Introduction

Recent observations of ultrafine aerosol in the atmosphere support model studies assuming ions as major nucleation agents. We present a model of ion induced aerosol formation based on experimental data in thermodynamic and aerodynamic models, and investigate the response of tropospheric aerosol production to variations in background ionization. We explain why aerosol production may both correlate and anticorrelate with the ionization rate, and show that the sign of the correlation depends critically on the gas phase concentration of sulfuric acid. For typical conditions, our model predicts a positive correlation of aerosol production with ionization throughout the troposphere. Only for very low concentrations of sulfuric acid or very low ionization rates, an anticorrelation can be expected. Our results indicate a significant response of ultrafine aerosol and surface area concentration to variations in ionization, such as resulting from the modulation of the galactic cosmic ray intensity by the 11 year solar cycle. Ion induced aerosol formation may thus play an important role in promoting the solar signal to the troposphere.

Analytical model

Consider a simplified system of neutral (A) and charged (A+) sulfuric acid clusters, each of which forms a sulfuric acid molecule, as shown in Figure 3. The reaction rates are given by:

\[ A + A_+ \rightarrow 2A \]

\[ A + A_+ \rightarrow A_2 + e^- \]

\[ A + A_+ \rightarrow A_2^+ + e^- \]

\[ A + A_+ \rightarrow A_3^+ + e^- \]

\[ \ldots \]

\[ A + A_+ \rightarrow A_n^+ + e^- \]

(1)

where \( n \) is the critical cluster size, \( I_n \) is the ion concentration, and \( \lambda \) is the ionization rate.

The solution for \( [A] \) in the gas phase is:

\[ [A] = \frac{n_{\text{neutral}}}{n_{\text{total}}} \]

(2)

where \( n_{\text{neutral}} \) is the number density of neutral clusters, and \( n_{\text{total}} \) is the total number density of clusters.

We will examine the influence of the number of sulfuric acid molecules in the neutral cluster on the production of supercritical clusters, using two different cluster profiles.

Figure 4 shows the number of sulfuric acid molecules in the neutral cluster as a function of cluster size. The neutral cluster size decreases as the ionization rate increases. A constant neutral cluster size is reached at a critical ionization rate.

For the constant neutral cluster size calculation, we use the rate equations:

\[ \frac{d[A]}{dt} = k_1[A][A_+] - k_2[A][A_+] - k_3[A][A_+] - \ldots - k_n[A][A_+] \]

(3)

where \( k_i \) are the ionization rates for each cluster size. The solution for \( [A] \) is:

\[ [A] = \frac{n_{\text{neutral}}}{n_{\text{total}}} \]

(4)

Discussion

The response of aerosol production to ion induced particle formation depends on several quantities: bulk production rate, sulfuric acid concentration, and initial cluster size. The neutral cluster size decreases as the ionization rate increases, resulting in a negative correlation. However, the aerosol production rate is also influenced by the ionization rate, resulting in a positive correlation. The trend is reversed in the upper troposphere, where the ionization rate is higher, leading to a negative correlation. The aerosol production rate in the lower troposphere is not significantly influenced by the ionization rate.

Figure 6 shows the aerosol production rate as a function of ionization rate. The aerosol production rate increases with ionization rate, reaching a maximum at a critical ionization rate, beyond which the aerosol production rate decreases.

Figure 7 shows the variation of aerosol production rate with ionization rate. The aerosol production rate increases with ionization rate, reaching a maximum at a critical ionization rate, beyond which the aerosol production rate decreases.

References