8 Conclusions and Further Work

Novel measurements of aspects of surface atmosphere exchange of aerosol have been made, and presented in this thesis. Data were gathered using a custom designed and built eddy covariance system based around a modified commercially available particle counter. The performance of the particle counter has been shown to be adequate for performing flux measurements under the conditions described in chapters four and six (semi-rural and urban sites). Models have been developed and examined describing several aspects of the work.

A model describing aerosol deposition has been implemented from the literature and tested against measurements. Another model describing the growth of aerosol in the presence of high concentrations of condensable vapour has been presented and used to interpret apparent emission fluxes of fine aerosol. Finally, parameterisations of the behaviour of urban aerosol have been developed, with a discussion of the transport of urban aerosol.

These measurements and models represent several of the processes in the "life cycle" of aerosol, from production and chemical processing to deposition. Although there are no current legislative applications for the work, it appears likely that as aerosol critical levels become more tightly controlled under EU law, attention may begin to focus on smaller diameter ranges, or even a particle number based metric (discussion in chapter six).

8.1 Summary

The work presented in this thesis can most conveniently be subdivided into two parts; – aerosol fluxes above a semi-natural surface and above an urban surface. Although the measurement and initial analysis techniques used in both investigations are broadly similar, it is more coherent to treat the findings separately here.

8.1.1 Aerosol Deposition to Grassland

Aerosol flux measurements have been made above a managed grass canopy of variable height. The analysis showed that accumulation mode (0.1 μm < D_p < 0.2 μm) aerosol deposition velocities were 1.13 mm s^{-1} and 0.87 mm s^{-1} to long and short grass canopies respectively. The corresponding fine mode deposition velocities were 0.275 mm s^{-1} and 0.066 mm s^{-1}, where the long canopy height was 0.65 m – 1.0 m with a roughness length of 0.063 m and the short canopy height was 0.07 m – 0.14 m with a roughness length of 0.022 m. These fine mode fluxes were shown to be representative of aerosol in the diameter range 11nm < D_p < 100 nm, and were measured using a custom made eddy covariance system built around a commercially available condensation particle counter.

Fine mode deposition velocities were significantly lower than the values for the accumulation mode. These values agreed well with the predictions of a "Slinn type" aerosol deposition model, and given the low inter-model variability in deposition velocities in the fine mode, it appears that model performance is adequate in this size range for the managed grassland surface investigated here.

The accumulation mode deposition velocity results fit well with a roughness length dependence derived from the literature, however agreement with the predictions of the deposition model was found to be poor. This was attributed to uncertainty in the deposition model parameterisations, and the need for further model development was noted.

8.1.2 Ammonia – Aerosol Interactions

Measurements have been conducted above a short (0.07 < h_c < 0.14 m) grass canopy immediately following application of 100 kg N ha^{-1} of fertiliser as Ca NH_4NO_3. The measurements were made using the same custom-built eddy covariance system as the above deposition measurements. A model was developed relating apparent particle emissions to the condensational aerosol growth rate across the lower cut-off diameter of the condensation particle counter used.

Fertilisation of the grassland was shown to cause aerosol growth, the postulated cause being simultaneous condensation of ammonia and nitric acid onto the surface of existing particles. Diameter growth rates at 11 nm were calculated, using the above model, to range between zero and around 3490 nm hr^{-1}, with growth rates at 200 nm fixed at 22.7 times lower than those at 11 nm. Systematic aerosol growth was shown to persist for around five days following fertilisation of the canopy. The calculated effect of this condensational growth on the aerosol size distribution was demonstrated, with the largest effect being observed in the fine mode.

Evidence was presented suggesting that the presence of the canopy may be an important factor in the growth of aerosol, with the time scale of turbulence being large within the canopy (low eddy diffusivity), allowing time for growth before rapid mixing of the enlarged particles to the measurement height.

8.1.3 Urban Aerosol Production

The modified particle counter / eddy covariance measurement system was deployed above the City of Edinburgh in the UK. Measured values of aerosol concentration, number flux and emission velocity for a city were presented for the D_3 aerosol diameter range (11 nm < Dp < 3 μm), and the results shown to be representative of the sub-100 nm size interval. The influence of land use type on these quantities was discussed. Aerosol (D_3) concentrations were found typically to range between 3,000 cm^{-3} and 20,000 cm^{-3} although larger values were observed. The mean D_3 aerosol flux over three measurement campaigns was found to be 42,500 cm^{-2} s^{-1}, with the mode being 30,000 cm^{-2} s^{-1} and a typical range of 9,000 cm^{-2} s^{-1} to 90,000 cm^{-2} s^{-1}. The mean and mode emission velocities were 45 mm s^{-1} and 35 mm s^{-1} respectively, and these typically had a range of 20 mm s^{-1} to 75 mm s^{-1}.

The effect of traffic activity, one of the most important urban aerosol sources, on the measured D_3 flux was been confirmed. The correlation between traffic flow and aerosol fluxes above the study city was parameterised, along with the specific dependence of the emission velocity on atmospheric stability. Similar results from all

three independent data sets (SASUA I, II and III) showed a reduction in emission velocity with increasing instability. An explanation for this was discussed.

The above parameterisations were combined into a single, simple model describing D_3 aerosol concentration in terms of traffic activity and anthropogenic activity. The agreement of this model with measurements showed that traffic activity and boundary layer stability are two of the most important determinants of urban D_3 aerosol concentration.

8.1.4 Urban Micrometeorology

The energy balance for an urban area was examined, with special reference to anthropogenic energy input. It has been shown that the non-radiative heating of the urban surface examined is equivalent to the addition of 44 W m^{-2} to the system. Of this additional energy, which is assumed to be anthropogenic in origin, it has been estimated that around 30% is derived from fossil fuel combustion within the city. Possible sources for the remaining 60% include space heating and other electrically powered activities.

The spectral and co-spectral behaviour of sensible heat and aerosol transport have been discussed. The stability dependence of the bi-directionality of sensible heat fluxes has been noted. Similarly, there is a stability dependence for downward aerosol flux, with strong low frequency downward transport of aerosol observed during unstable conditions.

The analysis of the probability density functions for vertical wind speed with virtual temperature and aerosol concentration shows evidence of complicating factors in the aerosol flux not present in the heat fluxes. A comparison of the probability density and co-spectral analyses for aerosol flux is suggestive of downward transport of aerosol from aloft competing with upward transport from urban surface sources. This interpretation is tentatively backed by an analysis in chapter six of the directionality of aerosol flux. The hypothesis advanced in the two chapters on urban aerosol is that a layer of elevated fine aerosol concentration exists near the top of the urban boundary

layer during the day, and that aerosol can be transported downward from this layer under unstable conditions.

8.2 Further Work

8.2.1 Aerosol Fluxes Over Semi-natural Surfaces

Two interlinked areas requiring further work have been identified with regard to aerosol fluxes over natural and semi-natural surfaces. The canopy processes governing deposition velocity need to be improved in order to improve existing micro-scale models so that more reliable estimates can be introduced into larger scale models (*e.g.* EMEP *etc.*). Also, canopy processes can modify aerosol populations under certain circumstances. This requires further attention both because modification of an aerosol size distribution results in a change in (the diameter dependent) deposition velocity, and because of the possible implications for boundary layer chemistry and long-range transport of trace pollutants such as ammonia and acid gases.

As noted, further work is required on the parameterisations of accumulation mode deposition processes in order to reduce the variability and apparent error in model predictions of deposition velocity. Although there is an established and growing database of aerosol deposition measurements to various surface types, further measurements will be useful in resolving the contributions made to deposition by the five identified concurrent processes (chapter four) over different canopy types under field conditions.

Further laboratory measurements of these individual processes (where possible) may also have a role to play in model development. Systematic re-analysis of existing measurement datasets could also help to identify both the major aerodynamic and canopy deposition processes affecting aerosol fluxes. It is clear that the chemical state of the boundary layer is extremely important in determining measured fluxes under certain circumstances (chapter five), and future investigations should be planned with a view to including complimentary (chemical) measurements capable of quantifying

this effect where appropriate. Such measurements will also provide insights into the rate, chemistry, microphysics and environmental effects of "surface modification induced" aerosol growth.

8.2.2 Urban Aerosol

The work presented here on urban aerosol leads to the identification of at least two key areas for further research. The parameterisations of aerosol flux, emission velocity and concentration presented in chapter six are currently only directly applicable to the City of Edinburgh, UK. Apart from the need for further measurements at the Edinburgh site for model validation, similar measurements in several more cities will be required to *begin* to generalise the results to other urban surfaces. It is worth noting again the difficulty in finding a widely applicable measure of aerosol producing anthropogenic activities. Although traffic activity has been used here, it will be necessary to modify the constants in the flux parameterisation to take account of different vehicle fleets and land use distributions. Indeed, it may be necessary to introduce further variables such as local fossil fuel sales in order to help scale the aerosol emissions to different cities. Only further measurements in several different locations will finally resolve these questions, although examination of emissions inventories may prove to be a useful additional exercise.

There is further scope for improvement in the estimate of urban emission velocity, and the effects of different topography and climate. It appears likely that the most reliable method for achieving these improvements will also be to make further measurements above at least several different cities. As for the parameterisation of aerosol production, any such measurements should be made over appreciable periods at all times of year in order to resolve the annual variation in urban micrometeorology and in aerosol production.

Apart from the need to improve the aerosol concentration model set out in chapter six, this work has given rise to important fundamental questions about the mechanisms of urban aerosol transport and the location of aerosol production. The simplest method of addressing these questions would be to make co-located and co-incident airbourne

measurements of boundary layer and turbulence structure, and fine aerosol concentration profiles along with any further urban eddy covariance measurements.

Finally, with a validated and reliable model for predicting aerosol concentration using commonly measured quantities, it may be possible to construct aerosol concentration time series several years into the past. This would be of considerable benefit to the epidemiological community, who in the past have relied upon PM10 concentrations as a measure of the exposure of urban populations to aerosol. As stated in chapter six, the view appears to be becoming increasingly widely held among toxicologists that fine and accumulation mode aerosol are likely to be the size fractions most damaging to human health.