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AN EXPERIMENTAL ASSESMENT OF CHANGES TO THE SIZE DISTRIBUTION OF AMMONIUM NITRATE UNDER ISOTHERMAL CONDITIONING

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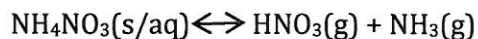
SUMMARY

The volatility of ammonium nitrate was assessed under laboratory conditions in terms of size distribution changes once subjected to isothermal conditioning. Three temperature regimes were chosen to coincide with what is perceived to be representative of normal indoor temperatures, whilst three particle sizes in airtight and accumulation modes were investigated. The size distribution for each regime was then measured before and after transportation through an evaporator and these changes assessed.

Results show that for 50nm ammonium nitrate particles, 70-80% losses were recorded through the evaporation chamber. The least volatile was found to be 200nm particulate at 15°C. Generally, at 25°C the greatest reactions were found as was expected from prior research, however an overall increase in particulate concentration at 100nm for all temperature regimes was surprising. Possible multiple physicochemical transformations during transportation through the evaporator, along with activation of impurities within the water could explain these particle gains. The lack of humidity data, inter- transportation temperature profiling and lateral gas measurements limit the effectiveness of this study.

INTRODUCTION

Secondary inorganic aerosol makes up a sizable fraction of total aerosol loading in the northern hemisphere, with ammonium nitrate (here-forth NH_4NO_3) contributing upto 30% of this (Ansari and Pandis, 1998). NH_4NO_3 is usually found in the accumulation mode of 100-300nm in diameter (Bergin et al., 1997), however they are volatile under normal atmospheric conditions and can deliquesce producing NH_3 and HNO_3 in the following reversible process



The balance of this process is known to be dependent on atmospheric pressure, temperature and relative humidity, factors that often differ from an outdoor to indoor environment. It is this volatility at what is considered normal ambient conditions which makes NH_4NO_3 challenging yet important to understand.

NH_4NO_3 originates from outdoor sources, mixing anthropogenically formed NO and NO_2 , predominantly from combustion sources which then oxidises to form nitric acid, which in the presence of ammonium is neutralised to form ammonium nitrate, (Lightstone *et al*, 2000). NH_4NO_3 is a major constituent of atmospheric aerosol, usually found over continental

landmasses and usually in the fine size range (Bergin et al 1997, lightstone et al, 2000, Nowak *et al*, 2010). Over the past decade there have been many attempts to model NH_4NO_3 over larger continental land masses to help complete emission inventories and to aid understanding of visual and radiative forcing effects (Schaap *et al* 2004, Adams et al., 1999). However awareness of NH_4NO_3 and its gas phase components of nitric acid and ammonia have been largely neglected as a contributing dynamic in indoor air pollution studies.

MEASUREMENTS

Three sizes of ammonium nitrate aerosol were chosen for analysis, 50nm, 100nm, and 200nm. These were then subjected to three temperature profiles 15°C, 20°C, and 25°C as they passed through a 2m evaporator. The reduction of aerosol size is indicative of the rate and degree of dissociation.

The volatility of NH_4NO_3 at 100nm is demonstrated in Figure 1. The dissociation rate is seen to be similar for both 15°C and 20°C where-as for 25°C the rate and extent of the size distribution change is observed to be more significant. Small losses were found between the time the NH_4NO_3 aerosol was created and the first DMA size selector (shown above labeled DFC).

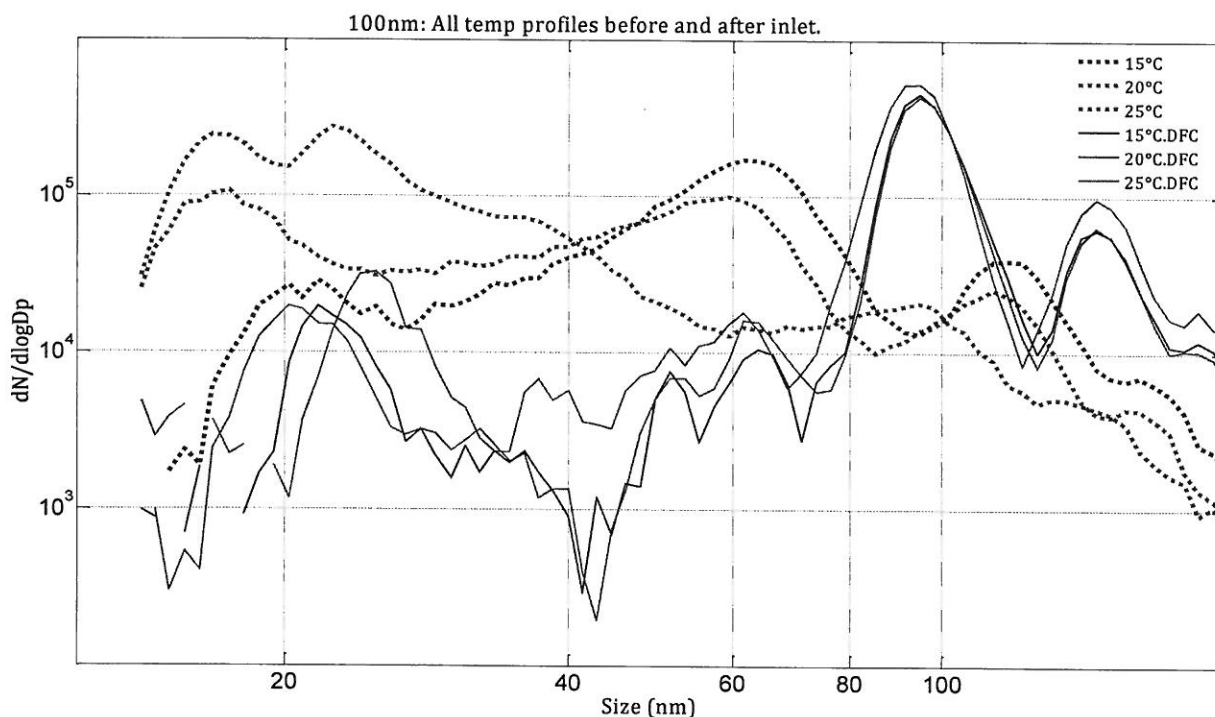


Fig. 1: Log Size distributions for monodisperse ammonium nitrate aerosol of 100nm is shown (hatched line) in relation to the original aerosol size distribution taken directly from the first DMA (DFC). Isothermal temperatures are represented by the red, green and blue lines.

DISCUSSION

The changes in the size regime of NH_4NO_3 are closely aligned to temperature. This is in accordance with prior research (Seinfeld and Pandis 1998, Dassios and Pandis 1999, Lunden et al., 2003). Between 70-80% of 50nm NH_4NO_3 disappeared during transportation through the evaporation chamber. It is probable that due to residence time through the experimental set-up, most of the particles deliquesced into their gaseous precursors. It is theorized by Dassios and Pandis (1999) that NH_4NO_3 particles smaller than 100nm can equilibrate between phases in just a few seconds under "typical" temperatures. This could explain the large losses seen here with transportation rate through the entire sampling set-up estimated to be between 3 and 4 minutes.

NH_4NO_3 volatility is evident once 100nm particles were produced. At 25°C reaction rates were at their greatest with a large transition of size distribution from 93nm measured directly from the first DMA down to 22nm after undergoing transportation through the evaporator.

For 200 nm, the most volatility was again recorded at the highest temperature. The modal peak at 25°C was very similar to that of 20°C however much greater losses occurred at 25°C, whilst at 20°C a net gain in concentration was recorded during transportation through the evaporator. At 15°C the NH_4NO_3 particles contracted by 28nm, thus displaying the least volatility of each size and temperate regime. This indicates that the larger the NH_4NO_3 particle the less prone to volatility it becomes, even at temperatures where deliquescence is expected.

CONCLUSION

Because of its volatility at ambient temperatures NH_4NO_3 is challenging, yet of importance to indoor air research, Particle size changes and transitional changes from solid-liquid-gas phases in a reversible mechanism make NH_4NO_3 and its component species ammonia and nitric acid a dynamic part of overall air mix within the indoor environment, both in terms of particulate residence time in the accumulation mode and the potential material damage caused by nitric acid sorption.

The evident overall reduction in particle sizes demonstrated here indicates either the removal or deliquescence of NH_4NO_3 to its gas phase precursors, a stripping of the nitrate off the particulate, or aqueous phase particulate undergoing evaporation. It is shown here that the smaller the particle and the higher the temperature, the greater the reduction in size and thus indicating increased volatility. However it is wrong to presume that the reaction is greater for the smaller size fractions as it is possible that the larger particles undergo various physiochemical transitions during the transportation through the evaporator.

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