

The authors investigated the effect of sulphur dioxide and ammonia on the secondary organic aerosol formation in the photooxidation of toluene/NO_x with or without Al₂O₃ seed particles. They presented new experimental results and showed that the synergistic effects between SO₂ and NH₃ in the heterogeneous process can greatly enhance the aerosol formation. While the authors presented valuable experimental data, they should provide detailed explanations to support their arguments and observations made in the manuscript.

Comments

One main question is: How much toluene is being reacted under different experimental conditions? This information is not given in the manuscript. If the amount of the toluene reacted is known, what are the aerosol mass yields in these experiments? How do the measured aerosol mass yields compare with literature results, if any?

Line 184, "Assuming the same aerosol density in these experiments, the presence of either NH₃ or SO₂ enhanced secondary aerosol formation markedly" Can the authors provide justification for this assumption?

Line 217, "A significant increase in new particle formation was observed in the presence of SO₂." Can the authors provide an explanation for this observation?

Line 232, "In experiment ATN, the concentrations of ammonium and nitrate decreased after about 2.3 hours of reaction, as shown in Fig. 3, which was consistent with the decreasing trend of particle concentration shown in Fig. 1. The reason for this phenomenon is unknown but we speculate that the generated NH₄NO₃ might partition back into the gas phase as reaction goes on." Can the authors perform any calculations or model simulations to support this hypothesis? What the concentration of the NH₃, SO₂ and NO_x as a function of time in these experiments?

Line 245, "In addition, the presence of organic matter might accelerate the deliquescence of generated inorganic particles (Meyer et al., 2009; Li et al., 2014), and provide moist surfaces for heterogeneous hydrolysis of N₂O₅, contributing to nitrate formation (Pathak et al., 2009)." What is the meaning of "the deliquescence of generated inorganic particles"? Please elaborate. Does the N₂O₅ form under the experimental conditions?

Line 256, "In Fig. 3, the generation of ammonium salt can be observed in the photooxidation of toluene/NO_x/SO₂ without introducing NH₃ gas. This indicated there was NH₃ present in the background air in the chamber, and also indicated that the effects of NH₃ on secondary aerosol formation might be underestimated in this study." As stated, in order to better assess the impact of NH₃ on the aerosol formation, it is important to know the background NH₃ concentration. The authors should give a reasonable guess or estimate on the background NH₃ concentration in their experiments. Could the authors estimate background NH₃ concentration based on their experimental results (e.g. STN data)?

Line 267, “in which the concentrations of NH_3 were more than twice the SO_2 concentrations and the oxidation products of SO_2 and NO_x were fully neutralized by NH_3 .” Any experimental data or calculations to support this argument.

Line 274, “In these experiments, the concentrations of toluene were reduced compared to the experiments in Table 1, and monodisperse Al_2O_3 seed particles with mode diameter about 100 nm were introduced into the chamber.” Any reason why a lower concentration of toluene is used for the Al_2O_3 seed particle experiments.

Line 279, “In the experiments carried out in the presence of Al_2O_3 seed aerosols, the decrease of NH_4NO_3 was not obvious in the experiment carried out in the absence of SO_2 under NH_3 -rich conditions, indicating that generation of NH_4NO_3 was highly dependent on the surface area concentration of the particles, which decreased the partitioning of NH_4NO_3 back to the gas phase, as discussed above”. In addition to describe the results in text, can the authors show the time variation of the chemical species in the SOA generated from the toluene oxidation in the presence of Al_2O_3 seed aerosols to support their argument?

Line 297, “A better correlation was found between secondary aerosol formation and particle surface area than that with particle volume, with details introduced in Fig. S1 in the supporting information, indicating an enhancement effect in the heterogeneous process rather than in bulk reactions”. From the Figure S1, the authors argue that there is a significant correlation between the aerosol formation and aerosol surface area. From that point, they suggest that the heterogeneous processes is important than the bulk reactions. However, a strong conclusion cannot be easily drawn from the simple correlation method and limited data points. Furthermore, a nice correlation between the aerosol formation and aerosol volume is observed. Can the authors further elaborate this point?

Line 330, “According to the consumption of toluene, OH concentrations in the photooxidation experiments were estimated to range from 1.6×10^6 molecules/ cm^3 to 2.7×10^6 molecules/ cm^3 . The reaction between these OH radicals and SO_2 contributed 35%-50% of the total SO_2 degradation in NH_3 -poor experiments, while this ratio was reduced to 25%-30% in NH_3 -rich experiments” Again, what is the amount of toluene and SO_2 reacted in the experiments? What the time variations of the toluene and SO_2 and other gas phase species (e.g. NO_x) in these experiments?

Line 341, “According to previous studies, NH_3 might provide surface Lewis basicity and liquid surface layers for SO_2 absorption and subsequent oxidation, and therefore, enhance sulfate formation (Yang et al., 2016; Tursic et al., 2004).” What is the physical state of the aerosols in the experiments? Aqueous droplets or solid particles?

Line 416, “The higher production of Factor 2 with higher SO_2 under an NH_3 -poor environment could be probably attributed to the well-known acid-catalysis effects of the oxidation product of SO_2 , i.e. sulfuric acid, on heterogeneous aldol condensation (Offenberg et al., 2009; Jang et al., 2002; Gao et al., 2004)” What is the pH or acidity of the aerosols in these experiments?

Line 427 “By inference and from the results of AMS measurements, aerosol water increased as the initial concentration of SO_2 increased, since more inorganic aerosol was generated. Liggio and Li (2013) suggest that dissolution of primary polar gases into a partially aqueous aerosol contributed to the increase of organic mass and oxygen content on neutral and near-neutral seed aerosols, which would also take place in the NH_3 -rich experiments and contribute to the generation of Factor 1.” What is the amount of aerosol phase water and how the aerosol water content change with reactions in these experiments?