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A review of measurement and modelling results of particle atmosphere–surface exchange

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ABSTRACT

Atmosphere–surface exchange represents one mechanism by which atmospheric particle mass and number size distributions are modified. Deposition velocities (v_d) exhibit a pronounced dependence on surface type, due in part to turbulence structure (as manifest in friction velocity), with minima of approximately 0.01 and 0.2 cm s^{-1} over grasslands and 0.1–1 cm s^{-1} over forests. However, as noted over 20 yr ago, observations over forests generally do not support the pronounced minimum of deposition velocity (v_d) for particle diameters of 0.1–2 μm as manifest in theoretical predictions. Closer agreement between models and observations is found over less-rough surfaces though those data also imply substantially higher surface collection efficiencies than were originally proposed and are manifest in current models. We review theorized dependencies for particle fluxes, describe and critique model approaches and innovations in experimental approaches, and synthesize common conclusions of experimental and modelling studies. We end by proposing a number of research avenues that should be pursued in to facilitate further insights and development of improved numerical models of atmospheric particles.

1. Introduction

The mass and number distribution of atmospheric aerosol particles are determined, in part, by atmosphere–surface exchange and in turn these properties influence the magnitude of direct and indirect climate effects (IPCC, 2001), visibility degradation (Malm, 2003) and detrimental health impacts (Pope and Dockery, 1999). A key process in atmosphere–surface exchange is dry deposition—atmosphere to surface exchange via contact. The relative importance of wet (and occult) versus dry deposition to particle removal is dependent on a plethora of atmospheric parameters (e.g. the precipitation climate), particle characteristics, and surface type, but dry deposition is a continuous process and likely contributes a significant fraction of particle removal in most environments (e.g. Foken et al., 1995; Takemura et al., 2000). Accordingly, inclusion of additional/improved parametrizations of processes affecting vertical transport and exchange of particles in air quality and climate modelling is recognized as a research need within international and national priorities (e.g. in the rubric of the North American Research Strategy for Tropospheric Ozone (NARSTO) (Hidy et al., 2000), and also the Intergovermental Panel on Climate Change (IPCC, Alley et al., 2007)). Improved understanding of the mechanisms of interaction between particles and vegetation has also returned to prominence with proposals to use trees as filtration mechanisms to reduce the exposure of urban populations to elevated concentrations of atmospheric particles (Donat and Ruck, 1999; Freer-Smith et al., 2005).

Several previous papers have articulated the state of knowledge regarding dry deposition (i.e. atmosphere to surface exchange by contact) during the last 20–30 yr. However, a decade has past since the publication of the review of particle fluxes over natural surfaces by Gallagher et al. (1997b), and there have been substantial advances in experimental techniques since Nicholson

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(1988a), Davidson and Wu (1990) and Sievering (1989) reviewed experimental methods for application to particle flux measurements. It is also over a decade since Dabberdt et al. (1993), Foken et al. (1995) and Businger (1986) described, more generally, issues that confront those involved in atmosphere–surface exchange studies. Further, it is over 5 yr since the overview of the status of knowledge on dry deposition by Wesley and Hicks (2000) appeared.

Here we present an update of these dry deposition reviews with a sole focus on particles and emphasis on experimental aspects of the vertical exchange over vegetated surfaces. For completeness we include some studies not conducted over vegetated surfaces in the literature synthesis presented in Table 1, but largely focus our discussion on vegetated surfaces to constrain this dialogue to surface types that are theorized to be dominated by deposition, as opposed to marine environments where the surface acts as a particle source in addition to a sink (e.g. Mårtensson et al., 2003) and urban environments where anthropogenic particle sources tend to dominate and emissions are typically observed (e.g. Dorsey et al., 2002). We document methodological and instrumentation advances and recent innovative application of theoretical and numerical tools, and describe the process-level insights derived from application of those techniques. We also briefly summarize operational options currently being utilized as part of monitoring networks. We conclude by articulating some of the remaining uncertainties in our understanding of the processes that dictate particle fluxes in the absence of precipitation and propose potentially fruitful research directions to be pursued.

2. Governing principles of dry deposition

Dry deposition describes transport of a property from the atmosphere to the surface by contact in the absence of precipitation. A key concept is that of deposition velocity $v_d(z)$ (i.e. the deposition velocity at a given height z) which is the flux of a constituent normalized by the concentration expressed as a partial density at that height z [shown as $C(z)$]:

$$v_d(z) = \frac{-F(z)}{C(z)}. \tag{1}$$

As discussed by Businger (1986), (1) implies that $F \to 0$ as the concentration at the surface $[C(0)] \to 0$. For $C(0) \neq 0$ one can also introduce the concept of a transfer velocity (v_t) (Chamberlain, 1966; Businger, 1986):

$$v_t(z_2, z_1) = \frac{-F}{C(z_2) - C(z_1)} \tag{2}$$

which can be employed for any height interval (i.e. z_1 can be taken as 0 or any other height).

The concept of particle v_d is also used extensively in modelling of dry deposition. In Table 1, where we summarize the results of many particle flux studies conducted during the past three decades, we employ a protocol that reports the terminology used in the original article from which data are drawn.

In practice the surface may act as a sink and/or source of both gases and particles. Hence fluxes may be bi-directional, or alternatively stated, the net exchange may have contributions from fluxes directed both to the surface (this flux is defined as negative) or from the surface.

For a rigorous treatment of material budgets in the atmospheric surface layer see Businger (1986), in brief, the conservation equation for scalars is:

$$\frac{\partial C}{\partial t} + \frac{\partial}{\partial x_i} u_i C = D \frac{\partial^2}{\partial x_i^2} C - \frac{\partial}{\partial z}\left(v_g C\right) + S. \tag{3}$$

Using Reynolds decomposition to decompose C and u into their average and fluctuating components ($C = \overline{C} + C'$ and $u_i = \bar{u}_i + u_i'$), the average of eq. (3) is:

$$\frac{\partial \overline{C}}{\partial t} + \frac{\partial}{\partial x_i} \overline{u_i}\,\overline{C} + \frac{\partial}{\partial x_i} \overline{u_i' C'} = D \frac{\partial^2}{\partial x_i^2} \overline{C} - \frac{\partial}{\partial z}\left(v_g \overline{C}\right) + \overline{S}. \tag{4}$$

In this equation the first term on the left-hand side is the local change in concentration (i.e. the storage change term), the second term represents advection by the mean flow, and the third term is the divergence of the turbulent flux. The terms on the right hand side describe transport by diffusion, and sedimentation, respectively. The final term in eq. (4) represents changes due to sources or sinks, which when considering the entire particle ensemble, derives solely from phase transfer assuming that biological sources and sinks are treated as boundary conditions (Raupach, 2001). Below we assume $\overline{w} = 0$, though this condition may not be realized in complex terrain, and as discussed in Section 4 buoyancy effects can cause violations of this assumption.

Under the assumptions of horizontal homogeneity ($\frac{\partial}{\partial x} = \frac{\partial}{\partial y} = 0$), steady state conditions ($\frac{\partial C}{\partial t} = 0$) and that there is no chemical source or sink of the scalar ($\overline{S} = 0$), eq. (4) becomes:

$$\frac{\partial}{\partial z} \overline{w'C'} = D \frac{\partial^2}{\partial z^2} \overline{C} - \frac{\partial}{\partial z}(v_g \overline{C}) \tag{5}$$

which can be integrated to give (Businger, 1986):

$$\overline{w'C'} = D \frac{\partial \overline{C}}{\partial z} - v_g \overline{C}. \tag{5a}$$

In practice, the storage and advective terms are proportional to measurement height (Fowler and Duyzer, 1989) and may not be zero and so use of eq. (4) may be required. Under conditions of low turbulence, the measured flux above the forest (i.e. the term on the left-hand side of eq. 5) may be very small, even when the other terms in the budget equation are non-negligible.

The influence of phase changes and particle dynamics on particle fluxes (and vertical flux divergence) has received only limited attention. In the case of a poly-dispersed particle ensemble, the continuity equation should be applied to a discretized form of the

Table 1. Synthesis of particle flux studies conducted over the past three decades. The abbreviations used for the instrumentation, chemical symbols and method are given below the table. Unless otherwise stated the values given for v_d are the mean, the number subsequent to ± is the standard error and numbers given in parentheses are the range

Reference	Method	Surface	Substance	D_p	v_d (cm s^{-1})	Instrumentation	Site	Data period	Comments
Allen et al., 1991	Gradient	Grass	S	0.1–2.0 μm	0.10 ± 0.03 σ = 0.18 Median = 0.08 (−0.33–0.57)	Filter sampler, IC	Colchester, England	06/88– 06/89	The authors noted 'no observed dependencies of deposition velocity on either atmospheric stability or friction velocity within the range of conditions encountered'
Buzorius et al., 1998	EC	Forest		>12 nm	−0.11 ± 0.01 (−1.5–1.0)	CPC	Hyytiälä, Finland	08/96	Suggested surface source may reflect particle formation within or close to canopy top.
Buzorius et al., 2000	EC	Forest		> 10 nm 10–20 nm	0.01–1.00 0.5–4.0	CPC, DMPS	Hyytiälä, Finland	03–05/97	Noted a tendency for v_d of 10–20 nm particles to increase with u_*
Buzorius et al., 2001	EC	Forest		> 10 nm	Flux [10^6 m^{-2} s^{-1}]: −1.05 ± 13.88	CPC, DMPS	Hyytiälä, Finland	04–05/98, 07–08/98, 03–04/99	
Davies and Nicholson, 1982	Gradient	Field	S		Median = 0.08 Unstable: 0.16 Neutral: 0.04 Stable: 0.025		England	79–80	Several cases with upward fluxes attributed to analytical uncertainties
Dollard and Unsworth, 1983	Gradient	Grass	Fog droplets	> 5 μm MMD ~ 20 μm	2.4–7.0[aa] Flux: 0.7–4.9 mg m^{-2} s^{-1}	Glass impactor	Leicestershire, England	01–04/81	$v_d \approx$ that for momentum Clear dependence of v_d on u_*
Donateo et al., 2006	EC	Grass, urban context		$PM_{2.5}$	Average over one week: −1–0.1[a] $PM_{2.5}$ mass flux: −0.5–0.5 μg m^{-3}	Optical (Mie pDR-1200)	Lecce, Italy	05/03, 01/04, 12/04	Strongly bi-directional fluxes. Noted measurements using the optical probe are strongly influence by humidity Emission due to anthropogenic activity
Doran and Droppo, 1983	Gradient	Grass	S		Mean: 0.2–0.3 Some cases of 0.3–0.4		Champaign, Illinois, USA	06/82	Several profiles exhibited inflection points
Dorsey, 2002	EC	Grass		11–100 nm 0.1–0.2 μm	0.0066–0.0275[bb] 0.087–0.113[bb]	CPC, ASASP, DMPS	Braunschweig, Germany	05–06/00	
Dorsey et al., 2002	EC	Urban		11 nm–3 μm	−2.0–7.5[c]	CPC	Edinburgh, Scotland	05 & 10/99, 10–11/00	
Droppo, 1979	Gradient	Arid vegetation	S		0.10–0.27	X-ray fluorescence of filter	Hanford, USA		
Duan et al., 1988	EC	Field/snow		0.15–0.30 μm 0.5–1.0 μm	0.034 ± 0.014 (−0.170–0.170) 0.021 ± 0.005 (−0.081–0.084)	ASASP CSASP	Rock Springs, Pennsylvania, USA	12/85	Under neutral and stable conditions normalized v_d/u_* was 0.006 for D_p ~0.15–0.30 μm and 0.0024 for D_p ~ 0.5–1.0 μm

Table 1. (cont'd).

Reference	Method	Surface	Substance	D_p	v_d (cm s^{-1})	Instrumentation	Site	Data period	Comments
Duyzer et al., 1994	Gradient	Forest	Sulfate Nitrate		−13.4–44.2 –0.5–31	Filter	Speulderbos forest, The Netherlands	92–94	More frequent upward fluxes for sulphate than nitrate Detection limit for sulphate v_d estimated to be 0.5–1.0 cm s^{-1}
Everett et al., 1979	Gradient	Grass	S		1.4 ± 0.4 1.2 ± 0.3 (stable) 1.7 ± 0.8 (unstable)	filter sampler, PIXE	Argonne, Illinois, USA	07/76	Report evidence for a strong dependence of v_d on stability, and that particulate S v_d is ∼ to that for momentum
Fowler et al., 1990	Gradient	Moorland	Cloud droplets	2–30 μm NMD ∼ 6–7 μm	2.1–3.9[cc]	FSSP	Great Dun Fell, England	03/87	Aerodynamic resistance contributes 60% of total resistance to transfer. Study used a weighing lysimeter combined with wind speed profile measurements from cup anemometers to evaluate u_* and z_0 and hence r_a
Freer-Smith et al., 2005	Leaf wash-off	Urban		PM_{10} $PM_{2.5}$ PM_1	0.57–5.35 0.81–11.04 25.43–36.24	Grimm, IC and AAS for composition to deposited particles	Brighton, UK	1998	Range of v_d to foliage shown is range across five tree species
Galbally et al., 1979	EC	Forest field	S S		0.2[d] 0.6[d]	FPSA	Berkshire, England	05–07/77	
Gallagher et al., 1988	Gradient	Moorland	Cloud droplets[e]	5–13 μm 13–25 μm 25–31 μm	0.5–5.8 5.1–8.9 4.9–6.6	FSSP	Great Dun Fell, England	06/87	For one specific sampling period in considering particles with D_p of 5–31 μm v_d was maximized for $D_p = 21$ μm. $v_d >$ momentum v_d for $D_p = 13$–24 μm, though authors note the difference observed for larger particles may be due to large statistical uncertainties.
Gallagher et al., 1988	EC	Coniferous forest	Cloud droplets	2–32 μm 3–50 μm	0.3–2.3[a] 0.2–3.3[a]	FSSP PVM	Dunslair Heights, Scotland	11/88	v_d increased with D_p. v_d for particles exceeded that for momentum for $D_p = 6$ μm Bi-directional fluxes observed.
Gallagher et al., 1992b	EC	Snow covered forest	Cloud droplets	3–13 μm 13−25 μm 25–31 μm	−4.4–5.2[a] 5.2–16.8[a] 10.9–50.9[a]	FSSP	Dunslair Heights, Scotland	03/90	Snowfall caused a two-fold decrease in the flux of cloud water to the canopy.

Gallagher et al., 1992a	EC, gradient	Forest, moorland	Cloud droplets[e]	3–31 μm	$\geq$ 80% of momentum derived v_d	FSSP, PVM	Dunslair Heights, Scotland Great Dun Fell, England	03/90	Over forest canopies v_d for $D_p >$ 17 μm exceeds v_d for momentum.
Gallagher et al., 1997a	EC	Forest		0.1–3.0 μm	0.01–10.00	ASASP	Speulder, the Netherlands	06–07/93	Bi-directionality of fluxes more pronounced for D_p: 0.18–0.24 μm than for D_p: 0.10–0.18 μm. v_d for D_p: 0.1–0.18, 0.18–0.24, 0.24–0.3, 0.3–0.5 μm all exhibit a strong dependence on u_*, though the precise nature of the relationship is a function of measurement height. Proposed the following relationship: $v_d = 0.0135 D p u^* (1 + (\frac{-300}{L})^{2/3})$ for $-0.04 < L^{-1} < 0, 0.1 \leq D_p \leq 0.5$ μm
Gallagher et al., 2002	EC	Heathland grass cropland Coniferous forest		0.1–0.2 μm 0.1–0.2 μm 0.1–0.2 μm 0.1–0.2 μm	0.071 ± 0.007[bb] 0.087–0.113 $\pm$ 0.01[bb] 0.138 ± 0.041[bb] 0.208 ± 0.052[bb]		Auchencorth Moss, Scotland Braunschweig, Germany Speulder, Netherlands	Variable	Based on synthesis of multiple data sets proposed $v_d = 0.581 \ln(z_0) + 1.86$ where z_0 is in m and v_d is in mm.
Gaman et al., 2004	REA	Coniferous forest		50 nm	0.43 ± 0.06	DMPS	Hyytiälä, Finland	09–10/01	Noted bi-directionality of flux. Some evidence for stability dependence of the flux: Mean v_d by stability class: Neutral: 0.48 $\pm$ 0.07 Stable: 0.24$\pm$ 0.02 Unstable: 0.15$\pm$ 0.8 (very few observations) For 50 nm particles: $v_d = 0.012 u_*$
Garland, 1981	Mixed	Grass		$\sim$0.04 μm $\sim$0.05–0.2μm $\sim$1.8	0.30 0.06 ± 0.03 0.05–0.12	CPC & radioactive tagging	England	1981	Synthesized three studies
Garland, 1981	Gradient	Grass		0.05–1.00 μm	< 0.1	CPC	England		Also observed some upward fluxes

Table 1. (Cont'd).

Reference	Method	Surface	Substance	D_p	v_d (cm s^{-1})	Instrumentation	Site	Data period	Comments
Gronlund et al., 2002	EC	Snow		> 10 nm	0.08–1.89	CPC,	Dronning	01/00	
		rock/snow		> 10 nm	0.2–2.4	DMPS	Maud Land, Antarctica		
Grönholm et al., 2007	REA	Pine forest		15 nm	Median v_d 1.49	DMPS	Hyytiälä,	01/04–01/05	Same system as Gaman et al.
				20 nm	1.18		Finland		
				25 nm	1.23				
				40 nm	0.94				
				50 nm	0.74				
				60 nm	0.62				
				70 nm	0.66				
				80 nm	0.99				
				100 nm	0.69–0.78				
				150 nm	0.25				
Held et al., 2003	DEA/ IDES	Forest	NO_3^- flux is predominantly upward.			TOF–MS	Waldstein, Germany	05/02	
Hicks and Wesely, 1979	Gradient	Pine forest	S				North Carolina USA	07/77	Surface resistance to 'fine particles' is proportional to u_*^p, where p = 1.0 ± 0.3.
Hicks and Williams, 1979	EC	Open lake			< 1		Great Lakes, USA		Particle instrumentation not described
Hicks et al., 1982	EC	Pine forest	S	0.03–0.1 μm	0.5–2.0 (day) (typical v_d = 0.7) < 0 (night) > 0.1 down in afternoon otherwise upwards	FPSA	Alamance County, North Carolina, USA	07/77	
				0.07 μm					
Hicks et al., 1983	EC	Grass	S		Diurnal average: 0.2 (daytime mean = 0.7) Range: 0.00–0.72	FPSA	Springfield, Ohio, USA	09/79	Strong diurnal variability
Hicks et al., 1986	EC	Grass	S		0.4 (−0.01–0.72) 0.8[d] (0.11–1.85)[d]	FPSA	South Charleston, Ohio, USA	09/79	

Hicks et al., 1989	EC	Forest	S other particles	0.05–0.10 μm 0.5–0.7 μm 0.7–1.2 μm	0.6 (0.0–1.0) 0.00–0.75 −1.2–0.9 −0.3–0.5	FPSA electrometer OPC OPC	Oak Ridge, Tennessee, USA	05/83	Several cases of upward fluxes particularly for $D_p \approx 0.7$ μm. Strong diurnal variability: Daytime average v_d for 50–100 nm particles ≈ 0.8 cm s^{-1}, night-time average $v_d \approx 0.1$ cm s^{-1}.
Hofken and Gravenhorst, 1982	Mass balance	Forest		MMD: 0.26 0.63 0.70 0.78 1.1 1.5 2.2 2.4	 1.8 1.1 1.0 0.9 0.7 1.0 1.0 1.3	Liquid washoff	Solling forest	1980	
Horvath, 2003	Mass balance	Forest	$(NH_4)_2SO_4$	$PM_{2.5}$	0.84 ± 0.25		Matra-Mountains, Hungary	Summer 2001	
Hummelshøj, 1994	Gradient	Grass		0.065–0.15 μm 0.15–0.4 μm 0.4–0.9 μm	$\bar{D}_p = 0.099$, $\bar{v}_d = 0.16$ $\bar{D}_p = 0.24$, $\bar{v}_d = 0.20$ $\bar{D}_p = 0.6$, $\bar{v}_d = 0.28$	OPC	Roskilde, Denmark	01/92	Also demonstrated the influence of stability (as characterised by the Richardson number) of fluxes
Katen and Hubbe, 1983	EC	Grass		0.15–2.50 μm	Negative and small	OPC	Champaign, Illinois, USA		
Katen and Hubbe, 1985	EC	Grass		0.15–0.21 μm 0.21–0.30 μm 0.30–1.50 μm 1.50–2.50 μm	−0.12–0.03 −0.18–0.18 −0.32–0.13 −0.52–0.20	OPC	Champaign, Illinois, USA	06/82	Strong evidence of bi-directionality in fluxes of all size ranges Variation of C'w' cospectra from those derived from u'w', T'w'
Kowalski et al., 1997	EC	Forest	Cloud droplets	2–32 μm	Flux [mg m^{-2} s^{-1}]: −7 (−35–25)	FSSP, PVM	Cheeka Peak, Washington, USA	07–09/94	
Kowalski and Vong, 1999	EC	Forest	Cloud droplets	2–32 μm	Flux [mg m^{-2} s^{-1}]: −19 [−50–(−5)]	FSSP, PVM	Cheeka Peak, Washington, USA	07–09/94	
Lamaud et al., 1994b	EC	Desert		0.2–2.0 μm	Mean < 0.1 0.05–0.65[bb]	Electrometer	Sahel, Niger	11/89	Proposed a linear increase of v_d with u_* Noted a strong dependence of $v_d u_*$ on 1/L

Table 1. (Cont'd).

Reference	Method	Surface	Substance	D_p	v_d (cm s^{-1})	Instrumentation	Site	Data period	Comments
Lamaud et al., 1994a	EC	Pine forest		0.05–1.0 μm	0.05–0.75	CCN electrometer	Pierroton, France	06/92	O_3 and aerosol v_d exhibit similar variability and magnitude (within $\pm$10% of each other). In near-neutral and stable conditions $v_d \approx 0.004u_*$
Lamaux et al., 1994	EC	Pine forest		0.05–1.0 μm	0.05–0.60	electrometer	Pierroton, France	06/92	
Longley et al., 2004a	EC	Urban		0.1–0.2 μm 0.2–0.5 μm 0.5–2.0 μm	1.3–3.0[c] 0.1–1.5[c] 0.3–0.8[c]	ASASP	Manchester, England	10/01	Variation in mean emission velocity (v_e) at street-level with D_p: D_p: 0.1–0.2 μm, $v_e \sim -1.5$–2.2 D_p: 0.5–1.0 μm, $v_e \sim -0.4$ Diurnal variability of v_e peaked in middle of the day and are most pronounced for 0.1–0.2μm
Longley et al., 2004b	EC	Urban		0.1–3.0 μm	Flux [# cm^{-2} s^{-1}]: 100–1000 (emission flux)	ASASP	Manchester, England	10/01	
Lorenz and Murphy Jr., 1989	gradient	Coniferous forest		0.5–1.0 μm 1.0–2.0 μm 2.0–5.0 μm	0.34 $\pm$ 0.35 (0.02–1.31) 0.78 $\pm$ 0.64 (0.03–2.71) 0.92 $\pm$ 0.76 (0.00–2.51)	OPC		9 months	v_d of 0.5–1.0 μm demonstrated to vary by a factor of 5 over the mean wind speed range of 2 to 4 m s^{-1}. r^2 of mean v_d by D_p: D_p = 0.5–1.0 μm v. U = 0.34 D_p = 1.0–2.0 μm v. U = 0.20 D_p = 2.0–5.0 μm v. U = 0.63
Meyers et al., 2006	REA	Agricultural field	SO_4^{2-}		2−3 −0.371 − 0.014 μg m^{-2} s^{-1}	Filter pack	Maryland, USA	06/01	Higher implied v_d-SO_4^{2-} than in previous studies over low roughness surfaces
Nemitz et al., 2000b	Gradient	Agricultural field	NH_4^+ NH_4^+ NO_3^- Cl^- SO_4^{2-}	MMD (μm): 0.42 0.68 1.61 0.34	0.17 $\pm$ 2.30 0.08 $\pm$ 1.98 1.01 $\pm$ 2.67 0.65 $\pm$ 3.87 0.49 $\pm$ 1.89	SJAC filter sampler filter sampler filter sampler filter sampler	North Berwick, Scotland	06/95	Noted importance of chemically induced flux divergence.
Nemitz et al., 2002b	EC	Moorland		> 11 nm 0.1–3.0 μm	−0.35–0.65 0.03–1.00	CPC ASASP	Auchencorth Moss, Scotland	05–10/99	Observed bi-directional fluxes and that mean v_d increased from 0.2 to 0.6 mm s^{-1} with u_* increasing from 0.15 to 0.55 ms^{-1}. For one case: v_d (mm s^{-1}) by D_p with u_* (ms^{-1}) 0.12–0.13: $v_d = 0.47u_*$ 0.15–0.16: $v_d = 1.07u_*$ 0.22–0.24: $v_d = 1.73u_*$ 0.4–0.45: $v_d = 4.55u_*$

Nemitz et al., 2004	EC gradient	Heathland heathland	NH_4^+	0.1–0.5 μm	0.1–0.9 (day) < 0.05 (night) 0.08–0.27 (night – day)	ASASP SJAC, filter sampler	Elspeet, the Netherlands	05–06/96	Bi-directionality of small (0.1–0.12 μm) particle flux. $\frac{v_d}{u*} = \left\{a_1(1+(-a_2L^{-1})^{2/3})\right\}, \quad L \leq 0$ $\frac{v_d}{u*} = a1, \quad L > 0$ $a_1 = 0.001$, $a_2[\text{m}] = 960D_p[\mu\text{m}]\text{-}88$
Neumann and den Hartog, 1985	EC	Grass	S	0.1–0.5 μm	< 0.05 0.21–0.33[d]	ASASP FPSA	Champaign, Illinois, USA	06/82	Average of data collected pertaining to sulphur fluxes: $v_d \approx 0.011u_*$
Nicholson and Davies, 1987	Gradient	Grass	S		0.07 ± 0.20 (−0.53–0.57) Stable 0.04 Unstable 0.21	Filter sampler, X-ray fluorescence	Norwich, England	06/79– 06/80	Only 78 profiles passed QA criteria. Upward fluxes frequently observed. Using data presented in the manuscript; Abs(v_d[cm s^{-1}]) = 0.0074u_*[ms^{-1}], $r^2 = 0.62$
Nilsson and Rannik, 2001	EC	Sea/ice		> 10 nm	0.19 (sea) 0.03 (ice) 0.029–0.091 (leads)	CPC, OPC, DMPS	Barents Sea, Arctic Ocean	07–08/96	
Nilsson et al., 2001	EC	Sea/ice		> 10 nm	Flux [m^{-2} s^{-1}]: 0.05*10^6 –20*10^6	CPC, OPC, DMPS	Barents Sea, Arctic Ocean	07–08/96	
Ould-Dada, 2002	Radioactivity	Model spruce canopy		MMD: 0.82 μm	Total canopy = 0.5 Soil = 0.015 Total = 0.515	Radioactivity	Wind tunnel		Demonstrated higher v_d for stems than needles.
Petelski, 2003	Gradient	Sea		1–20 μm	Flux [m^{-2} s^{-1}]: 10^3–10^7	CSASP	the Baltic Sea the Arctic Ocean	07–08/00, 07–08/01 10/00, 05/01, 10/01	
Pryor et al., 2007a	EC	Deciduous forest		> 10 nm	Flux [m^{-2} s^{-1}]: −7*10^7–5*10^7	CPC	Sorø, Denmark	05–06/04	Noted a significant fraction of particle flux estimates are not statistically different from zero according to error statistics from Wyngaard (1973)
Pryor, 2006	EC	Deciduous Forest		20–30 nm 60–70 nm	0.45 0.15	CPC, NMD independently from SMPS	Sorø, Denmark	05–06/04	For NMD = 50–60 nm Mean $v_d = 0.008u_*$ Median $v_d = 0.006u_*$ Where v_d and u_* are in ms^{-1}.
Pryor et al., 2007b	EC	Pine forest		GNMD (nm) 25 35 45 55 65 75 85	Mean v_d 0.43 0.37 0.31 0.31 0.27 0.26 0.24	CPC and DMPS	Hyytiälä, Finland	01/04–12/04	

Table 1. (Cont'd).

Reference	Method	Surface	Substance	D_p	v_d (cm s^{-1})	Instrumentation	Site	Data period	Comments
Rannik et al., 2000	EC	Forest		> 10 nm	0.01–1.50 (Modal diameter: 15–35 nm)	CPC, DMPS	Hyytiälä, Finland	09/00	Inference of decreasing v_d with particle size for modal diameters of 15–35 nm
Rannik et al., 2003	EC	Forest		10–500 nm	Flux [10^6 m^{-2} s^{-1}]: Median: −1.33 (5^{th} to 95^{th} percentile: −19.9–8.6)	CPC, DMPS	Hyytiälä, Finland	08–10/00, 04–10/01	$^1/_4$ of all observations fail QA criteria due to non-stationarities
Ruijgrok et al., 1997	ED	Coniferous forest		MMD 7–20 μm		FSSP	Speulderbos forest, The Netherlands	12/92–02/93	v_d of fog droplets = $0.195u_*^2$ (r = 0.83)
Schery et al., 1998	REA	Grass/ desert	Radon progeny	1.0–2.7 nm	7.3 ± 2.5 (5−35)	filter sampler, scintillation detector	Albuquerque, New Mexico, USA	05–11/96	Detection limit for v_d of ∼ 2 cm s^{-1}
Sievering, 1981	Gradient	Lake		0.1–2.0 μm	0.02–2.34 $0.11 < D_p < 0.2$ μm, $v_d = 0.4 \pm 0.1$ $0.3 < D_p < 0.6$ μm, $v_d = 0.2 \pm 0.1$ $0.6 < D_p < 1.5 \mu$m, $v_d = 0.7 \pm 0.3$	ASASP	Lake Michigan, Illinois, USA	05–09/77, 05–12/78	On average v_d for particles ∼ half that for momentum
Sievering, 1982	Gradient	Field		0.15–0.30 μm	0.38 ± 0.29 (0.1–1.9)	ASASP	Boulder, Colorado, USA	09–10/79	$z_0 < 0.1$ cm s^{-1}. Possible confounding influence of re-suspension leading to reduced gradients
Sievering et al., 1982	EC	Coastal/sea		0.2–0.5 μm	0.9 ± 1.1[c] (0.1–5.5)[c]	ASASP	Fowey Rocks, Florida, USA	02–03/81	Noted importance of particle dynamics in dictating measured fluxes
Sievering, 1983	EC	Field		0.15–2.50 μm	Overall mean: 0.05 0.37 ± 0.04 (1 case, stable) 1.19 ± 0.18 (1 case, unstable)	OPC	Boulder, Colorado, USA	06/82	Significant number of upward fluxes noted. Dominance of low-frequency fluctuations on total flux. During a single case study day correlation of v_d with $u_* = 0.57$. Strong influence of stability.

Sievering, 1986	Gradient	Field	S Al Ca Fe	0.1–5.0 μm	Mean +/−70% confidence interval 1.0 ± 0.5 (night) 2.9 ± 1.1 (day) 3.1 ± 2.8 3.5 ± 2.2 4.6 ± 3.0 5.3 ± 2.1 3.3 ± 1.9 4.7 ± 2.4	filter sampler, PIXE	Boulder, Colorado, USA	06/82	$6 < U < 10$ ms^{-1}
Sievering, 1987	EC	Field		0.09–2.50 μm	For $D_p = 0.2\mu$m Unstable: v_d: 1.2 ± 0.2 ms^{-1} Stable: v_d: 0.37 ± 0.04 ms^{-1}	ASASP	Boulder, Colorado, USA	05–08/82	$6 < U < 10$ ms^{-1}
Speer et al., 1985	EA	Grass	sulphur	Problems in concentration measurements.		filter sampler, IC	Champaign, Illinois, USA	06/82	
Tasdemir and Kural, 2005	SS	Urban	Ca, Ca, Pb, Ni		Pb: 2.3 ± 1.7 Ni: 11.1 ± 6.4	Mylar strips. Inductively coupled plasma atomic emission spectroscopy	Bursa, Turkey	10/02–06/03	
Vermeulen et al., 1997	EC	Forest	Fog Droplets	3–50 μm	−1–11[a]	PVM, FSSP	Speulder, the Netherlands	12/92, 02/93	v_d of fog droplets $= 0.195{u_*}^2$ (r = 0.83) v_d of fog droplets = $0.5v_d$(momentum)-0.0028
Vong and Kowalski, 1995	EC	Coniferous forest	Cloud Droplets	2–47 μm	0.0–2.6[a]	FSSP, PVM	Cheeka Peak, Washington, USA	04–05/93	Occasions with upward fluxes noted for $D_p < 8$ μm possibly due to splash droplets formed from impaction of large raindrops
Vong et al., 2004	EC	Grass		0.31–2.30 μm 0.52 μm	0.3 ± 0.07	OPC	Shedd, Oregon, USA	05–06/00	Pronounced influence of stability: Mean v_d for D_p of 0.34, 0.52 and 0.84 μm: Unstable = 0.65, 0.5, 0.75 cm s^{-1} Neutral = 0.60, 0.35,0.65 cm s^{-1} Stable = 0.22, 0.18, 0.22 cm s^{-1}
Wesley et al., 1977	EC	Grass		0.05–0.10 μm	0.1–1.2	Electrometer	Wood River, Illinois, USA	02–03/76	Light winds
Wesely and Hicks, 1979	EC	Snow grass		0.05–1.0 μm 0.05–1.0 μm	< 0.2 0.02–1.00		USA		

Table 1. (Cont'd).

Reference	Method	Surface	Substance	D_p	v_d (cm s^{-1})	Instrumentation	Site	Data period	Comments
Wesely and Williams, 1980	EC	Lake		0.03–0.10 μm 0.3–1.0 μm	flux upwards flux downwards		Lake Michigan, Illinois, USA		
Wesely et al., 1982	EC	Coastal/ sea		0.3–1.0 μm	Upward fluxes 4–28[c]	electrometer	Fowey Rocks, Florida, USA	03/81	Upward fluxes linearly dependent on u_*
Wesely et al., 1983a	EC	Forest and Grass	S		Synthesis of particulate sulphur in unstable conditions and the relationship to u_*; u_*: v_d: 0.25: 0.18 ± 0.02 0.3: 0.18 ± 0.02 0.14: 0.26 ± 0.11 0.18: 0.53 ± 0.18 0.3: 0.41 ± 0.05 0.22: 0.1 ± 0.07 0.57: 0.76 ± 0.08 0.44: −0.05±0.24	FPSA	Eastern North America		Evidence for upward fluxes in stable conditions, and for a strong influence of stability on flux magnitudes.
Wesely et al., 1983b	EC	Forest/ snow	S	0.05–0.10 μm 0.01–2.00 μm	0.8 (−0.41–0.90) −0.08 ± 0.18 (−1.28–2.48)	Electrometer FPSA	Durham, North Carolina, USA	01–02/81	
Wesley et al., 1985	EC	Grass	S	0.1–2.0 μm	0.22 ± 0.06[bb]	FPSA	Champaign, Illinois, USA	09/81, 06/82	Proposed dependence on u_* for $D_p = 0.1\text{-}1.0$ μm: $v_d = 0.002u_*$, $L \geq 0$ $v_d = 0.002u * [1 + (\frac{-300}{L})^{2/3}]$, $L < 0$
Wyers and Duyzer, 1997	Gradient	Coniferous forest	SO_4^{2-} NO_3^- NH_4HSO_4		0.7 ± 0.3 1.2 ± 0.4 1.1	Filter sampler, IC, denuder	Speulder, the Netherlands	04–12/93	Median v_d of NH_4HSO_4 = $0.0444u_*^{1.47}$ with $r^2 = 0.988$

Key

[aa]bulk deposition velocity $= v_t + v_g$

[a]turbulent deposition velocity v_t

[bb]surface v_d, v_d with influence of r_a removed

[b]for the nucleation mode particles (10–20 nm)

[cc]sedimentation velocity (v_g)

[c]emission velocity v_e ($= F_c/C$)

[d]both sulphur gases and sulphur particles

[e]Concentrations of SO_4^{2-}, NO_3^- and Cl^- were analysed from deposited water using ion chromatography.

Abbreviations used for flux approach:

EA = Eddy Accumulation

EC = Eddy Covariance

Gradient = Gradient sampling

REA = Relaxed Eddy Accumulation

DEA = Disjunct Eddy Accumulation

IDES = Irregular Disjunct Eddy Sampling

SS = Surrogate Surface

Chemical abbreviations

$(NH_4)_2SO_4$ = ammonium sulphate

NH_4HSO_4 = ammonium bi-sulphate

S = sulphur

NH_4^+ = ammonium

NO_3^- = nitrate

Cl^- = chloride

SO_4^{2-} = sulphate

Al = aluminium

Ca = calcium

Fe = iron

Abbreviations of instrumentation:

ASASP = Active Scattering Aerosol Spectrometer Probe

CPC = Condensation Particle Counter

CSASP = Classical Scattering Aerosol Spectrometer Probe

DMPS = Differential Mobility Particle Sizer

FPSA = Flame Photometric Sulphur Analyzer

FSSP = Forward Scattering Spectrometer Probe

IC = Ion chromatography

OPC = Optical Particle Counter

PIXE = Proton Induced X-ray Emission

PVM = Particulate Volume Monitor

SJAC = Steam Jet Aerosol Collector

TOFMS = Time-Of-Flight Mass Spectrometer $\bar{F}\ \Im_x$.

size distribution (i.e. specific to D_p), and it can readily be seen in that case for a given D_p, $S \neq 0$ would derive from changes in the particle ensemble due to particle dynamics (nucleation, coagulation, condensation/evaporation). The degree to which S deviates from 0 (and the magnitude of the vertical flux divergence due to phase transitions) is a function of the chemical climate (Nemitz and Sutton, 2004; Nemitz et al., 2004) and the particle ensemble (Pryor and Binkowski, 2004).

3. Numerical and theoretical modelling

In many operational air quality models dry deposition of atmospheric particles is quantified as the product of the modelled concentration and a value of v_d that is often modelled using the concept of the resistance analogue which is most commonly presented in the following form (Zufall and Davidson, 1998):

$$v_d(z) = \frac{1}{r_a(z) + r_b + r_c} + v_g. \tag{6}$$

In this approach, dry deposition is conceptualized as three resistances in series representing:

(1) The aerodynamic resistance to transfer $r_a = f(\frac{U}{u_*^2}, L)$.
(2) The resistance to transfer across the quasi-laminar surface layer $r_b = f(\frac{1}{u_*}, D)$.
(3) The resistance to surface uptake $r_c = f$(surface structure, presence of water or films).

in parallel to a second pathway—gravitational settling.

Particle exchange, when conceptualized using the resistance approach, is viewed as occurring as a sequence where the particle is first vertically transported towards the surface via turbulent diffusion and sedimentation and then across a pseudo-laminar sublayer largely by Brownian diffusion and finally by interaction with the surface. For larger particles, interception and impaction may circumvent r_b and as $D_p \to \infty$, v_g increasingly dominates the total flux. Davidson and Wu (1990) present a useful summary of some properties that influence the atmosphere–surface flux and proposed formulations for r_a, r_b and r_c.

Using the resistance analogy approach (eq. 6), v_d should be calculated by particle size (i.e. the observed or modelled particle size distribution should be discretized) since v_g and r_b are functions of D_p. Further the diffusivity of a particle of given size should be computed from the slip-flow corrected Stokes–Einstein relation:

$$D(D_p) = \frac{kT_a C_c}{6\pi \nu \rho \frac{D_p}{2}} \tag{7}$$

$$C_c = 1 + \frac{\lambda}{D_p}\left[2.514 + 0.8\exp\left(-0.55\frac{D_p}{\lambda}\right)\right] \tag{8}$$

(Seinfeld and Pandis, 1998).

Several authors (Kramm et al., 1992; Venkatram and Pleim, 1999) have noted that the electrical analogy presented in eq. (6) is imperfect in the context of particle dry deposition modelling because it is inconsistent with mass conservation. Sehmel (1973) derived an alternative formulation for monodispersed particles which was extended to a poly-dispersed ensemble by Kramm et al. (1992). Using similar arguments, for particles of a given size, Venkatram and Pleim (1999) proposed the following:

$$v_d(z) = \frac{v_g}{1 - e^{-r(z)v_g}} \tag{9}$$

$$r(z) = \int_0^z \frac{dz}{K_p(z)}, \tag{10}$$

where $r(z)$ and v_g are computed as a function of D_p, and v_g is considered to be height invariant.

The most commonly used process-level model for computing size-resolved particle v_d is that proposed by Slinn (1982) and it is formulated as shown below. Note that in this form it is implicit that the particle size distribution has been discretized and that the formula will be applied for multiple size classes (i.e. D_p):

$$v_d = v_g + C_D U_r \left[1 + \frac{U_h}{U_r}\left(\frac{1-\varepsilon}{\varepsilon + \sqrt{\varepsilon}\tanh\gamma\sqrt{\varepsilon}}\right)\right]^{-1} \tag{11}$$

$$C_D = \left(\frac{u_*^2}{U_r^2}\right) \tag{12}$$

$$\frac{U_h}{U_r} = \frac{u_*}{\kappa U_r}\ln\frac{1}{z_0} \tag{13}$$

while this model has been extensively used, it is based upon a number of assumptions which may frequently be violated in practice. Among these assumptions are the following:

(1) The model applies an eddy diffusivity (K) to describe vertical transport in the canopy, which implicitly assumes local transport down local concentration gradients. Further, in order to obtain an analytical solution, Slinn (1982) assumes that both K and the canopy resistance term (parametrized using the drag coefficient C_d, surface area of vegetation per unit volume α_c, and wind speed u) are constant in the upper region of the canopy, for the vertical extent of the tree corona (i.e. in the canopy, where l, the in-canopy mixing length, is assumed constant). The numerical models of Peters and Eiden (1992) and Wiman and Ågren (1985) also rely on application of K theory and parametrization of K as a function of the in-canopy mixing length, but unlike the model of Slinn (1982), $K(z)$ is not a constant inside the forest. For the forest case study considered by Peters and Eiden (1992) and the numerical model they present, differences in $v_d(D_p)$ computed assuming K has a constant value in the canopy (as assumed by Slinn (1982)) and for K varying with height (as in their model) were generally relatively modest, but for D_p of 0.1–0.5 μm the model run with a height invariant K showed values that are up to 50% lower than those derived using the K scaling they proposed.

(2) The mass density solution is obtained using the momentum analogy (which recovers the average horizontal wind profile when $\epsilon = 1$) and constitutes only the horizontal component of

the particle flux. Additionally, the overall accuracy of the solution is only as good as the average horizontal wind profile to which the particle flux analogy is made and is dependent on the appropriateness of the analogy (Raupach et al., 1996). This wind profile is formulated in terms of the within-canopy eddy length scale and the surface roughness length. Both of these are difficult to assign with a high degree of confidence. The latter may be estimated observationally and under near-neutral stratification and the former likely scales with leaf width and leaf area density (Goudriaan, 1977).

(3) With the exception of v_g, the influence of particle size on v_d is accounted for only through the collection efficiency coefficients. However, the particle size plays an important role earlier in the process in terms of the size distribution of the available particle budget (i.e. strong turbulence present in the upper canopy maintains larger particles airborne which in the absence of turbulence would settle by gravity).

In eq. (11) the collection efficiency is comprised of components deriving from Brownian motion, interception, and impaction and is corrected for particle rebound. The semi-analytical descriptions of particle collection efficiencies used by Slinn (1982) were largely derived from wind tunnel studies in the late 1950s and 1960s (Chamberlain, 1967). These descriptions are as follows:

$$E_B = \frac{c_v}{c_d} Sc^{-2/3} \tag{14}$$

$$E_{IN} = \frac{c_v}{c_d}\left[\overline{F}\left(\frac{D_p}{D_p + A_1}\right) + (1 - \overline{F})\left(\frac{D_p}{D_p + A_2}\right)\right] \tag{15}$$

$$E_{IM} = \frac{St^2}{1 + St^2} \tag{16}$$

$$St = \frac{\tau u_*}{c A_2} \tag{17}$$

$$\tau = \frac{v_g}{g} \tag{18}$$

$$v_g = \frac{\rho D_p^2 g C_c}{18\mu}. \tag{19}$$

Assignment of appropriate values for model parameters such as A_1, A_2 is also a source of uncertainty in model-derived particle size resolved v_d.

The model of Slinn (1982) has been widely used due to its analytic form, and hence ease of implementation but, as indicated above, alternative particle deposition models are also available. Most require numerical solvers (e.g. Peters and Eiden, 1992; Wiman and Ågren, 1985; Wiman et al., 1985), or consider only a specific portion of the particle size distribution (Bache, 1979a,b). In all cases they are either variants of the Slinn (1982) model (Zhang et al., 2001), or maintain a number of simplifying assumptions invoked by Slinn (1982). For example, the models developed by Bache (1979a,b), Giorgi (1986), Peters and Eiden (1992), Wiman and Ågren (1985) and Wiman et al. (1985) maintain an eddy diffusivity based approach.

Multiple alternative formulations for E_B, E_{IN} and E_{IM} have been derived subsequent to the work of Slinn (1982) which substantially alter their magnitude and v_d for a given D_p (Zhang et al., 2001; Gallagher et al., 2002; Pryor and Binkowski, 2004) but the pronounced minimum in particle v_d for $D_p \sim 0.1$–2 μm is a persistent feature of this model [and others (Peters and Eiden, 1992)] when applied to forests (Figs. 1 and 2). In this region of the particle size distribution none of the collection efficiencies formulated by Slinn (1982) are particularly efficient (Fig. 2). E_B dominates for the smaller particles and declines rapidly with increasing D_p, while E_{IN} and E_{IM} are of larger magnitude for $D_p > 1$ μm.

An example of the variability in the efficiency terms derived from differing formulations drawn from the literature is given in Fig. 2. Particle impaction (E_{IM}) is parametrized as a function of the Stokes number which is the ratio of the stopping distance of a particle to the characteristic dimension of the obstacle. Some formulations for *St* used in dry deposition models tend to emphasize the nature of the flow field in determining the magnitude of *St* (Binkowski and Shankar, 1995) while the formulation of Slinn (1982) focuses on the individual obstacles (leaves). For some formulations of *St* and E_{IM} drawn from the literature, the magnitude of the impaction efficiency for $0.1 \leq D_p \leq 1.0$ μm exceeds the interception efficiency as computed using the formulation of Slinn (1982) and using input parameters from the Speulderbos conifer forest in the Netherlands (Ruijgrok et al., 1997) while for alternative formulations $E_{IN} \gg E_{IM}$ in this size range (Fig. 2b). As illustrated in Fig. 2 use differing efficiency formulations change the slope of the dependence of the removal efficiency on D_p and thus can alter the magnitude and location of the minimum v_d, but the presence of the minimum v_d persists (Pryor and Binkowski, 2004).

Figure 2c shows results from the model by Slinn (1982) and that of Zhang et al. (2001) for simulations of a forest with the characteristics of the land use land cover class 1—an 'evergreen-needleleaf' forest as described by Zhang et al. (2001). Also shown are v_d estimates derived from observations drawn from experiments conducted in forest environments (Table 1). As shown, v_d for submicron D_p are underestimated by the model of Slinn (1982) relative to the observations, while the model of Zhang et al. (2001) shows rather better agreement with the observations but tends to overestimate v_d for $D_p < 0.2$ μm except in the case of data derived using application of an on-line REA system at the Scots pine forest at Hyytiälä in Finland (Grönholm et al., 2007). While the agreement between the model of Zhang et al. (2001) and data from Grönholm et al. (2007) is notable, it is worth mentioning that the mean surface roughness length around the Hyytiälä site is considerably higher than the z_0 assumed for this

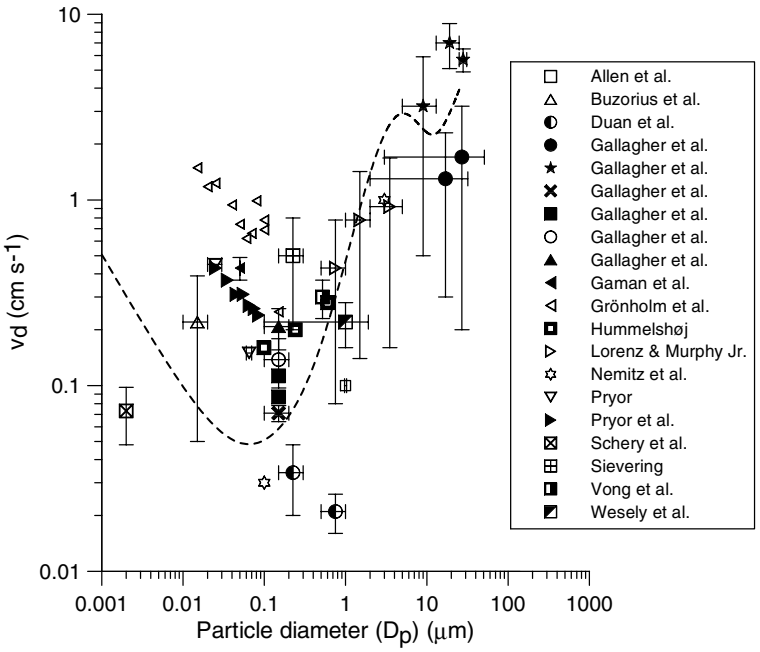

Fig. 1. Synthesis of studies that have sought to relate average particle v_d to particle diameter (D_p). The symbols are shown at the arithmetic mean of the particle size range and the error bars show the range of D_p sampled and the error bounds computed by the original researchers. Note that this figure differs from that presented by Gallagher et al. (1997b) in terms of the data sources and also that this figure summarizes data across a range of vegetation types, while that of Gallagher et al. (1997b) focussed on high surface roughness. The symbols encode the surface type (squares represent grass, triangle forests, stars moorlands/heathlands and circles arable/crops). A description of the studies from which data are presented is given in Table 1. The dashed line shows particle v_d from the model of Slinn (1982) for a representative forest.

land use class by (Zhang et al., 2001) (1.2 m versus 0.8 m), and that when a z_0 of 1.2 m is used in the model the simulated values of v_d increase beyond those that characterise the observations. A striking aspect of Fig. 2c is that the model as formulated by Zhang et al. (2001) exhibits minimum v_d for substantially larger D_p than the model of Slinn (1982), or the models of Wiman and Ågren (1985) or Peters and Eiden (1992), leading to an apparent underestimation of v_d for particles of approximately 1 μm diameter by the model of by Zhang et al. (2001) relative to the limited data sets available.

Because of the complexity and lack of coherent/inclusive models, many operational air quality models assume one v_d for all particles irrespective of size and/or composition (e.g. Park et al., 2001; Sickles II and Shadwick, 2002) or calculate v_d accounting only for atmospheric variations (Luo et al., 2002) or surface characteristics (Brook et al., 1999) rather than particle properties. Particle concentrations over the entire globe for past and possible future climate states computed using off-line particle models driven by general circulation model output for limited particle ensembles typically used a fixed v_d by particle origin (Takemura et al., 2003), and even one of the new generation of couple general circulation models with embedded particle dynamics—ECHAM5-HAM (Stier et al., 2005) uses the empirical formulation of Wesely et al. (1985) for v_d over land [where $v_d = (u_*, \mathrm{L})$, see Section 5.3].

4. Advances in observational approaches

With a few exceptions (Duan et al., 1988; Sievering, 1987, 1989) prior to the 1990s, particle fluxes were derived from differential methods (e.g. dry deposition was derived as the difference between throughfall and wet deposition (Parker, 1990; Rea et al., 2001), using surrogate surfaces (Goldenberg and Brook, 1997) particularly to quantify the flux of trace elements in coarse mode particles (Zufall et al., 1998; Sabin et al., 2006), or via gradient approaches (Sievering, 1986; Lorenz and Murphy Jr., 1989; Hummelshøj, 1994).

Gradient approaches employ first-order closure and the analogy with molecular diffusion to infer the flux from a vertical concentration gradient and a turbulent diffusion coefficient which describes the ease of transport (Panofsky and Dutton, 1984):

$$F = -K\frac{\mathrm{d}C}{\mathrm{d}z}. \tag{20}$$

The principle of similarity is then used to avoid the need to specify K for particles under the assumption that the eddy diffusivities for particles, momentum and/or heat are identical. The simplest case from a measurement perspective is to invoke the momentum analogy under which the flux can be derived from:

$$\frac{\tau_m}{F} = \frac{-\rho K_m \frac{\mathrm{d}\overline{U}}{\mathrm{d}z}}{-K_p \frac{\mathrm{d}\overline{C}}{\mathrm{d}z}}, \quad \text{i.e.} \quad F = \frac{\tau_m}{\rho}\frac{\overline{\mathrm{d}C}}{\overline{\mathrm{d}U}} \tag{21}$$

$$\tau_m = -u_*^2\rho. \tag{22}$$

Use of the heat analogy likely is more robust to assumptions regarding similarity at least for some portions of the particle size distribution (Pryor et al., 2007a), but can be problematic in near-neutral stability when $\frac{\mathrm{d}T_a}{\mathrm{d}z} \rightarrow 0$.

In the case of particles with non-negligible settling velocities, eq. (20) can be extended by assuming vertical transport of particles of a given size from turbulent transport and particle settling

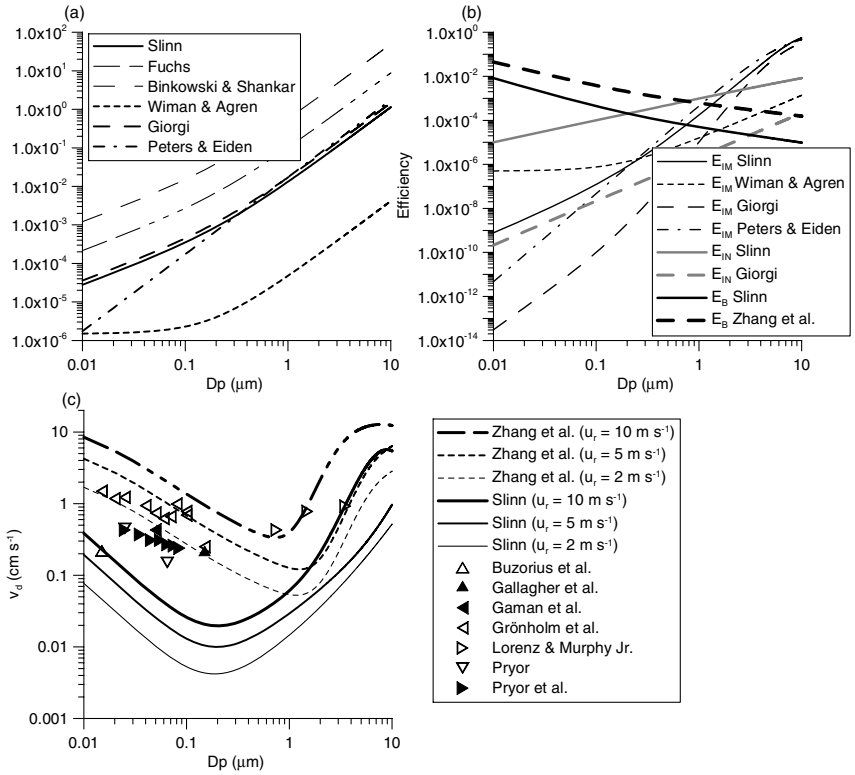

Fig. 2. (a) Stokes number and (b) surface collection efficiencies computed using different formulations available in the literature (Slinn, 1982; Wiman and Ågren, 1985; Giorgi, 1986; Peters and Eiden, 1992; Binkowski and Shankar, 1995). The formulations are shown in detail below the figure. Frame (c) shows deposition velocities by particle size computed using the models of Slinn (1982) and Zhang et al. (2001). The model of Zhang et al. (2001) uses E_{IM} from Peters and Eiden (1992) and E_{IN} from Giorgi (1986).

Source	Slinn (1982)	Fuchs (1964)	Binkowski and Shankar (1995)	Wiman and Ågren (1985)	Giorgi (1986)	Peters and Eiden (1992)
Formula	$St = \frac{\tau u_*}{A_2}$	$St = \frac{\tau U_0}{d_c}$	$St = \frac{u_*^2 v_g}{g v}$	$St = \frac{m_p B u_h}{1+\frac{f_c}{D_p}}$, $B = \frac{C_\lambda}{3\pi\mu D_p}$	$St = \frac{\frac{v_g}{g} u_*^2}{d_c}$	$St = \frac{\rho_p D_p^2}{9\mu d_c}$

Source	Slinn (1982)	Wiman and Ågren (1985)	Giorgi (1986)	Peters and Eiden (1992)
Formula	$E_{IM} = \frac{St^2}{1+St^2}$	$E_{IM} = \frac{St}{3+St}$	$E_{IM} = \left(\frac{St}{0.6+St}\right)^{3.2}$	$E_{IM} = \left(\frac{St}{0.8+St}\right)^2$

In these calculations A_2 is the characteristic dimension of large collectors (2.0 mm, assumed $= d_c = r$).

are additive (Csanady, 1973):

$$F = -K_{\mathrm{p}} \frac{\mathrm{d}C}{\mathrm{d}z} - v_{\mathrm{g}} C. \tag{23}$$

The number of ambient studies using gradient sampling to determine particle exchange has declined in recent years (see Table 1). This may be due in part to the difficulty in measuring very small gradients over high roughness surfaces (Rattray and Sievering, 2001), differences in footprints with height, and uncertainties in the form of stability corrections to the flux-profile relationship even for heat and momentum (Högström, 1988). However, this technique has been applied to quantify specific chemical components of the particle flux (Nemitz et al., 2000b) and for large particles and cloud water/fog droplets (Dollard et al., 1983; Gallagher et al., 1988; Kowalski and Vong, 1999).

These issues, combined with the evolution of particle measurement techniques (McMurry, 2000a, b), mean application of alternative flux techniques is increasingly commonplace. Now the majority of ambient particle flux measurements over vegetated surfaces conducted by micrometeorologists rely on

application of the eddy covariance technique (Table 1), where fast (typically several Hz) measurements of particle concentration and vertical wind velocity are used to derive the turbulent flux. Formally

$$F = \overline{w'C'} = \lim_{T \to \infty} \frac{1}{T} \int_{t_0}^{t_0+T} w'(t)C'(t)\mathrm{d}t, \tag{24}$$

eq. (24) is operationalized as:

$$F = \overline{w'C'} = \frac{1}{t_2 - t_1} \int w'(t)C'(t)\mathrm{d}t. \tag{25}$$

The primed quantities are instantaneous deviations from the time averaged mean which is typically computed over integration times of up to 1 hr.

For a discussion of the general issues pertaining to eddy covariance determination of scalar fluxes we direct the reader to Businger (1986) and Finkelstein and Sims (2001). We constrain our current discussion to issues specific to particle fluxes. Particle concentration measurements used in eddy covariance may either represent total or size-resolved number concentrations and are most commonly, but not exclusively, obtained using condensation particle counters (CPC) or optical particle counters (OPC). The major challenge that confronts use of eddy covariance is that the accuracy of fluxes is dependent on the counting statistics of the particle instrumentation (Wyngaard, 1973; Duan et al., 1988; Buzorius et al., 2000; Pryor et al., 2007a). This limitation generally constrains the use of eddy covariance to consideration of particles with diameters below ∼0.1 μm except under circumstances when larger particles are sufficiently numerous to generate robust statistics or instrumentation is operated with very high flow rates to increase the number of detected particles (Sievering, 1983; Duan et al., 1988; Vong and Kowalski, 1995). The limitation imposed by counting statistics is equally true of the use of differential mobility systems to pre-select a subset of particles on the basis of size for presentation to CPC (Buzorius et al., 2003).

The accuracy of derived fluxes is also dependent on stationary behaviour of the scalars which typically limits the integration period (T) to ≈ 1 hr (Kaimal and Finnigan, 1994). Particle concentrations exhibit non-stationary behaviour more frequently than other scalars which may further restrict T (Lamaud et al., 1994a,b; Buzorius et al., 1998; Rannik et al., 2003). Hence, another major question in making particle flux measurements using eddy covariance is how long is long enough? That is, what is the ideal integration period for flux calculation (Lenschow et al., 1994). Wyngaard (1973) and Lumley and Panofsky (1964) sought to determine the averaging time necessary to generate useful approximations of turbulent properties, and assert the error or uncertainty (given as the standard deviation on the ensemble statistics) on fluxes $\overline{w'x'} = \overline{w'u'}$, $\overline{w'C'}$ ... is given by:

$$(\delta F_x)^2 = \frac{2\Im_x}{\mathrm{T}}\left[\overline{(w'x')^2} - \overline{w'x'}^2\right]. \tag{26}$$

To address this question in an operational context we computed the fraction of derived fluxes that exceed the uncertainty (or error estimates derived as above) for particle, momentum, heat and humidity fluxes for varying T using data collected at two forest sites:

(1) The 80-yr-old beech (*Fagus silvatica L.*) stand at Sorø (55°29′N, 11°38′E, 40 m above sea level) (Pilegaard et al., 2003).

(2) The SMEAR II station is located in a relatively expansive homogeneous Scots pine stand (*Pinus sylvestris* L.) next to the Hyytiälä forest station in southern Finland (61°51′N, 24°17′E, 181 m above sea level) (Kulmala et al., 2001).

We consider T of 10 min to 3 hr and assume (Wyngaard, 1973):

$$\Im_x = \Im_u = \Im_w = \Im_C = \frac{z}{\overline{u}}. \tag{27}$$

We further assume that the fraction of computed fluxes that exceed the uncertainty bounds may be used as a metric of the quality of the fluxes, and in keeping with the procedure most commonly used at Fluxnet sites conduct the coordinate rotation separately for each period. The results indicate that obtaining robust particle fluxes at any integration period is more challenging than momentum and heat fluxes (Fig. 3). It may also be inferred that an integration period of 30 min may be 'optimal' for particle fluxes (at least at Hyytiälä and likely Sorø) and that increasing T leads to only very limited improvement in particle flux estimation. According to eq. (26) the error scales linearly with height; thus, it should also be noted that at both sites the effective z (computed to account for the displacement height) are of the order of 10–15 m. Hence these findings are applicable to relatively near-canopy measurements. The assessment of a 'significant' flux is rather subjective. Here we use a threshold, $|F| - \delta F > 0$, noting that for a Gaussian distribution 68% of data values lie within $\pm$ 1σ of the mean.

As the integration time increases beyond 30 min (i.e. $T >$ 30 min) the fraction of particle number fluxes for which $|F| > \delta F$ rapidly asymptotes (Fig. 3). From Eq. (26) it can be seen that $(\delta F)^2$ scales with $1/T$, hence the inference is that as $T \to \infty$ there are compensating effects manifest in the particle flux data that act to prevent δF increasing beyond the level attained for $T = 30$ min. The results from the other scalars exhibit a higher fraction of the fluxes exceed δF for a given integration period (T) and the fraction of fluxes for which $|F| > \delta F$ show continued increases for T beyond 30 min. We postulate that differences in the behaviour between particle fluxes and the other scalars derive principally from differences in the scales and spatial variability of sources/sinks of particles versus the other variables (Lenschow, 1995).

The accuracy of eddy covariance derived flux estimates is also dependent on application of flux corrections (Fairall, 1984; Buzorius et al., 2000; Pryor et al., 2007a) to remove the confounding influence of covariance of the saturation ratio with the vertical wind velocities (Kowalski, 2001), the WPL-terms for

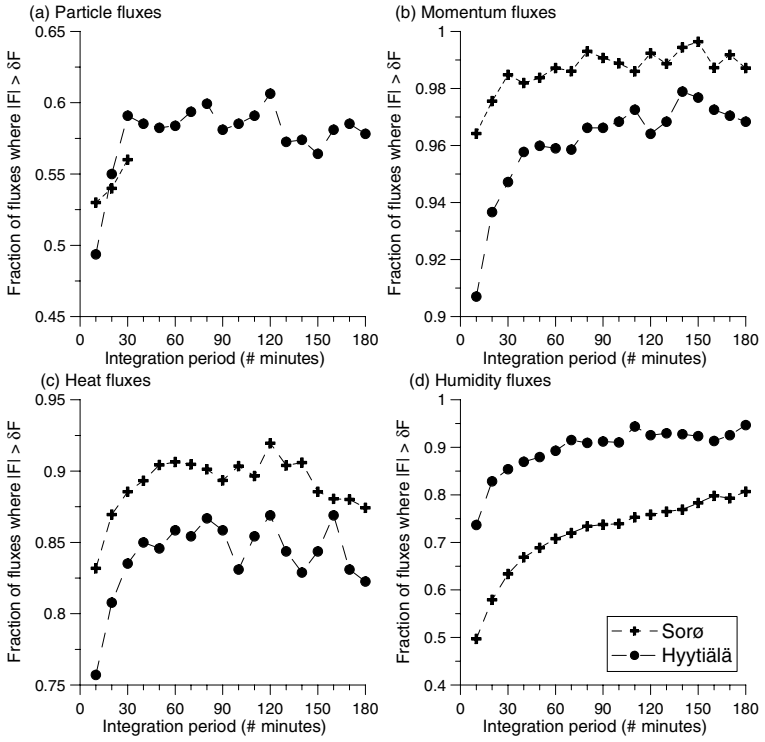

Fig. 3. Analysis of the influence of integration period (run length) on the statistical significance of flux measurements. Results are shown using observations from two forest sites (Sorø which is a mixed deciduous forest and Hyytiälä a pine forest). Results are shown for four parameters (a) particle number flux, (b) momentum flux, (c) sensible heat flux and (d) humidity fluxes. With the exception of the particle number fluxes at Sorø, the calculations were based on continuous measurements obtained over a 3 month period in early 2004. The particle flux measurements from Sorø are derived from a much shorter data set collected over a 3-week period in May–June 2004, and hence are considerably less robust. The differences in results for the humidity measurements at both sites derive from differences in experimental set-up. While humidity measurements from both sites were derived from closed-path Licor systems, the tubing at Hyytiälä is considerably shorter than at Sorø leading to reduced attenuation of the signal.

density effects (Webb et al., 1980), and the correction for truncation of the particle spectrum due to the frequency response of instrumentation (Horst, 1997). In some cases these corrections may be of comparable magnitude to the raw flux estimates. In a recent study of data collected at the Sorø forest site the mean magnitude of the WPL correction to particle fluxes had an average value of <1% of the flux (Pryor et al., 2007a). In this same study the average flux underestimation due to the attenuation of the high frequencies was computed to be 13% of the raw flux, and the average correction due to correlation of fluctuations in the saturation ratio with vertical wind speed was 37% of the raw flux (Pryor et al., 2007a). The particle flux community has not yet identified preferred and universally applied methods for making flux corrections or deriving robust uncertainty estimates, but just such a protocol would make a substantial contribution to advancing particle flux research.

As a result of the challenges in using eddy covariance for determining particle fluxes other studies have focused on development/application of alternative micrometeorological approaches such as Eddy Accumulation (EA), proposed by Desjardins (1977). Relaxed Eddy Accumulation (REA) is a derivative of this approach that was first proposed by Hicks and McMillen (1984) and formulated in detail by Businger and Oncley (1990). In REA as w exceeds a threshold ('dead-band') velocity, air is differentially sampled at a constant flow rate. Thus the flux is determined from differential sampling of particle concentrations in updrafts and downdrafts (Oncley et al., 1993):

$$F = b\sigma_w(C_{\text{up}} - C_{\text{down}}). \tag{28}$$

The REA technique is based on a number of model assumptions (such as similarity of scalar transport) and as noted by Kramm et al. (1999) should be considered a 'one-and-a-half-order closure scheme'. Uncertainties associated with the REA-derived fluxes may be computed using work by Kramm et al. (1999) who formulated the mean relative error in the REA derived flux ($\frac{\delta F}{F}$) (derived using the sensible heat analogy) using Gaussian error propagation principles as:

$$\frac{\delta F}{F} = \sqrt{\left(\frac{\delta H}{H}\right)^2 + 2\left(\frac{\delta C}{\Delta C}\right)^2 + 2\left(\frac{\delta \theta}{\Delta \theta}\right)^2}. \tag{29}$$

REA may be applied either as a measurement technique (Schery et al., 1998; Nemitz et al., 2000a; Gaman et al., 2004; Meyers et al., 2006), or as a post-processing tool (Pryor et al., 2007a). Application of REA to particle fluxes as a measurement technique is technically challenging, and sampling artefacts may be introduced for particles, due to the need for fast switching of airflows and/or particle losses in storage reservoirs. Both features are often associated with REA setups. Use of REA as a post-processing tool applied to high-frequency data (Pryor et al., 2007a, b) may have advantages over the eddy covariance approach because although δF is dependent on accurate determination of b (Ruppert et al., 2006), it is a function of the relative accuracy of mean particle concentrations differentially sampled by the sign of the vertical velocity rather than higher moments of the probability distribution as is the case in eddy covariance. To evaluate this in a practical context, we computed half-hour particle number fluxes at the Hyytiälä site using data collected during

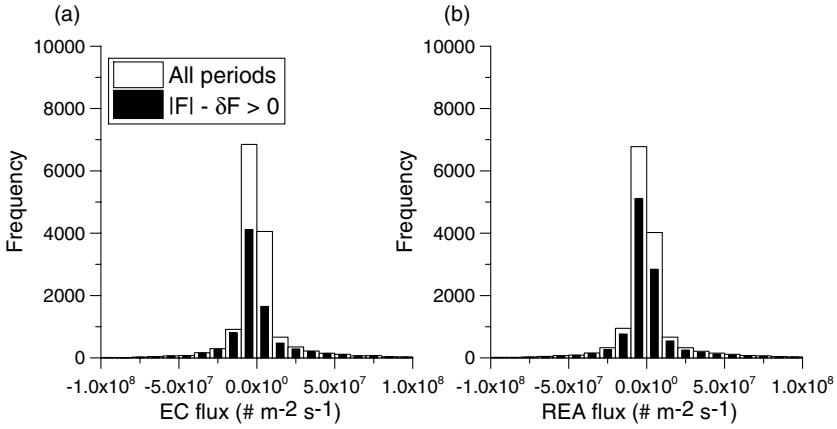

Fig. 4. Particle number fluxes computed using a half-hour integration period and 12-months of data from the Hyytiälä forest site. Frame (a) shows the results of application of eddy covariance, while (b) shows the results of application of the REA technique as a post-processing tool. In each frame the white bars show histograms of the flux estimates from every half-hour period. The black bars represent the frequency of flux magnitudes where only fluxes that exceed the uncertainty bounds (δF) are included. Note the range of flux magnitude has been truncated to show the middle 95% of realizations for readability.

January to December, 2004. We computed fluxes using EC and REA applied as a post-processing tool (using the sensible heat analogy). The raw fluxes were then subject to the flux corrections outlined above following the techniques applied in Pryor et al. (2007a). We then applied the uncertainty bounds derived using eqs (26) and (29). The results indicate that the mean ratio of EC to REA fluxes differs from 1 by less than 0.1%, indicating some confidence that the assumptions on which the REA methodology are met (i.e. scalar similarity). Further, more of the REA derived fluxes exceed the uncertainty computed using eq. (29) (Fig. 4). Note that in this analyses we deem the eddy covariance and REA derived fluxes to be statistically significant fluxes if the absolute magnitude of the flux exceeds the uncertainty computed using eqs (26) and (29), respectively (i.e. $|F| - \delta F > 0$). It should also be reemphasized that use of REA either as a direct measurement technique or applied in a post-processing context is dependent on scalar similarity to derive b which does not hold in all circumstances (Kramm et al., 1999; Ruppert et al., 2006) and, as mentioned above, does not alleviate the need to apply flux corrections associated with, for example, vertical humidity gradients and particle hygroscopicity.

We are aware of only one study that applies spectral techniques to obtain particle fluxes over vegetated surfaces (Pryor et al., 2007a). However, the inertial-dissipation method that has been largely pioneered for use in air–sea momentum and heat exchange studies (Fairall et al., 1990) is also potentially applicable to particle fluxes and terrestrial surfaces. This method, like REA, invokes Monin–Obukhov similarity theory. It also employs universal laws for inertial subrange turbulence to derive particle fluxes from the $-5/3$ slope of the high-frequency region of the power-spectra, S(*as a function of frequency* (f):

$$u_* = \left[\frac{f\,S_U(f)}{\alpha}\right]^{1/2}\left[\frac{2\pi}{\overline{U}}\frac{\kappa z}{\phi_\varepsilon} f\right]^{1/3} \tag{30}$$

$$|F| = \left[\frac{\alpha}{\beta}\frac{\phi_\varepsilon}{\phi_{NC}}\frac{f\,S_C(f)}{f\,S_U(f)}\right]^{1/2} u_*^2 . \tag{31}$$

See Edson et al. (1991) for details of the technique and an analysis of the uncertainties in flux estimates associated with use of the inertial-dissipation technique.

As shown by eq. (31) a potential limitation of this approach is that the sign of the flux needs to be determined externally to the calculation of the flux magnitude. An additional constraint on application to particle fluxes is that truncation of the frequency spectrum by instrument response times and other effects leads to much noisier power spectra than for other passive tracers (Duan et al., 1988; Pryor et al., 2007a).

As discussed above and shown in Fig. 1, recent estimates of v_d for sub-200 nm diameter particles derived using an on-line REA system with particle sizing by a DMPS at the Hyytiälä forest station in southern Finland (Grönholm et al., 2007) are considerably higher than estimates at this site derived using eddy covariance with inferred particle diameter from an independent DMPS system (Buzorius et al., 2000; Pryor et al., 2007b). The source of this discrepancy is currently unknown but it may be linked to physical causes such as increased roughness length due to logging activities close to the flux tower or variations in meteorological conditions between the various studies, or it may reflect methodological differences, such as the following:

(1) Each micrometeorological method relies on different statistical properties of the particle data and thus the associated uncertainties differ (cf. eqs 26 and 29), and each flux estimate may reflect different flux footprints. Source area dimensions are considerably higher for scalar concentrations than scalar fluxes (Schmid, 1994), hence over inhomogeneous surfaces fluxes computed using techniques that reflect different moments of the particle number probability distribution may differ as a result of differences in the associated source areas.

(2) Differences in the application of various flux corrections (Pryor et al., 2007a) by different authors.

(3) Issues pertaining to particle transfer efficiencies in the REA system, difficulties in deriving robust statistics of the particle counts when size selection is employed, or difficulties in assigning a representative median diameter from particle size

spectra derived using an SMPS or DMPS system to the flux estimates derived using EC from a CPC.

As discussed in Section 6, further analysis and intercomparisons of different micrometeorological flux techniques is warranted.

Despite advances in particle measurement technologies and theoretical advantages of the micrometeorological techniques described above for flux estimation, such studies have tended to focus on instrumentation to measure fluxes of particle numbers, without knowledge of the chemical speciation. The measurement of composition resolved aerosol fluxes has until recently relied on gradient techniques, or the use of either surrogate surfaces (Golomb et al., 2001), or mass balance approaches (Ivens et al., 1990; Draaijers et al., 1994; Horvath, 2003). See also the summary in Davidson and Wu (1990). Additionally, most routine monitoring networks utilize particle phase chemical concentrations from filter packs with models of particle v_d to estimate mass fluxes (Hicks et al., 2001) without detailed size distribution data. An important issue with such approaches that has long been noted is that long integrative filter pack measurement techniques may suffer from site specific volatility artefacts especially for nitrate aerosol which may be lost from filter surfaces (Weber et al., 2003) so net fluxes for these species may be underestimated, and may not compare well with those derived from micrometeorological approaches.

Recently, the advent of Aerosol Mass Spectroscopy (AMS) has facilitated development of approaches to measure chemically speciated particle fluxes by eddy covariance (Nemitz et al., 2006). Also the requirement of fast time-response analysers in the eddy covariance method has been relaxed by the use of disjunct eddy covariance (DEC) (Dabberdt et al., 1993). DEC has been applied to trace gas fluxes (Rinne et al., 2001) but has yet to be used for particle fluxes. Application of the disjunct eddy accumulation approach (Rinne et al., 2000) with instrumentation capable of sizing and chemically resolving individual particles (Held et al., 2003) is, however, a step in that direction.

In closing this section on experimental methods it should be noted that other flux estimation procedures have been developed (e.g. the flux variance approach, Tillman, 1972; Albertson et al., 1995; Wesely, 1998) but to the author's knowledge they have not been widely applied to particles (Lamaud et al., 1994b). Further we reemphasize that errors in particle flux measurements depend on the particle climate and prevailing meteorology in the environment where the measurements are being taken, the micrometeorological technique used to compute the flux and the instrumentation applied. As noted herein particle instrumentation is rapidly evolving but for a summary of some technical aspects of currently applied instruments we direct the reader to the excellent synthesis of McMurry (2000a and references therein), and for an update on aerosol mass spectrometers to the analysis of Allan et al. (2003).

5. Particle flux dependencies: new insights from experimental data

From eq. (6) and the following discussion it can be inferred that there are three dominant driving forces for dry deposition of submicron diameter particles—particle diameter, friction velocity and/or surface roughness, and stability. Figures 1 and 5 present these dependencies based on some of the observational studies over vegetated surfaces summarized in Table 1. The sections below articulate experimental research pertaining to these flux dependencies and particle rebound (Paw U, 1983) and resuspension. Note that the formulations presented in this section are empirical in nature and generally are not theoretically derived.

5.1. Particle diameter

On the most fundamental level, as shown in Fig. 1, in accord with the increase in the terminal fall velocity, supermicron particles exhibit higher v_d than do submicron particles. However, there is some evidence that the magnitude of the difference between v_d for $D_p > 2$ μm and $D_p \sim 0.1$–2 μm may vary by surface type, and specifically that these differences are smallest for forests. Also, in contrast to models such as that of Slinn (1982), observational studies over forest canopies do not appear to indicate substantially different mean v_d for D_p of 20–100 nm (Gaman et al., 2004; Pryor, 2006; Grönholm et al., 2007; Pryor et al., 2007b) than for D_p of approximately 100–800 nm (Gallagher et al., 1997a, 2002; Lorenz and Murphy Jr., 1989). There is large scatter and uncertainty bounds on the experimental measurements and some of the variability between studies may reflect the accuracy of the particle D_p measurements, possible stability effects and issues pertaining to whether the reported D_p relates the wet or dry diameter. Nevertheless, the observations over forests do not replicate the clear v_d minimum for $D_p \approx 0.1$–2 μm that models such as that by Slinn (1982) exhibit. There has recently been partial validation of the model of Slinn (1982) for particles with diameters (D_p) ≤ 0.1–0.2 μm over forests (Gallagher et al., 2002), and for particles with $D_p = 0.1$–3 μm in a moorland setting (Nemitz et al., 2002b). However, it is notable that agreement between measurements and the model of Slinn (1982) could only be achieved if coefficients used in the Slinn (1982) parametrizations were increased significantly, close to their limits, which suggests that the physical underlying principles may be in error or that processes may be missing from the model formulations. Further, these measurements (from the work of Gallagher et al., 2002 and Nemitz et al., 2002b) were conducted with the same instrumentation and experimental approach using the combined efforts of both researchers. Although they used independent data analyses to reach the same results, they require further independent verification.

It should also be noted that the more recently collected data compiled in Table 1 and shown in Fig. 1, imply slightly lower v_d for $D_p \sim 0.1$–2 μm, than were obtained using inferential

techniques and reported by Gallagher et al. (1997b) (cf. Fig. 1 with fig. 3.6 in Gallagher et al., 1997b). Nevertheless, as first noted over 20 yr ago (Wesely et al., 1977; Sehmel, 1979; 1980; Hicks et al., 1982; Gallagher et al., 1997b) and discussed below, we assert that there are still substantial and systematic discrepancies between process-level models of particle dry deposition and available measurements over forested surface, particularly for $D_p \approx 0.1$–1 μm. Given that forests are likely the most effective sink for submicron particles, this discrepancy has important implications for modelling particle size distributions and concentrations across a range of spatial scales. The discrepancy for high-roughness (i.e. forests) between measurements and models, and specifically the lack of a pronounced size-dependence in particle v_d, is in contrast with the measurements of Gravenhorst and Hofken (1982). Their data, collected using cascade impactors, clearly indicated a size-dependence of canopy filtration by particle diameter ($D_p \sim 0.26$–2.4 μm mass median diameter) in the comparison of above and below canopy particle concentrations.

Several postulates have been proposed to explain the discrepancy between micrometeorologically derived observations and process level models of the particle flux over high roughness vegetated surfaces:

(1) *Observational errors:* The discrepancy could be explained if the minimum were over a relatively narrow size range not resolved by the measurements or possibly due to instrument performance issues. Indeed, this may be offered as a partial explanation in the case of data from the Hummelshøj (1994) study which employed an optical particle counter (OPC) and was conducted over grassland. For the analogue circuitry employed in OPC from that era there is certainly a possibility of coincidence errors due to high particle concentrations at the small sizes, however particle concentrations were comparatively low during this study, and there is convergence of data from varying particle regimes (Table 1 and Fig. 1).

(2) *Chemically induced flux divergence:* While ambient measurements reflect the net removal or introduction of particles into the atmosphere, models such of that by Slinn (1982) only reflect the contribution of surface removal. Hence, other processes that lead to a reduction in, for example, the particle number concentration or mass of particle-bound components will lead to a discrepancy between the measurements and models. Gas-particle partitioning has long been recognized as one such process (e.g. Kramm and Dlugi, 1994). But Pryor and Binkowski (2004) demonstrate that, using the model formulation of Slinn (1982) or alternative published formulations for the components of dry deposition and for reasonable size distributions, coagulation may also cause substantial evolution of the particle size distribution below typical flux measurement heights and the actual receptor surface.

(3) Incorrect or inadequate treatment of the profiles of meteorological parameters responsible for particle transfer to and through the canopy. In studying vertical turbulent transfer inside tall vegetative canopies, Slinn (1982) assumed the diffusivity coefficient is constant with height for most of the vegetated canopy, except for a small layer near the ground. Moreover, as discussed above, the absorption function (represented by the product of the drag coefficient, wind speed, leaf area index and leaf area density) is also assumed to be constant throughout the vegetation layer. These assumptions lead to an exponential concentration profile inside tall vegetation (Ruijgrok et al., 1997). Although (in an averaging sense) the treatment of vertical turbulent transfer in the above manner is essentially correct, a more elaborate parametrization scheme is needed to represent the compound effect of both the vegetation and forest floor acting as particle sinks (Birsan, 2005).

(4) Under-estimation of collection efficiencies in the model of Slinn (1982). Gallagher et al. (1997a) demonstrated that observed v_d of 0.1–1.0 μm diameter particles to the Speulder forest were profoundly underestimated by the model of Slinn (1982), leading the authors to assert that the discrepancy may reflect errors in the collection efficiencies used in this model. Conditional sampling of particle fluxes over a deciduous forest by the particle ensemble number geometric mean diameter (GMD) also imply higher surface collection efficiencies than are employed in the Slinn model (Pryor, 2006). However, as noted in Section 3, changing the formulation of the efficiency terms to others within the published shifts the modelled v_d minimum in diameter space but does not remove it.

(5) Exclusion of additional deposition pathways or dependencies within the models. Most models of particle dry deposition to vegetated surfaces treat surface uptake in terms of impaction by inertial forces, interception, Brownian diffusion and gravitational settling (e.g. Slinn, 1982). However, there are additional forces or processes that may be important under specific circumstances. Those processes which have been the subject of recent intense research include electrostatic forces which were shown in one laboratory study (Tammet et al., 2001) to substantially increase the dry deposition of 10–200 nm diameter particles under low wind conditions.

As shown in Fig. 1 there is tremendous variability between measurements over superficially similar surfaces. Hummelshøj (1994) used the aerodynamic gradient method to compute particle v_d over a grass-field with a roughness length of $\leq$10 mm and found an increase in v_d as a function of size ranging from 0.16 cm s^{-1} for D_p of 0.065–0.15 μm ($\bar{D}_p \approx 0.099$ μm), to 0.20 cm s^{-1} for D_p of 0.15–0.4 μm ($\bar{D}_p \approx 0.24$ μm), and 0.28 cm s^{-1} for $D_p = 0.4$ and 0.9 μm ($\bar{D}_p \approx 0.6$ μm). These data show relatively good agreement with previous studies over grassland, particularly when the influence of stability is considered. However, Nemitz et al. (2002b) conducted eddy-correlation measurements over moorland (with $z_0 \approx 10$ mm) and found mean v_d of 0.03 cm s^{-1} for $D_p \approx 0.1$ μm, increasing to v_d of 1 cm s^{-1} for $D_p \approx 3$ μm. Thus the v_d for $D_p \approx 0.1$ μm from these two studies differ by a factor of five. As discussed herein this

may reflect subtle differences in the vegetation morphology or static stability of the atmosphere during the two studies, or the influence of chemical interactions.

With respect to the influence of particle diameter on v_d for smaller particles, models of Slinn (1982) and Peters and Eiden (1992) predict an order of magnitude higher v_d for 10 nm diameter particles than 100 nm particles for representative forests (see Fig. 1). However, little experimental data are available to confirm these projections. In a recent study, particle number fluxes over a deciduous forest were conditionally sampled by the prevailing GMD of the particle size distribution. It was found that the median v_d for particle ensembles with a GMD of 20–30 nm is 4.5 mm s^{-1}, decreasing to 1.5 mm s^{-1} for particle ensembles with a GMD of 60–70 nm (Pryor, 2006), while analyses of data from a pine forest imply declines in v_d of a factor of approximately two over this size range (Grönholm et al., 2007; Pryor et al., 2007b).

5.2. Friction velocity and surface roughness length

While the form of the relationship between v_d and u_* clearly varies with surface type, stability and particle diameter, virtually all studies synthesized for this review indicated an increase in particle v_d with u_* (Table 1). Gaman et al. (2004) applied a REA system to determine the number flux of 50 nm particles to a Scots pine forest and found the average v_d of 50 nm particles increases with u_* from approximately 5 mm s^{-1} at $u_* < 0.5$–15 mm s^{-1} for $u_* > 1.0$ m s^{-1} leading the authors to propose the following relationship:

$$v_d = 0.012\, u_* \tag{32}$$

Data from a deciduous forest in Denmark indicate a mean v_d for particle ensembles with number GMD of 50–60 nm increases from 1–1.8 mm s^{-1} at $u_* < 0.4$ m s^{-1} to 7.5 mm s^{-1} at $u_* > 1$ m s^{-1} (Pryor, 2006). Mean inferred v_d for sulphur containing particles having a mass-median diameter ~800 nm were 1 cm s^{-1} for $\overline{u_*} = 0.49$ m s^{-1}, and increased to 2.9 cm s^{-1} for $\overline{u_*} = 0.59$ m s^{-1} in a gradient based study over low vegetation by Sievering (1986). Although part of this increase may be due to stability effects, these data also imply a strong dependence on u_*, as did data collected using the gradient technique over the Speulder forest (Erisman et al., 1997; Wyers and Duyzer, 1997) leading to proposed v_d for specific ions with the form: $v_d = c_1 u_*^{c_2}$, where c_1 and c_2 are ion-specific coefficients. Over moorland median v_d for $D_p = 0.40$–0.45 μm increased from 0.3 mm s^{-1} to over 2 mm s^{-1} as u_* increased from 0.1 to 0.5 m s^{-1} (Nemitz et al., 2002b). Deposition of fog droplets (3–50 μm diameter) from eddy covariance was also found to vary with u_* (Vermeulen et al., 1997):

$$v_d = 0.195 u_*^2 \tag{33}$$

However, this study measured total cloud/fog liquid water content and there was no droplet size dependence information provided with the fluxes.

When data are synthesized over a range of experimental studies it appears that at very low u_* ($u_* < 0.15$ m s^{-1}) the relationship particle-v_d and u_* is non-linear (it appears to scale more closely with u_*^2), but as u_* increases the relationship becomes more linear (Fig. 5). This may imply a decreased role of r_a at high u_* and an increase in the relative importance of r_b which scales as $\frac{1}{u_*}$. However, the relative importance of storage and advective terms in dictating the particle flux under low u_* conditions may also be reflected in the apparent u_* dependence, as has been shown to occur for CO_2 exchange (Aubinet et al., 2005).

Table 1 supports earlier assertions that higher roughness surfaces exhibit higher particle v_d (Gallagher et al., 2002). This is expected principally due to a reduction in the aerodynamic resistance to transfer and accordingly there is abundant evidence for dependence of particle v_d on surface roughness or friction velocity. A synthesis of studies considering 100–200 nm diameter particles yielded the following relationship: $v_d = 0.581\ln(z_0)+1.86$, where z_0 is in m and v_d is given in mm s^{-1} (Gallagher et al., 2002).

5.3. Stability

There are insufficient data to allow definitive characterization of the dependence of particle fluxes on stability conditions, and indeed it should be noted that this dependence may be indirect and expressed via the dependence of u_* on atmospheric stratification. Although there is some evidence for an increase in particle v_d in unstable conditions and reduction with increasing thermal stratification (Everett et al., 1979; Sievering, 1983, 1987; Wesley et al., 1985; Hummelshøj, 1994; Lamaux et al., 1994; Fontan et al., 1997; Gaman et al., 2004; Vong et al., 2004) and during the daytime relative to the night (Hicks et al., 1983) few studies have been able to quantify the influence of stability with a high degree of statistical certainty. This may reflect the dominance of other processes in dictating the resistance to transport, and/or it may be a result of the relatively large statistical uncertainties in flux estimation. Wesely et al. (1985) and Walcek and Taylor (1986) first proposed the following relationships based on their measurements of sulphate particles with $0.1 < D_p < 0.3$ μm over grass:

For Monin–Obukhov length (L) > 0 m (stable to neutral conditions):

$$v_d = 0.002 u_*. \tag{34}$$

For $L < 0$ m (unstable to neutral conditions)

$$v_d = 0.002 u_* \left[1 + \left(\frac{-0.3 z_i}{L}\right)^{2/3}\right]. \tag{35}$$

The general form of these equations is implied by scaling analysis presented by Wyngaard (1973), and although research subsequent to Wesely et al. (1985) has postulated other values for the constants, the general form of the dependencies has been maintained (Lamaud et al., 1994a). Gallagher et al. (1997a) who

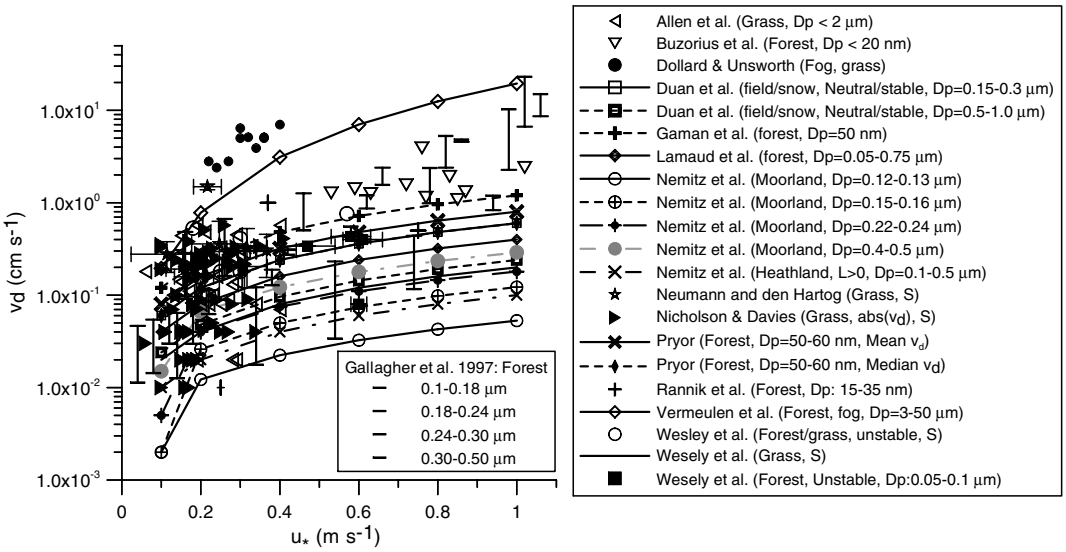

Fig. 5. Synthesis of studies that have sought to relate particle v_d to friction velocity (u_*). Where studies have reported 'point' data they are shown as individual points, where studies have either formulated the relationship or provided sufficient information for an equation to be formulated the resulting relationship is shown as a continuous line with symbols added to aid clarity. In this case the equations are only plotted for the range of u_* observed during the study from which the data are drawn. The data from Gallagher et al. (1997a) are given for bin averaged u_* with the range of observed v_d shown by the error bars. These data are given for four D_p ranges shown in order from lowest D_p to highest D_p. The data from the study of Rannik et al. (2000) also show the uncertainty bounds. Some of the discrepancies between individual studies are the result of variations in particle size (and composition), stability conditions and observational uncertainties. Even in the face of such variations there is clear evidence for an increase in v_d with increasing u_*.

proposed the following relationship for $0.1 \leq D_p \leq 0.5$ μm for a coniferous forest for -0.04 m$^{-1} < L^{-1} < 0$ m^{-1} (unstable to neutral conditions):

$$v_d = 0.0135 D_p u_* \left[1 + \left(\frac{-300}{L}\right)^{2/3}\right]. \quad (36)$$

Nemitz et al. (2004) also kept the same functional form in their research over heathland again for $0.1 < D_p < 0.5$ μm, leading them to propose:

For $L \leq 0$ m (unstable to neutral conditions):

$$\frac{v_d}{u_*} = \left\{a_1\left(1 + (-a_2 L^{-1}\right)^{2/3}\right\}. \quad (37)$$

For $L > 0$ m (stable to neutral conditions):

$$\frac{v_d}{u_*} = a_1. \quad (38)$$

With $a_1 = 0.001$, $a_2 = 960$ D_p-88, with a_2 having units of m, and D_p in μm.

The results of these different formulations are shown in Fig. 6. While there is no theoretical basis for this specific form of dependence on Monin–Obukhov length, this form exhibits asymptotic behaviour with increasing unstable conditions consistent with a decline in the relative importance (and in the limit case the absence of) the aerodynamic resistance.

At this juncture it appears the data are insufficiently robust to allow calculation of stability corrections for flux profile relationships. There is, however, a measurable effect of stability with the v_d being greatly decreased by highly stable conditions (small positive values of L), and greatly increased in unstable conditions (small negative values of L).

5.4. Resuspension

Several studies have observed bi-directional fluxes even in the absence of an obvious surface-based particle source. Emissions of supermicron particles are likely attributable to wind-driven resuspension (Nemitz et al., 2002a; Gillette et al., 2004a,b). Gillette et al. (2004a,b) used wind tunnel measurements to differentiate the effects of aerodynamic mechanisms (defined as viscous and turbulent mechanisms) from mechanical processes resulting from the receptor grass striking a stationary object in determining resuspension. They found, for spherical particles of $D_p = 2$–10 μm, a threshold wind speed of over 12 m s^{-1} was required for resuspension, and that aerodynamic effects were dominant for the smaller particles, while the aerodynamic and mechanical processes were of approximately comparable importance for the larger particles. These and other experiments have also observed an exponential decrease in resuspension through time (Nicholson and Branson, 1992; Ould-Dada and Baghini, 2001; Gillette et al., 2004a,b) implying a finite source of material that can be resuspended. For example, in the experimental

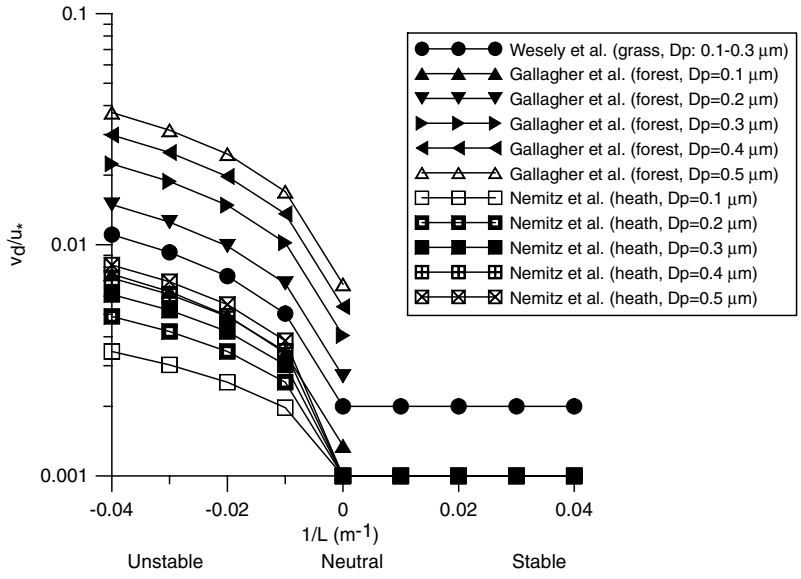

Fig. 6. Normalized v_d (by u_*) for submicron diameter particles under varying stability conditions (as reflected in the Monin–Obukhov length) as postulated from three studies conducted over different surface types. For details of the studies see Table 1.

research of Nicholson and Branson (1992) 15% source depletion of 22 μm diameter particles from a concrete surface was observed in the first 10 seconds for an ambient wind speed of 6.5 m s^{-1}.

Parametrizations for resuspension are likely to be strongly dependent on surface morphology. Recent size-segregated flux measurements conducted in an urban context indicate that the flux increases with increasing wind speed, following a power-law relationship consistent with previous studies (e.g. Nicholson and Branson, 1992). In addition, the size-spectrum of the resuspension flux peaks at around 3 μm. This shape is very likely the result of the competing effects of resuspension efficiency, which is known to increase with particle size, and the number size distribution of available material on urban surfaces, which decreases with increasing particle size. Overall, the following proposed formulation for the mass flux (μg m^{-2} s^{-1}) by resuspension can be fitted to the measurements (Nemitz et al., 2002a, modified):

$$\frac{\mathrm{d}F}{\mathrm{d}\log D_p} = \exp[-68.69 + 73.39 \times (1 - \exp(-0.730 D_\mathrm{p})]U^{20.12\exp(-0.506 D_\mathrm{p})}, \quad (39)$$

where D_p is given in μm, and U was measured at 65 m above street level.

For smaller particles resuspension likely plays a minor role and emission fluxes are more likely the result of particle dynamics (e.g. nucleation, growth or evaporation) or primary emissions principally from combustion processes.

6. Concluding remarks and identification of outstanding research questions

Technical developments over the last decade have substantially improved our ability to quantify particle fluxes experimentally and to assign uncertainties to those flux estimates. However, despite tremendous strides in our understanding of particle fluxes over the last decade, there remain substantial uncertainties. Below we articulate a few of the aspects of particle fluxes that would benefit from further investigation:

(1) Landscape heterogeneity has the potential to greatly and systematically affect atmosphere–surface energy and chemical exchange (Walcek et al., 1986; Draaijers et al., 1994; Wilson et al., 2001; Zhang and Brook, 2001; Hasager et al., 2003; Reithmaier et al., 2006). 'Edge effects' may be even more pronounced for particles, and are caused by advection of pollutants into the forest edge and, secondarily, by enhancement of turbulence due to the roughness discontinuity (Wiman and Ågren, 1985; Wiman et al., 1985; Ruck and Adams, 1991; De Jong and Klaassen, 1997; Dai et al., 2001). The specific activities of Pb-210 in dried soil samples have been used to infer that particle deposition at the exposed edge exceeds that in the open by $\geq$50% (Branford et al., 2004). Scaling for landscape heterogeneity across model grid-cells is a very challenging task but one that is of tremendous importance to the air quality and climate modelling communities and is a key issue in reconciling measurements and models. Hence, the influence of forest edge effects and other vegetation discontinuities on flux estimates merits further attention as does the influence of forest thinning on particle fluxes. Initial work in this field (Vesala et al., 2005) suggests that of the two competing effects on particle deposition (reduction in receptor area vs. an increase in the penetration of turbulence into the canopy), it is the reduction in the surface area to which particles deposit that dominates the induced change in particle deposition. However, more research—both experimental and numerical—needs to focus on assessing the magnitude and spatial extent of edge effects and landscape heterogeneity in determining particle fluxes.

(2) Relatively little is known about the partitioning of particle deposition between vegetation and the underlying ground, and the profile of the particle concentration gradient and flux through vegetated canopies. It is likely that the contribution of the ground to the total flux can be considerable depending on the arrangement of tree crowns and number density (Donat and Ruck, 1999) or on the vegetation type under consideration (Little, 1977). Further knowledge of this distribution would greatly benefit both process-level understanding and scientists focussing on the ecosystem impact of nutrient fluxes.

(3) We echo the assertions of Gallagher et al. (2002) that progress in understanding particle exchange is 'hampered by the lack of a consistent methodological approach for interpreting measurements from very different techniques' and would extend this to consideration of and quantification of micrometeorological flux methods as applied to atmospheric particles. While some of this research is ongoing (Pryor et al., 2007a,b) and can certainly benefit from flux methodology homogenization efforts being conducted under Fluxnet (Aubinet et al., 2000), more is required to generate specific understanding of the sources of uncertainty and methods for developing robust uncertainty estimates such that there can be greater comparability of disparate studies and improve understanding of the sources of model vs. measurement discrepancies. Further, there needs to be much greater transparency with respect to how individual researchers treat their data.

(4) While the majority of studies over vegetated canopies have focused on dry deposition—that is, the atmosphere–surface exchange being directed towards the vegetated surface—particle resuspension (Nicholson, 1988b; Ould-Dada and Baghini, 2001) has long been recognized as a mechanism by which vegetated surfaces can act as sources of particles. Few studies have applied the eddy covariance technique to particle resuspension (Nemitz et al., 2002a). However, it is likely that detailed useful information on re-suspension and saltation processes might be derived using modern instrumentation adaptable to the micrometeorological techniques discussed in this review. Such information may prove extremely useful for those who advocate use of trees for filtering of urban air (Ould-Dada and Baghini, 2001). There is also increasing evidence for particle formation at or close to canopies and hence of bi-directionality of the particle flux as a result (Buzorius et al., 1998; Pryor et al., 2007a). Improved understanding of such processes requires cohesive field experiments with simultaneous measurements of micrometeorological parameters, as well as particle and gas concentrations and fluxes.

(5) Despite the importance of determining size-resolved particle flux (and deposition velocity) magnitudes, few studies have sought to quantify the size dependence of submicron diameter particle v_d. Such measurements are necessary to evaluate particle deposition models. Assuming uncertainties due to low particle counts can be overcome, development of techniques for differentiating both the size and composition of the particles being transferred [e.g. applications of Aerosol Time-of-Flight Mass Spectrometers (ATOFMS)] will also greatly benefit process-level understanding, atmospheric chemistry models, and ecosystem modellers who seek to determine the fate and effects of dry deposited particles.

(6) Observations over high-roughness vegetated surfaces (forests) generally do not support a pronounced minimum of v_d for particles in the diameter range 0.1–1 μm manifest in models. The Slinn (1982) model employs semi-analytical descriptions of particle collection efficiencies against which all subsequent field observations have been compared which originally came from the wind tunnel studies by Chamberlain in the late 1950s and 1960s. Although significant challenges confront those who seek to undertake wind tunnel measurements (Ould-Dada, 2002), new wind tunnel studies that make use of the latest advances in particle instrumentation may provide valuable insights. If these studies can be conducted under controlled stability conditions they could greatly benefit the search for improved estimates of collection efficiencies and stability corrections for application to particle fluxes. Other processes that may contribute to deposition efficiency in the natural environment (but not in wind tunnels), including electro- and thermophoresis, remain largely unstudied but also merit additional attention.

(7) There is still much we do not understand concerning particle interaction with biological surfaces. Biological surfaces actively exchange mass with the atmosphere and additionally exhibit complex micromorphology which may enhance deposition of non-spherical particles. A macrometeorological perspective is likely required for development of model parametrizations of particle fluxes suitable for inclusion in atmospheric-chemistry-transport models. However, microscale analyses may be necessary for understanding biophysical aspects and consequences of particle and hydrometeor deposition (Jagels, 1991) and, possibly, in explaining variations in flux magnitudes across superficially similar land cover types (Bache, 1979a; Davidson et al., 1982). Physical entrapment by structural features of leaf surfaces are implicitly incorporated within deposition models such as that of Slinn (1982) via use of characteristic foliage length scales. However, the precise mechanisms of interaction between particles and leaf surfaces remain somewhat elusive (Hosker and Lindberg, 1982). Preferential deposition of 500 nm diameter particles to and around stomata of coniferous needles due to enhanced microroughness of the epicuticular waxes was observed in a wind tunnel study (Burkhardt et al., 1995) and may greatly affect the ultimate fate of transported chemicals and biological response (Jagels, 1991) such as via regulation of water exchange (Burkhardt et al., 1995). However, few mechanistic studies have been conducted on the influence of microscopic roughness and other leaf/surface properties in regulating/mediating particle deposition and the consequences of these interactions for atmospheric studies remain uncertain. Integration of microscale 'leaf-level' analyses with atmospheric flux experiments may yield critical insights into both the ultimate fate of deposited particles and the importance of leaf surface properties and physiology

in dictating atmosphere–surface exchange. These aspects might also be successfully investigated with more wind tunnel studies making use of modern particle measurement techniques.

(8) Small particles are transported by eddies in a manner similar to gas molecules, but large particles are not. Challenges thus remain before we can have confidence in treating the motion of heavy particles (Wilson and Sawford, 1996) or particles undergoing physical transformation processes as well as in reliably estimating particle flux footprints. We are unaware of any current research pertaining to footprints (source areas) of large particles, although there are numerous articles on particle dispersion. Footprints may also be sensitive to coagulation, formation and phase transition processes and hence these processes should be implemented in Lagrangian and closure models or large-eddy simulations.

As a final note we would urge that there must be integration of experimental and numerical research to facilitate operationalization of insights gained in the experimental domain.

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8. Appendix: Nomenclature

$A(z)$ = absorption function at height z

A_1 = characteristic width of 'small' collectors in the canopy

A_2 = characteristic width of 'large' collectors in the canopy

b = Businger coefficient determined by the probability distribution of w, the dead-band width and sampling height (Ammann and Meixner, 2002)

c = a constant ($\approx$1)

$C(z)$ = particle concentration at height z (e.g. C(0) = concentration at $z = 0$), also abbreviated as C

C_c = Cunningham slip correction factor

C_D = overall canopy drag coefficient

C_{up} and C_{down} = average concentrations from REA samples collected when w is positive and negative, respectively

c_v = viscous drag

c_d = total drag

c_v/c_d = ratio of viscous to total drag [0.33, (Slinn, 1982)]

d = displacement height

D = molecular diffusivity of C in air

D_p = particle diameter

E_B = efficiency of collection by Brownian motion

E_{IM} = efficiency of collection by impaction

E_{IN} = efficiency of collection by interception

$F(z)$ = flux at height z (also abbreviated as F)

$\bar{F}$ = fraction of total particle interception by 'small' collectors in the canopy

g = gravity

$\frac{\delta H}{H}$ = the relative accuracy with which the sensible heat flux can be determined

k = Boltzmann constant

$K(z)$ = eddy diffusivity at a given height z, also abbreviated to K, where a subscript denotes the parameter under consideration [particles (K_p), momentum (K_m), or heat (K_H)]

l = characteristic eddy length in the canopy

L = Monin-Obukhov length

m_p = particle mass

N_A = Avogadros constant

P = atmospheric pressure

R = particle rebound

$r(z)$ = total resistance at height z

$r_a(z)$ = aerodynamic resistance to transport at height z, also abbreviated as r_a

r_b = resistance to transfer across the quasi-laminar surface layer

r_c = resistance to surface uptake

R_g = gas constant

S = source (positive) or sink (negative) of C

$S()$ = power-spectra (as a function of frequency)

Sc = Schmidt number (ν/D)

St = Stokes number

T_a = air temperature

T = averaging period

t_0 = start time

U and u = horizontal wind speed (U_h, is u at h, where h is the canopy height). U_r is wind speed at a reference height, r

U_0 = wind velocity away from obstacles

u_i = wind speed in 3 directions (u, v, w)

u_* = friction velocity

$v_d(z)$ = deposition velocity at a given height z, also abbreviated as v_d

v_g = settling velocity

v_t = transfer velocity

w = vertical wind speed

x_i = the 3 directions used to define the wind speed components (x, y, z)

z = height

z_i = inversion base height

z_0 = roughness length for momentum.

α and β = Kolmogorov constants describing the intensity of the inertial range spectra for velocity spectra and all other scalar spectra respectively

α_c = surface area of vegetation per unit volume

δF = standard deviation (used here as a measure of uncertainty) on the ensemble flux statistics

$\delta\theta$ = uncertainty of the temperature measurements

δC = uncertainty on the measurements of C_{up} and C_{down}

ΔX (where X is C or θ) = $\bar{X}_{\text{up}} - \bar{X}_{\text{down}}$

ε = collection efficiency R

ϕ_ε and ϕ_{NC} = normalized dissipation functions [f thermal stability)]

γ = parameter characterizing the wind profile through the forest

λ = mean molecular free path

κ = von Karman constant

μ = dynamic viscosity of air

ν = kinematic viscosity of air

ρ = air density

ρ_{p} = particle density

σ = standard deviation

σ_w = standard deviation of vertical velocity, w

τ = particle relaxation time

τ_m = momentum flux

$\Im_x$ = averaging time for flux estimation

Overbar is used to denote time averages.

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