient flow (Stull, 1988).

- *Low-level jets*: Stull (1988) defines a low-level jet (LLJ) as a thin stream of fast-moving air in which wind speeds are greater than those above (and below) by 2 m s^{-1}. A variety of mechanisms may be responsible for such a wind speed maximum (commonly LLJs are ascribed to an inertial oscillation whereby winds accelerate to supergeostrophic values in the absence of friction at the top of the SBL). As a nocturnal LLJ develops above the SBL the localized increase in shear may generate bursts of turbulence (Beyrich *et al.*, 1996b; Corsmeier *et al.*, 1997).
- *Mesoscale wind systems* including land/sea breezes and slope/valley wind systems: in regions of complex terrain, thermotopographic wind systems (at night these are usually of the drainage or katabatic variety) permit the development of wind shear and the onset of turbulence (Haeger-Eugensson, 1999). The reversal of local wind systems (e.g., the switch from up-valley to down-valley winds) also causes mechanical turbulence through the NBL (Salmond and McKendry, 2002). Differences in the thermal characteristics of these flows may also result in the generation of convective turbulence if cool air is advected over a warmer surface.
- *Breaking gravity waves*: stable atmospheric environments support the development of buoyancy (gravity) waves (Stull, 1988). Turbulence can be generated by wave breaking or modulated by gravity waves which have a large amplitude compared to the depth of the boundary layer.
- *Density currents*: These are fast-moving bands of cold air initiated by distant cold fronts, drainage flows or mesoscale disturbances. As a result localized turbulence can occur within the density current as it passes over warmer ground (Sun *et al.*, 2002).

Few studies, however, have been able to associate turbulence measured at the surface directly with the source aloft (Poulos *et al.*, 2002). Indeed the identification of large-scale controlling mechanisms to account for the presence of intermittent turbulence have so far even eluded the participants of the CASES-99 project (Coulter and Doran, 2002) which is considered one of the most comprehensive nocturnal field experiments in terms of the vertical and horizontal scale of boundary layer and turbulence measurements.

III Analysis of turbulence in the very stable NBL

Turbulence measurement techniques and traditional data processing methods (such as the Fourier Transform) have limitations when applied to turbulence in the very stable NBL (Mahrt, 1985). Turbulence is typically measured using micrometeorological instruments mounted on a tower above a particular surface. Turbulence in the very stable NBL is weak and often near the limits of instrument sensitivity. To date, there are no accepted, objective and systematic methods to distinguish between instrumental noise and plausible physical behaviour (Vickers and Mahrt, 1997). Critical values must therefore be chosen to exclude contaminated data. Inappropriate selection of data may lead to sampling errors or result in the systematic exclusion of low wind speed conditions thereby creating a bias towards strong wind cases (and excluding the very stable NBL).

Information from high-frequency micrometeorological instruments can be used to calculate surface fluxes (transport of heat, moisture, momentum or pollutants per unit area per unit time) from an area upwind of the tower – the flux 'footprint' (Schmid, 1994; Horst and Weil, 1994; Kharabata and Schuepp, 1999). However, under cloudless nocturnal conditions typically associated with the very stable NBL, significant fluxes and vertical advection patterns can preferentially develop at isolated locations corresponding to stationary updrafts (Sun *et al.*, 1997). This is accentuated in regions of complex terrain where stationary features can be generated with convergence of drainage flows or by surfaces with different thermal properties (Mahrt, 1998). Isolated towers cannot detect these stationary eddies (Lee and Black, 1994) resulting in underestimation of turbulence in the very stable NBL.

Temporal variability in turbulence (expressed both in terms of intermittency and nonstationarity) represents perhaps the most significant challenge to the interpretation of atmospheric data and the calculation of turbulent fluxes in very stable conditions (Smedman *et al.*, 1995; Foken and Wichura, 1996; Howell and Sun, 1999). Fluxes describe the turbulent transport of a quantity per unit area with time. In order to calculate a turbulent flux accurately, data recorded at a minimum of twice the frequency of the shortest wavelength (the Nyquist frequency) are averaged over a specified time period. The turbulent component is then generated as the difference between the mean state of the variable through the time period and each individual data point. The flux is equal to the mean product of the turbulent component of (for example) a scalar quantity with the turbulent component of either the horizontal or vertical transport term. This is the basis of the eddy-covariance method of calculating scalar fluxes.

Defined as changes in the mean state over the chosen time period, nonstationarity may result from a variety of atmospheric forcings including mesoscale circulatory systems (Mahrt *et al.*, 1996), diurnal trends in the boundary layer (Mahrt, 1998) and variations in cloud cover (McMillen, 1988). Nonstationarity can affect the computed flux at scales larger than turbulence. Although when averaged over many records the effects may be negligible, at an individual scale contamination can be pronounced (Mahrt, 1998). Nonstationary records are ideally omitted from the data record because the calculation of the turbulent and mean component of the flux is flawed. This renders flux calculations in nonstationary data sets particularly sensitive to averaging length, detrending and filtering procedures (Mahrt, 1985). The relative importance of the various sources of errors in the data set cannot be easily isolated (Mahrt, 1998).

Quantitative estimates of fluxes are also contaminated with sampling problems generated by the intermittency of the turbulence (Mahrt, 1985). Observational studies by Weber and Kurzeja (1991) identify periods of isolated turbulence lasting 5–30 minutes near the surface in the NBL. Thus within any given 30-minute sampling period only a small portion of signal may be turbulent. This limits the number of eddies sampled and hence the statistical validity of the flux calculation. This also makes the physical interpretation of the spectra and cospectra difficult because the characteristics of the turbulence spectra are obscured by nonturbulent portions of the time period (Mahrt, 1985).

Recent studies by Hartogensis *et al.* (2002) in the CASES-99 field experiment and de Bruin *et al.* (2002) in Uppsala, Sweden, have demonstrated for the first time the potential value of a different type of instrument for measuring turbulence in the very stable NBL – the scintillometer. The basic premise behind scintillometery is that atmospheric turbulence attenuates the radiation from two parallel laser beams. This results in changes to two parameters measured by the receiver from which turbulent fluxes of heat and momentum can be calculated using Monin-Obukov Similarity theory. In this way, scintillometers offer the ability to make path-averaged measurements of turbulent fluxes of heat and momentum, and provide an alternative approach to obtaining more spatially representative data sets in the NBL.

Scintillometers have been widely used in the daytime convective boundary layer in rural areas but their potential application in the NBL has been largely ignored. The increased spatial sampling of these measurements compared with those from standard eddy correlation instruments enables the use of shorter averaging times for flux calculations. This is a significant advantage when considering the nonstationary, intermittent characteristics of turbulence in the very stable NBL. As a result both de Bruin *et al.* (2002) and Hartogensis *et al.* (2002) highlight the superiority of the technique over traditional eddy-covariance techniques when considering turbulent fluxes over timescales of 10 minutes or less in the NBL.

New analytical techniques for quantitatively analysing turbulent data have also emerged in the last 20 years that have had a significant impact on our ability to quantify turbulent fluxes. Perhaps the most promising of these is wavelet analysis. Wavelet analysis differs from other quantitative analytical tools that have been used to analyse turbulent time series (such as Fourier analysis) primarily in that it is a local transform, and the analysis takes place at a variety of different scales. This enables information about the temporal location of different features (characterized by different frequencies) within the data set to be retained. In this way the wavelet analysis can be said to 'zoom in' on a particular feature of the signal and studied locally with a level of detail corresponding to the scale of the feature. Thus the technique has been described as a 'mathematical microscope' (Hubbard, 1998) and can be used to filter the signal, thus effectively isolating the turbulent signal from other processes, such as gravity waves or background noise that contaminate the data set.

The local characteristics of the transform also render the technique ideal for calculating fluxes and analysing turbulence in nonideal conditions when the time series is expected to be nonstationary. Wavelet analysis essentially acts like a local moving average window and thus is less sensitive to long-term nonstationarities in the data set and well suited to the intermittent turbulence characteristic of the very SBL. Howell and Mahrt (1997), Howell and Sun (1999) Cuxart *et al.* (2002) provide just a few examples of the successful application of wavelet techniques to the analysis of turbulence in the nocturnal boundary layer. Wavelet analysis has also been used to provide insights into the complex interactions between gravity wave activity and turbulence (Sato and Yamada, 1994).

IV Pollutant impacts

High concentrations of photochemical pollutants are typically associated with anticyclonic conditions when strong temperature inversions and light winds limit vertical and horizontal dispersion. During the day under these conditions convective turbulence can be expected to mix pollutants emitted within the boundary layer and concentrations of pollutants are typically comparatively uniform with height. At night, however, the layered structure of the very stable NBL can result in a much more complex vertical distribution of pollutants. For example, since the SBL and the RL are essentially decoupled from each other, pollutant emissions, chemical (production and removal) processes and horizontal and vertical transport processes within the two layers may result in quite different pollutant profiles with time through the night. Pollutants emitted into the surface layer (such as carbon monoxide, nitrogen monoxide and carbon dioxide from vehicular emissions) may become trapped close to the surface within the nocturnal inversion and can be expected to increase with time. This is a result of continuous emissions of pollutants into a comparatively small volume of air that is characterized by limited horizontal and vertical dispersion. In contrast, concentrations of these pollutants are likely to remain low in the residual layer due to the absence of emission sources and isolation of the RL from the SBL. Other pollutants emitted from chimneystacks above the nocturnal inversion into the RL may be stored within the residual layer aloft and not penetrate into the SBL. Thus it is very important to study the pollutant concentration of the NBL in three dimensions (vertical as well as horizontal) in order to get a true picture of pollutant dispersion in the very stable NBL (Hastie *et al.*, 1993).

Some of the earliest studies of variation of pollutant concentration with height in the NBL were those of van Dop *et al.* (1977), Harrison *et al*. (1978) and Garland and Derwent (1979). These studies (focused on ozone air pollution) demonstrated that ozone concentrations increased with height through the NBL (Figure 1a, profile 2). Ozone, a highly volatile secondary pollutant, is formed in the atmosphere as a result of the photodissociation of nitrogen dioxide. This chemical reaction can occur only in the presence of ultraviolet light and thus there are no known sources of ozone in the NBL. However, ozone is rapidly removed from the surface layers of the boundary layer as a result of deposition onto the earth surface or chemical titration with nitrogen monoxide (a pollutant primarily associated with vehicle exhaust emissions). Thus we might expect to see ozone concentrations decrease to near zero with time at the surface (Figure 1b), while ozone concentrations aloft are expected to remain at concentrations which resemble the convective boundary layer from the previous day (Figure 1a, profile 2).

However, detailed studies of hourly variations in ozone concentration near the surface in the very stable nocturnal boundary layer reveal that superimposed upon the general trends in concentration described above a number of temporally localized spikes in concentration are frequently observed (Reitebuch *et al.*, 2000; Seibert *et al.*, 2000). These spikes in ozone concentration (also known as nocturnal ozone maxima) have been observed in the very stable NBL in a range of different environments from the rural plains of Germany (Corsmeier *et al.*, 1997), inner urban areas of Essen (Strassburger and Kuttler, 1998), mountaivalley location of Freiburg (Baumbach and Vogt, 1999; Kalthoff, 2000) the Alps (Prevot *et al.*, 2000), Jerusalem and Tel Aviv (Steinberger and Ganor, 1980), to the northeastern USA (Samson, 1978), Chicago area (Coulter, 1990) and the Lower Fraser Valley, British Columbia (Salmond and McKendry, 2002).

These temporally localized spikes in surface concentration are typically thought to be the result of vertical mixing due to intermittent turbulent coupling between the SBL and RL (Figure 1, profile 4). Wavelet analysis has helped to provide insight into both the characteristics of the turbulent bursts and their relationship to vertical mixing processes (Salmond, 2005). For example, the results from wavelet analysis of a vertical velocity time series (Figure 2b) are given in the scalogram in Figure 2a. The scalogram provides a plot of the wavelet coefficients for a variety of different scale analyses. Lighter colours indicate a high degree of correlation between the wavelet shape at any given scale and point in time. Since the wavelet chosen for analysis in Figure 2 is the haar wavelet, we would expect to see lighter colours associated with marked jumps in the time series. Comparison of Figure 2a with Figure 2b shows a cluster of lighter colours at high frequencies (small scales) between 0005 and 0015 Pacific Daylight Time (PDT) indicating the presence of turbulent activity at this time on the night of 31 August and 1 September 1998.

As shown in Figure 2c the burst of turbulent activity coincides with a localized increase in ozone concentration. Given that no coincident shift in wind speed or direction occurred at the surface at this time, these results strongly suggest that increased ozone concentration at the surface resulted from vertical mixing processes associated with the burst of turbulence. Indeed this event formed part of a series of intermittent bursts of turbulence that lasted between 2300 and 0130 PDT. These ideas are explored further in Salmond (2005). The spatial extent of this turbulent event can be deduced from the network of ozone monitors throughout the Lower Fraser Valley during this time period (Figure 3). Spikes of varying magnitudes were observed simultaneously at Burnaby South, Surrey East, Langley, Aldegrove, Chilliwack and Golden Ears. At other sites, the absence of a nocturnal spike is potentially attributed to (a) chemical destruction of ozone by NO, (b) a lack of vertical mixing, or (c) a lack of available ozone in the RL for down-mixing (Salmond and McKendry, 2002).

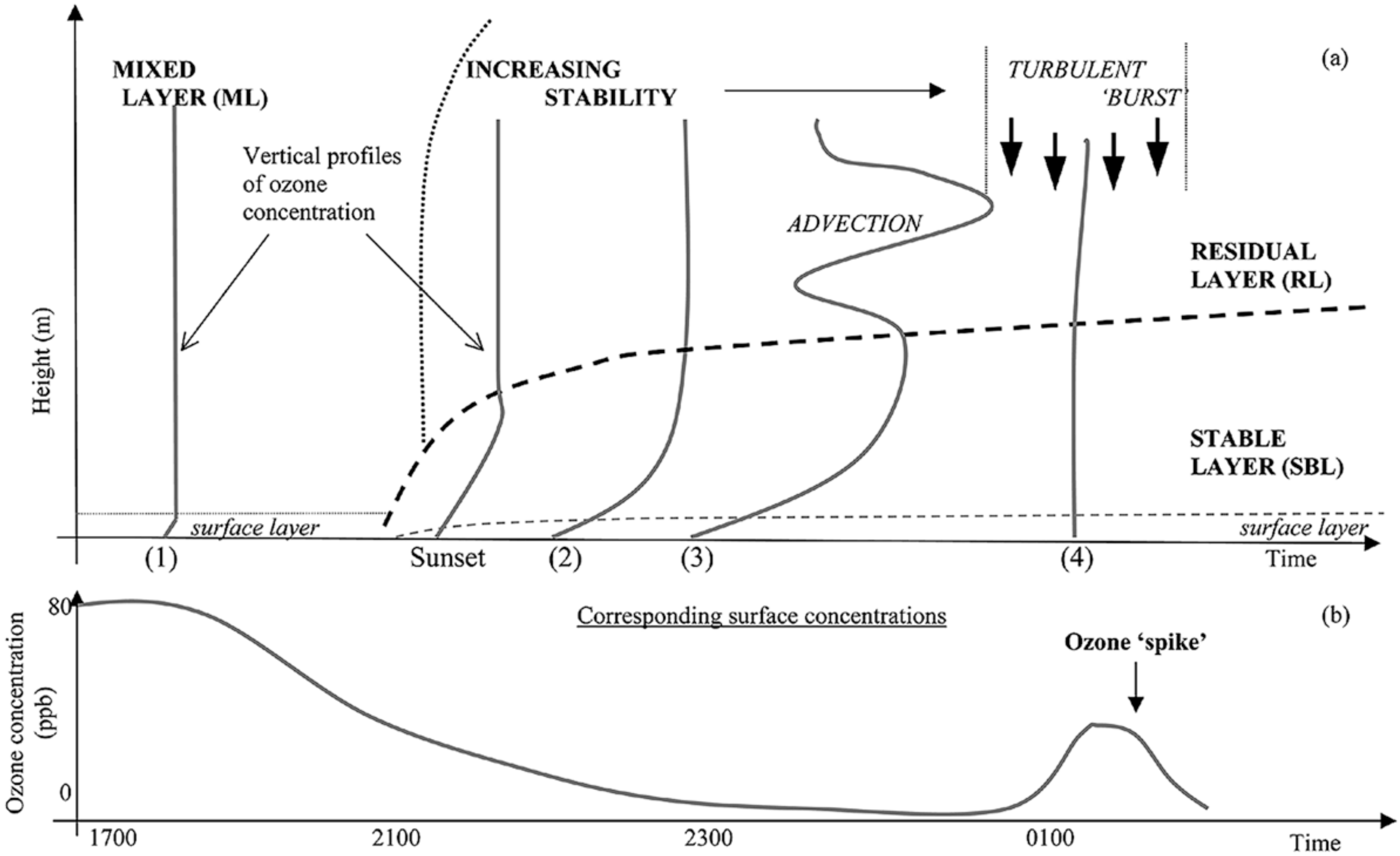

Figure 1 (a) A conceptual model to describe the evolution of the nocturnal boundary layer (NBL) and vertical distribution of ozone and (b) the corresponding changes in surface ozone concentration

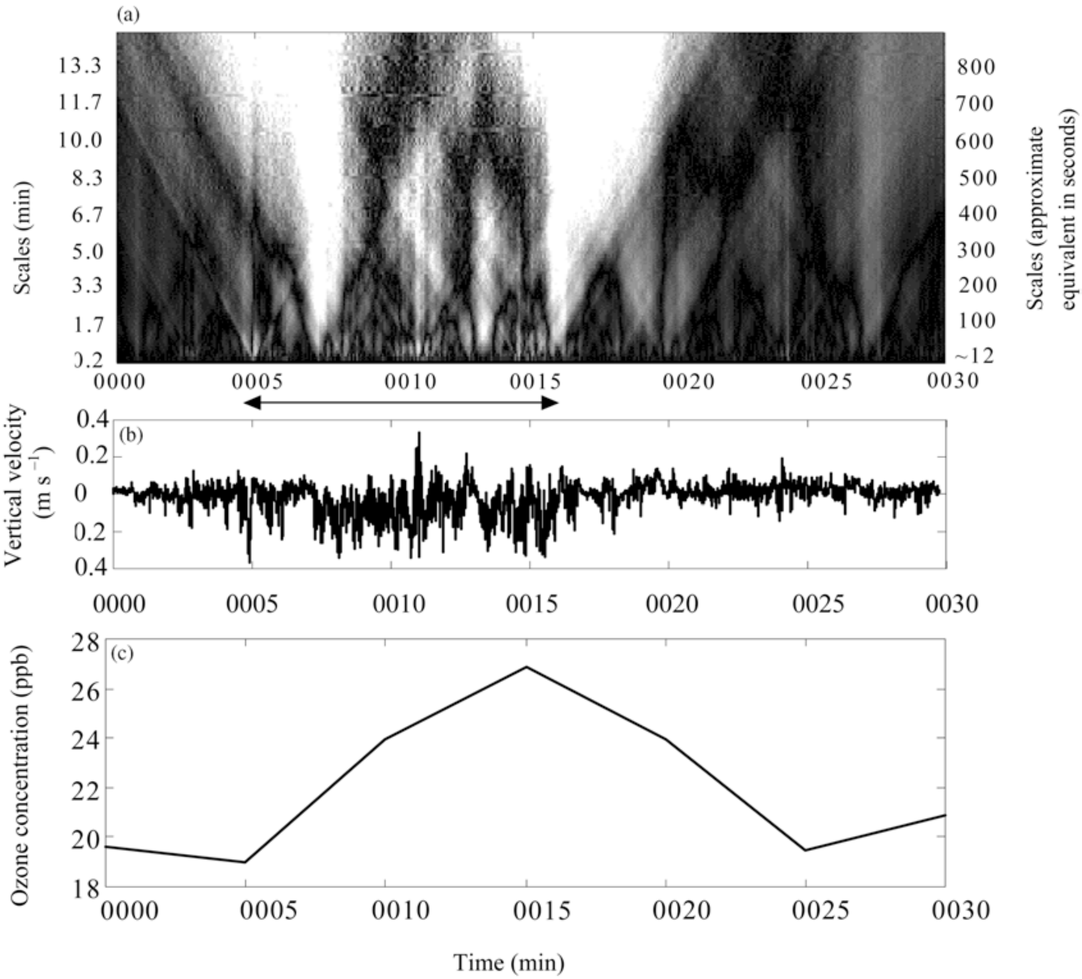

Figure 2 (a) Time series of vertical velocity, (b) scalogram of vertical velocity and (c) surface ozone concentrations at Aldergrove at 0000–0030 PDT on 1 September 1998

Although ozone maxima are the most extensively studied, localized nocturnal spikes in a range of different pollutants have been observed. These include volatile organic compounds (Prevot *et al.*, 2000), isoprene (Starn *et al.*, 1998), hydrogen peroxide (Das and Aneja, 1994), carbon monoxide (CO) (Moxley and Cape, 1997), nitrogen oxide (NO) and particulate matter smaller than 10 microns in diameter (PM_{10}) (McKendry, 2000). Results indicate that some of these spikes result from the building up of surface emissions in the very stable surface layers as well as vertical transport from reservoirs aloft. An example of such an event from the LFV is shown in Figure 4. After a warm summer day with elevated ozone concentrations, NO and PM_{10} concentrations (primarily from automobile emissions into the surface layer) are observed to peak before midnight as the very stable NBL develops and dispersion is significantly reduced. This buildup is abruptly

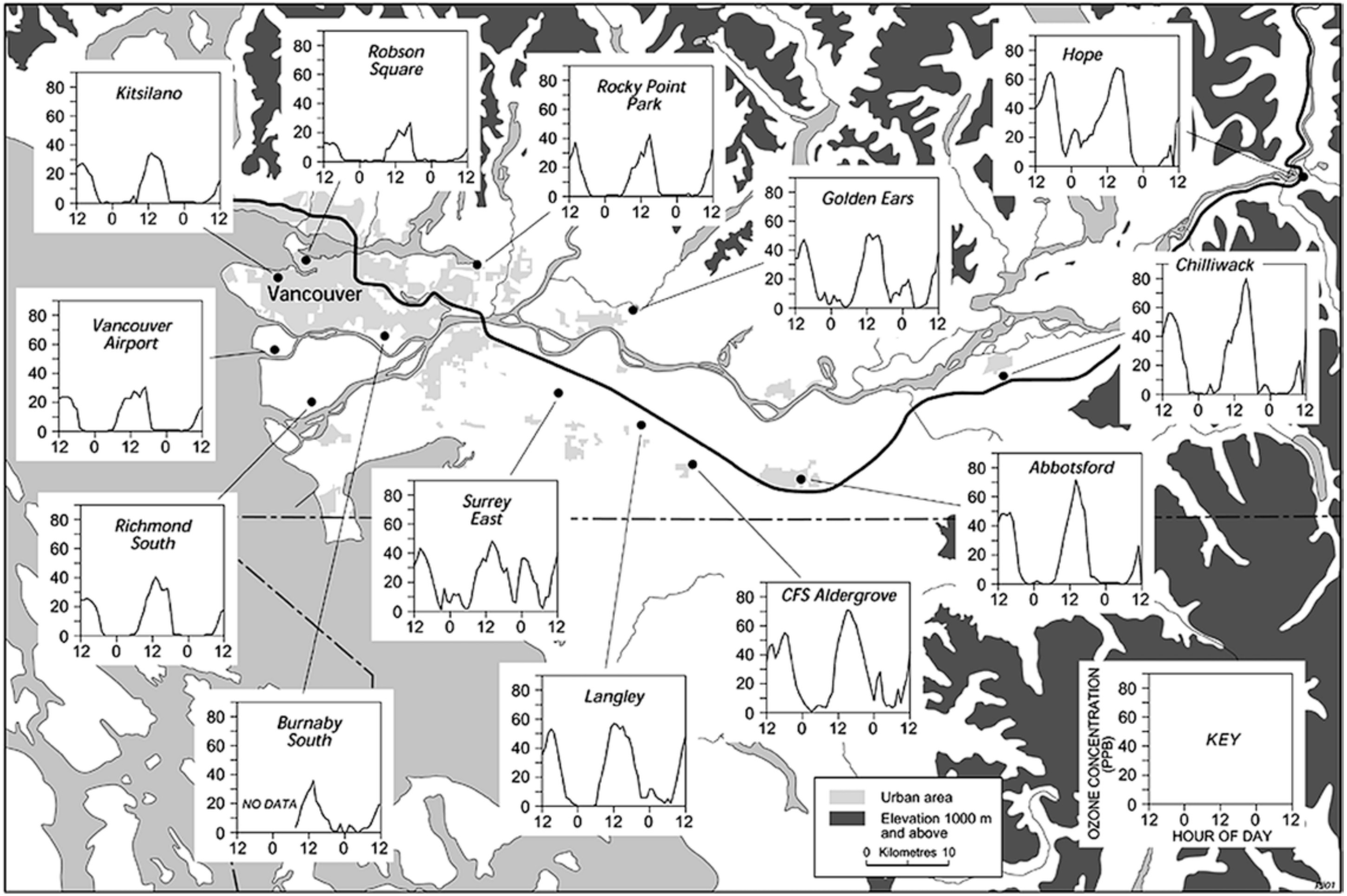

Figure 3 Map to show the variations in O_3 concentration recorded at 10 of the Greater Vancouver Regional District O_3 monitoring sites between 1200 PDT on 30 August and 1200 PDT on 1 September 1998
Source: reprinted from Salmond and McKendry (2002) with permission from Elsevier.

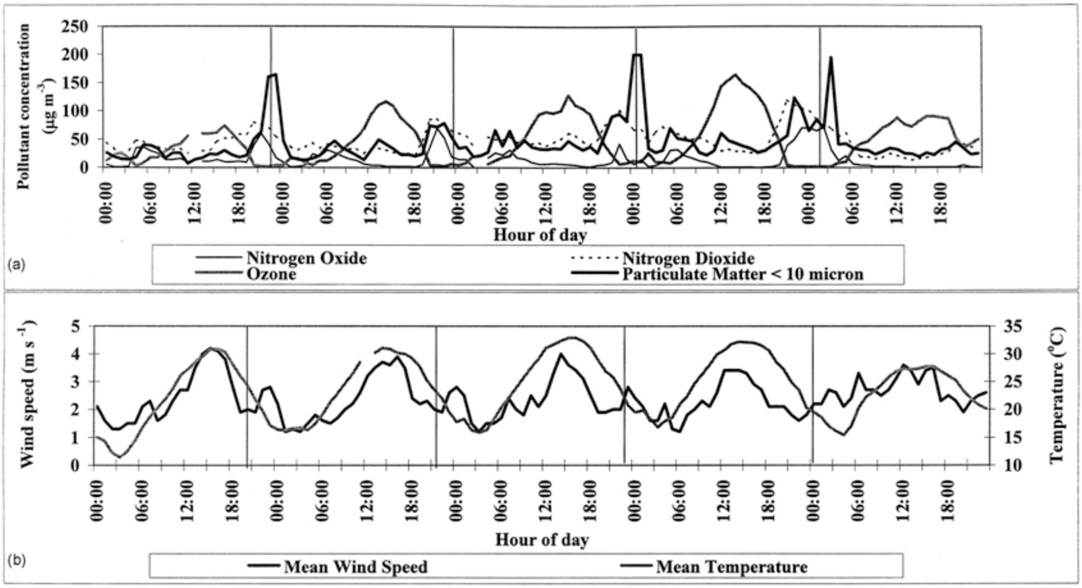

Figure 4 (a) Time series of PM10, ozone, nitrogen monoxide and nitrogen dioxide concentration for Abbotsford and (b) wind speed and temperature for nearby sites, between 27 June and 1 July 1995

curtailed with the onset of vertical mixing that mixes NO and PM_{10} upwards and brings ozone to the surface from the residual layer aloft creating a 'spike' in surface ozone concentrations (McKendry, 2000).

The occurrence and potential significance of such 'spikes' in nocturnal pollutant concentrations has been frequently overlooked in the literature. This is in part due to the localized, intermittent characteristics of the pollutant transport processes and in part due to the averaging techniques used to describe air quality. However, these marked increases in concentration may have environmental and health consequences for local populations. Elucidation of vertical mixing processes operating in the very stable NBL also has significant implications for pollutant concentrations in the residual layer.

Due to logistical issues, only a few examples exist of measurement programmes that have considered diurnal concentrations in the residual layer and most of these studies are from mountain areas where surface sites are located within the residual layer due to their elevation. For example, studies by Zaveri *et al.* (1995) in the Southern Appalachians, Vecchi and Valli (1999) in the Alps and Millan *et al.* (2000) along the Spanish coast of the western Mediterranean basin suggest that, while diurnal cycles of ozone concentration are pronounced at sites located at low altitudes in the valley or near sea level, little diurnal variation in concentration is observed at altitudes considered to be predominantly located within the RL. Thus pollutant concentrations (especially ozone levels) are frequently treated as invariant in time and space in the residual layer (Neu *et al.*, 1994).

However, an increasing number of studies suggest that this is an oversimplification, and that, particularly in areas of complex terrain, ozone concentrations can be highly variant in the RL and strongly affected by local patterns of advection (Banta *et al.*, 1997; Berkowitz *et al.*, 2000; Salmond and McKendry, 2002). For example, Berkowitz *et al.* (2000) note that relatively large increases in nocturnal surface ozone levels are preceded by the horizontal transport of aged plumes rich in ozone aloft. This three-dimensional complexity of pollutant transport and vertical mixing (shown in Figure 1, profile 3) is well illustrated in the following case study from the Lower Fraser Valley, British Columbia, Canada.

In Figure 5, the evolution of the vertical distribution of ozone, temperature and wind throughout the night at CFS Aldegrove (derived from tethered balloon flights), illustrates the local three-dimensional complexity of such events. The temporal sequence may be summarized as follows.

- 2000–2300 hours: development of a very SBL accompanied by decreasing near surface O_3 concentrations associated with surface deposition and chemical destruction. Winds are from the west and up-valley (+u component). High ozone (>80 ppb) concentrations persist above 400 m AGL.
- 2300–0300 hours: winds switch to down-valley (−u component) and associated shear instability promotes vertical mixing from RL aloft (seen as bursts of turbulence in Figure 2). O_3 concentrations of 80 ppb appear at 100–200 m AGL with lower concentrations from 250 to 350 m. Surface

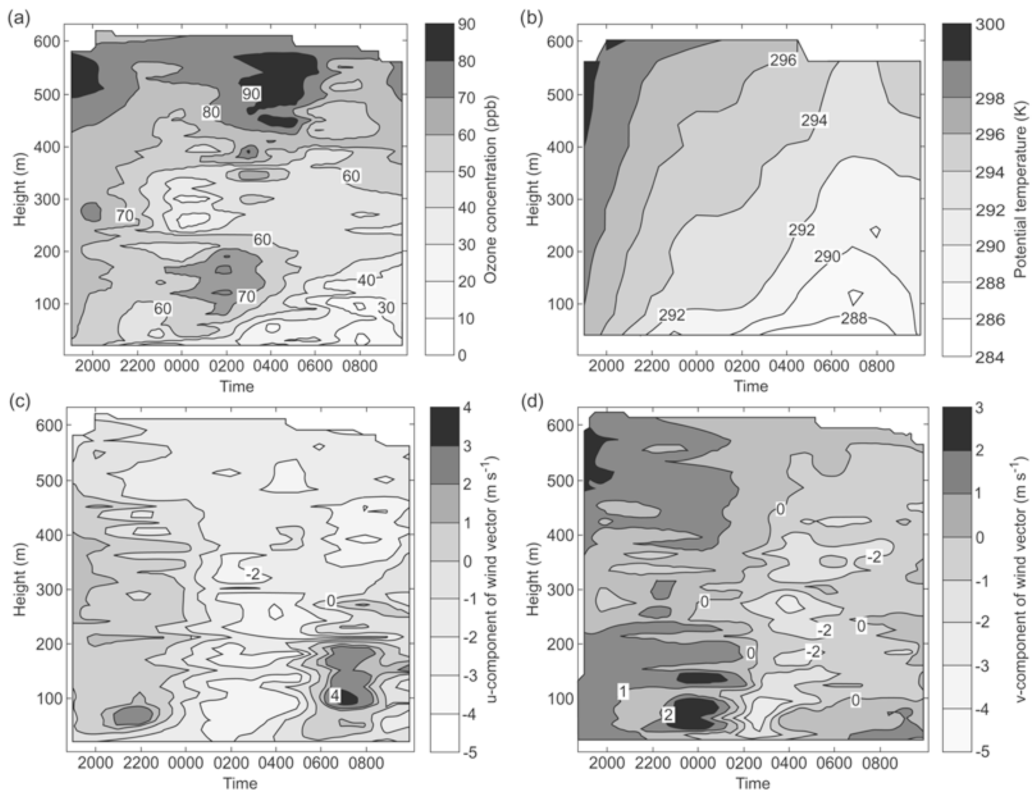

Figure 5 Contour plots of (a) ozone concentration, (b) potential temperature, (c) u-component of wind vector (+u represents up-valley winds and −u represents down-valley winds) and (d) v-component of wind vector (cross-valley winds) for 31 August to 1 September 1998
Source: adapted from Salmond and McKendry (2002).

cooling is inhibited (note near horizontal isentropes) as warm air from aloft is mixed downward.
- 0300–0700 hours: despite persistence of high ozone concentrations in RL the very SBL re-establishes in the nocturnal down-valley flow and low concentrations are observed at ground level. In the absence of shear instabilities associated with the reversal of the mesoscale wind regime, no further turbulent down-mixing events occur.

This example illustrates that within the residual layer pollutant concentrations may be highly variable with both time and height. Plumes of high ozone concentrations are intermittently advected from a variety of different directions. Large increases in ozone concentrations recorded at the surface were coincident with both ozone concentrations of more than 80 ppb in the residual layer and significant wind shear generated by the reversal of local thermotopographically generated winds. The timing of turbulence *vis-a-vis* advection of ozone aloft was found to be a critical factor determining the magnitude of ozone spikes at the surface.

The concentrations of pollutants in the RL are known to play an important role in determining the characteristics of diurnal cycles of surface photochemical pollutants (Hastie *et al.*, 1993; Zaveri *et al.*, 1995; Beyrich *et al.*, 1996a; Berkowitz *et al.*, 2000; Zhang and Rao, 2000). This is because at dawn, following the breakdown of the stable NBL and resulting entrainment of the residual layer, pollutants stored in the RL during the night become well mixed into the daytime convective boundary layer. For example, recent research suggests that more than half the daily surface concentrations of ozone can be accounted for by entrainment from pollutant reservoirs aloft (Millan *et al.*, 2000). Further, Banta *et al.* (1998) note that during periods of poor air quality in Nashville, Tennessee, nighttime advection of the urban plume is slow compared to other conditions, and urban pollutants are stored aloft to be mixed to the ground the following day rather than advected downwind into the surrounding rural areas. However, little is known about the impact of intermittent vertical mixing events in determining mean pollutant concentrations in the RL.

Clearly the processes determining pollutant transport and storage in the very stable NBL may have significant implications for the daily temporal autocorrelation of ozone concentrations during pollution episodes (Gusten *et al.*, 1997). Understanding the interaction between advection and pollutant storage in the residual layer with turbulent mixing processes is therefore critical to identifying the importance of mixing events on diurnal pollutant cycles.

V Discussion and conclusions

The strongly layered structure and weak, patchy and intermittent characteristics of turbulence in the very stable NBL present a particular challenge to meteorologists and air pollution modellers alike. However, in the last 20 years a combination of extensive (three-dimensional) field campaigns, the development of new instruments such as the scintillometer and introduction of new analytical tools such as wavelet analysis have started to provide real insights into the structure and characteristics of turbulence in the very stable NBL.

In this review, attention is also drawn to developments in our understanding of pollutant distribution and transport in the very stable NBL. Recent studies have shown that the bursts of turbulence characteristic of the very stable NBL may temporarily couple the stable surface and residual layers facilitating the exchange of pollutants between the two layers. Such mixing events not only contribute to ground-level pollutant exposures but also may affect regional pollutant mass budgets by depleting the 'reservoir' of pollutants stored in the nighttime residual layer.

The emergence of a literature reflecting various geographical settings suggests that such processes are commonplace and consequently may have important air pollution implications. However, the examples from the Lower Fraser Valley suggest that the analysis of such events is not straightforward, especially in regions of complex terrain where nocturnal near-surface flows tend to be intermittent and meandering in form. Furthermore, discontinuous layers of pollutants and chemical reactions between different pollutants in the atmosphere further hinder interpretation.

Although much has been learned about the nocturnal transport and dispersion of air pollution, considerable scope for further research remains. Of most urgent need are studies addressing the following.

- *Budgets*: most of the studies cited here have concentrated on observations at single sites. Regional studies are required to assess the role of the RL with its stored pollutants and vertical mixing processes on regional pollutant budgets. With wavelet analysis as an emerging tool, detailed studies exploiting turbulence measurements are now more practical.
- *Climatology:* to date, few studies have investigated the detailed climatology of nocturnal mixing events. The location, timing (synchronicity) and seasonality of such events across a variety of geographical settings remain an important priority.
- *Health Studies*: the impact of nocturnal spikes on indoor exposure, human health and the local environment is as yet undetermined.
- *Modelling:* time-dependent, three-dimensional models incorporating complex chemistry are essential to the development of a forecasting capability and form an important test bed for pollutant abatement strategies. The complex processes described herein imply an urgent need for high-resolution simulations that capture the details of vertical mixing, local flows and advection of pollutant layers.

Although there are significant challenges associated with the analysis of vertical mixing processes in the very stable nocturnal boundary layer the examples presented here clearly demonstrate that intermittent turbulence can have a significant impact on ground-level concentrations of pollutants in the very stable NBL. Further, this review demonstrates that, from the perspective of diurnal pollutant budgets, the processes operating in the NBL which determine the three-dimensional patterns of pollutant advection, storage, transport and deposition cannot be ignored.

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