Exploring the interactions between atmospheric ions and micrometre sized aerosols

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Abstract. The interaction of atmospheric ions and micrometre diameter aerosols has been explored using two different numerical modelling techniques. Firstly, a simple box-model has been used to study the dynamics of ion-aerosol interactions and the sensitivity to a number of parameters. Secondly, a coupled dispersion and ion-aerosol interaction model has been used to study the evolution of a charged aerosol released from a source. This more complex model includes the effect of the dilution of the released aerosol which can be a key parameter controlling the rate of change of the aerosol charge distribution.

1. Introduction
Attention has focused in the past on the interaction of atmospheric ions and particles with diameters in the sub-micrometre range because of their importance in climatic processes [1]. However, larger aerosols have the ability to carry greater electrostatic charges and also interact with atmospheric ions. Whilst previous work has investigated the interaction between corona ions from high voltage power lines and atmospheric aerosols [2, 3], and also on the charging of radioactive aerosols [4], this work explores the interaction of micrometre sized aerosols and atmospheric ions. In particular, this work is focused on the study of the decay of aerosol charge from anthropogenic releases as they interact with atmospheric ions.

Two modelling approaches have been used in this work. The first, a well-mixed box-model, was used to study the dynamics of ion-aerosol interactions. The model was used to explore the sensitivity to a number of the system parameters. The second approach includes the effect of dispersion on an aerosol source, combined with ion-aerosol interactions. The model performance was compared with experimental measurements of atmospheric ion concentrations and used to predict the combined dispersion and charge evolution from an aerosol source.

2. Ion-aerosol dynamics box-model
2.1. Introduction
A box-model was created which tracked the concentration of positive and negative ions and the components of a charged aerosol distribution. Aerosols of 1 micrometre diameter can potentially carry charges of up to $10^4$-$10^6$ elementary charges depending on the composition of the aerosol and the polarity of the charge [5]. To describe a population of aerosols a probability distribution of charges can be used. In the box-model a discrete distribution was used, which tracked the concentrations of aerosols with a given number of charges.
2.2. Model implementation

The rate of change in ion concentration for positive or negative ions was calculated using equation (1), following Harrison and Carslaw [1].

\[
\frac{dn_\pm}{dt} = q - \alpha n_\pm - \sum_{j=-\infty}^{\infty} \beta_{\pm,j} n_\pm Z_j
\]

Where \( n_\pm \) is the concentration of positive or negative ions, \( q \) is the rate of production of ions, \( \alpha \) is the rate of recombination of positive and negative ions, \( \beta_{\pm,j} \) is the rate of ion-aerosol reattachment for a positive or negative ion reacting with an aerosol of charge \( j \) electrons and \( Z_j \) is the aerosol concentration for aerosols with a charge of \( j \) electrons. A mono-disperse aerosol was assumed and only combination with singly charged ions was considered.

The rate of ion-aerosol attachment was calculated using equation (2) after Mayya and Sapra [6] and Gunn [7] for the continuum regime.

\[
\beta_{\pm,j} = \frac{je\mu_\pm}{\varepsilon_0 [\pm \exp(\pm 2\lambda j)]^{\frac{1}{2}}} \]

Where \( \lambda = e^2/8\pi\varepsilon_0 akT \), \( \mu_\pm \) is the electrical mobility of the positive and negative ions respectively, \( e \) is the charge on an electron, \( \varepsilon_0 \) is the permittivity of free space, \( a \) is the aerosol radius, \( k \) is the Boltzmann constant, and \( T \) is the temperature.

The rate of change in aerosol concentration was calculated using equation (3) after Hoppel and Frick [8].

\[
\frac{dZ_j}{dt} = n_+ \beta_{+,j-1} Z_{j-1} - n_+ \beta_{+,j} Z_j + n_- \beta_{-,j+1} Z_{j+1} - n_- \beta_{-,j} Z_j
\]

Values for \( q \), \( \alpha \), \( \mu_\pm \), and \( n_\pm \) were taken from [1]. Limits for the maximum and minimum number of charges carried by the aerosol were typically set to values of +300 and -300 respectively.

A computer model was implemented using the scripting language Perl with graphical output created using Perl/Tk. A simple Euler scheme was used to calculate successive ion and aerosol concentrations from the starting conditions. The resulting ion and aerosol concentrations were written to a file at each time step. For the purposes of ease of interpretation of the data, the aerosol charge distribution was displayed graphically. At each time step, the concentration of aerosol with each discrete charge was drawn as a pixel, and its colour modulated by its concentration. High concentrations were expressed as red, with lower concentrations as blue and zero as black. The charge distribution at each time step was displayed as a horizontal line, with negative charges to the left and positive charges to the right. Progressive time steps were drawn below the last, producing a picture of the charge distribution evolution over time. An example of the graphical output of the model is shown in Figure 1 below.
2.3. Box-model results

The model was used to explore a number of parameters that influence the change in aerosol and ion concentrations. The standard deviation and mean of the aerosol charge distribution were calculated for each time step. Figure 2 shows the change in the standard deviation of the aerosol charge for five different aerosol concentrations for an aerosol of 1 µm diameter. It can be seen that for the lower concentrations, \(1 \times 10^9 \text{m}^{-3}\) and \(1 \times 10^8 \text{m}^{-3}\), there is little difference in the decay of the charge distribution. However, at higher concentrations there is a decreasing rate of decay. This is to be expected from equation (1), since higher \(Z\) results in reduced ion concentrations, slowing the rate of neutralisation. The ion concentration therefore becomes a limiting factor in these cases.
The mean of the aerosol charge distribution for the same models showed a small, increasing negative bias in all cases due to the higher mobility assumed for the negative ions [1]. The mean values for the higher concentrations were still changing at the end of the 30 minute simulation.

3. Coupled fluid flow and ion-aerosol dynamics model

3.1. Introduction
In an external environment, the decay of an aerosol concentration and charge distribution will be influenced by the effects of dilution due to turbulent mixing, in addition to the ion-aerosol dynamics. To capture both processes a coupled fluid flow and ion-aerosol model has been developed and used to study an example case of the dispersion of a charged aerosol.

3.2. Model implementation
A commercial computational fluid dynamics (CFD) solver, Fluent ® v6.2.16, was used to model the dispersion from an aerosol source. The fluid flow treatment was supplemented by the tracking of a discrete distribution for the aerosol charge distribution. A reaction scheme was implemented to include the ion-aerosol attachment coefficients for each of the tracked elements of the aerosol charge distribution. Ionisation and ion-ion recombination were also included. As for the box-model, the aerosol was assumed to be mono-disperse.

The use of a discrete distribution for the aerosol charge distribution required the tracking of a scalar for each aerosol charge combination. For large standard deviations of charge, this required the tracking of a very large number of scalars. The computational requirements become unmanageable for a large number of scalars and the CFD solver has an absolute limit on the number of scalars that can be used. To model wider distributions, the individual concentrations of aerosol-charge combinations were combined into a series of bins. The concentrations in these bins were then tracked and the reaction dynamics were adjusted accordingly.

The model was tested against ion concentration measurements from a wind tunnel trial of a test aerosol release. Experimental measurements of positive and negative ion concentrations were made at 40m downwind from the source and compared with modelled values. The modelled percentage decreases in ion concentrations fitted well within the range observed in the experimental data.

An example scenario of external dispersion of an aerosol source was studied and results are presented below. The model domain was 2.1 x 0.5 x 0.1 km (L:W:H). A logarithmic wind profile with a velocity of 5m s⁻¹ at 10m above the ground was assumed at the inlet. The roughness length for the velocity profile was assumed to be 0.01m. The RNG k-ε turbulence model was used to model the turbulence statistics.

3.3. Coupled fluid flow and ion-dynamics model results
Figure 3 shows an example set of results from the coupled model. The graph shows the concentration of aerosol and the standard deviation of the aerosol charge distribution plotted along the plume centreline. For comparison the standard deviation for the Boltzmann equilibrium charge distribution for the same diameter aerosol is also shown in Figure 4. These results show that the dispersion of the aerosol by the atmospheric flow happens more rapidly than the decay of the charge distribution.
Figure 3. Decay of aerosol concentration and standard deviation of aerosol charge distribution with distance from source for a mono-disperse aerosol of 2µm diameter. The graph is plotted along plume centreline. The standard deviation of the Boltzmann charge distribution is also shown for comparison.

It is also possible with this modelling approach to output data on the shape of the aerosol charge distribution and its spatial variation. Figure 4 shows the aerosol charge distribution evolving with downwind distance, also along the plume centreline. The narrowing of the distribution with increasing distance is clear. It can also be seen that there is a bias to the negative side of the distribution, due to the higher mobility of the negative ions.

Figure 4. Plot showing the aerosol charge distribution and its change with downwind distance. The aerosol distribution is tracked with a bin size of 25 elementary charges.
4. Conclusions
The interaction of ion and micrometre sized aerosols has been studied using two different modelling approaches. A box-model capable of tracking the dynamics of ion-aerosol interactions has been developed and used to explore the sensitivity of aerosol charge decay to a number of parameters. The use of visual output allowed the dynamics to be quickly assessed. The total aerosol concentration was seen to be an important variable above a threshold of $1 \times 10^7 \text{m}^{-3}$ under these conditions.

A coupled fluid flow and ion-dynamics model was used to explore the interaction of dispersion and ion-aerosol dynamics. A commercial computational fluid dynamics code was adapted to track the evolution of the aerosol charge distribution and atmospheric ion concentrations. For an example case, the dilution of the aerosol concentration was more rapid than the decay of the aerosol charge.

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