1 Introduction

This thesis contains a broad investigation of several aspects of the interaction between aerosol and various surfaces. The diversity of material covered necessitates an early separation into distinct themes. These are addressed systematically in this chapter and throughout the rest of the thesis. Briefly, the separate areas of work included:

- Construction of an eddy covariance aerosol flux measurement system, capable of measuring fine mode aerosol fluxes using a modified, commercially available particle counter.

- Use of the eddy covariance system to measure fine mode aerosol deposition velocities to a variable length semi-natural canopy. Accumulation mode fluxes were also measured with a separate system.

- Use of the system to investigate the interaction between ammonia and the aerosol phase following fertilisation of a managed grass canopy.

- Measurements of fine mode aerosol fluxes above a city.

- Various modelling as an aid to interpretation of the above measurements.

While the modelling is integrated with the results, it has been necessary to treat the measurements separately from each other. An additional chapter of introduction to some common micrometeorological concepts is also included. Accordingly, this chapter begins with a short general outline of aerosol exchange, and goes on to give background for the sections on aerosol growth and urban aerosol. Each section here is brief, as the background for the individual topics is covered in more depth as they are presented.

1.1 Aerosol Exchange

1.1.1 Deposition

Knowledge of the rate of aerosol exchange with surfaces is important for a variety of reasons. Aerosol emission and deposition rates are important in determining both atmospheric aerosol concentration and pollutant loadings to ecosystems. For example, in the EMEP framework, and many other such models, the deposition flux is calculated as the product of air concentration and an empirically determined deposition velocity (chapter two). Chapter four of this thesis is devoted to measurements of deposition velocity and comparison with a mechanistic deposition model, as it is this type of measurement that is used to parameterise aerosol exchange in regional scale models.

Much work on aerosol deposition was conducted in response to concerns over acid deposition and its effects, especially on forest ecosystems (*e.g.* Wyers and Duyzer, 1997). However, as sulphur emissions have declined, most notably in Western Europe, attention has shifted to other effects of aerosol deposition. One of these is possible eutrophication of nitrogen sensitive ecosystems (*e.g.* Sutton *et. al.*, 1997), possibly by long-range ammonia transport associated with aerosol. Although the likely application of results from regional transport models has changed subtly, the difficulty in adequately representing aerosol in such models remains. The largest uncertainty in the use of models such as EMEP for predicting aerosol loadings is the parameterisation of deposition velocity for different surfaces under different meteorological conditions (Hummelshøj, 1993). Although EMEP currently runs on a 50 km × 50 km grid, it is still necessary to properly parameterise aerosol deposition in order to have confidence in the predicted deposition loadings and air concentration depletions.

1.1.2 Micrometeorological Deposition Measurements

The earliest widely reported eddy covariance (chapter two) measurements of aerosol deposition velocity were made by Wesely *et. al.* (1977) using an electrical particle counter with a response time of around 0.8 s. These measurements were made 5 m above scrub grass land, and resulted in deposition velocity estimates of the order of 1.0 – 10.0 mm s^{-1}. Further measurements began to be reported in the literature from 1982 onwards, *e.g.* Sievering (1982; although this was a gradient measurement using optical counters), Neumann and Den Hartog (1985), Katen and Hubbe (1985) and Sievering (1988). Gallagher et. al. (1997 a) give a comprehensive review of these measurements up to around 1995, along with a review of the performance of mechanistic aerosol deposition models.

Since the above review, several deposition velocity measurements have been published. Eddy covariance and gradient measurements above a coniferous forest canopy were presented by Gallagher *et. al.*, (1997 b) and Wyers and Duyzer (1997) along with a modelling effort by Ruijgrok *et. al.* (1997) and a summary papers from Erisman *et. al.* (1997 a, b) as part of the Netherlands "Aerosol Project". Results from this and more recent projects are presented in chapter four.

A recent development in aerosol deposition measurements is the use of condensation particle counters (chapter 3) to measure fine mode aerosol fluxes (down to around 10 nm in diameter). Several of these measurements are shown in chapter four along with new results for fine mode aerosol deposition to a grass canopy.

1.2 Ammonia – Aerosol Interactions

Measurements of ammonia induced fine aerosol growth are introduced in chapter five. These are the first such direct measurements of aerosol growth above an anthropogenically modified canopy. Other similar work includes the BIOFOR project, which aimed to examine biogenic aerosol formation above forests (*e.g.* Rooke, 2001; this reference is the foreword to the Tellus BIOFOR special issue). Some of the modelling methods used in chapter five were derived from this work. Some work has also been published recently on particle formation in the coastal environment (*e.g.* O'Dowd, 2001), but the focus of this work is rather different, – in the coastal environment it is thought that particle formation is more important than growth of existing particles.

Previous investigations of aerosol – ammonia interactions have been primarily based upon measurements of acid gas and ammonia concentrations (*e.g.* Nemitz *et. al.*, 2000), and have been aimed at verifying ammonia gradient measurements rather than investigating the effects of ammonia emission on the aerosol population.

1.3 Urban aerosol

The fine mode aerosol flux system has been deployed above a city, measuring urban aerosol emission. A thorough introduction to the local effects of urban aerosol is given at the start of chapter six. However, there have been no previous measurements of aerosol emission above a city, so the chapter is necessarily rather independent of the existing literature. The remainder of the required background to this chapter is given in chapter two.