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Secondary particle formation from gaseous emissions from livestock buildings

The agricultural animal husbandry is involved in gas emissions such as ammonia, nitrogen oxides and volatile organic compounds. The emitted gases are suspected to be responsible as precursors for the formation of so called secondary particles. In general secondary inorganic aerosols belong to particle sizes of < 0,2 µm. In rural areas secondary aerosols may play a larger role in the total dust concentration, larger than primary particles with a larger diameter do. The aim of a research project is to estimate the importance of secondary particles generated from livestock buildings on the particle matter load in North Rhine-Westphalia and to identify the processes which lead to secondary particle formation. This article should give a summary of the state of knowledge for the terms of formation, identify knowledge gaps and describe design and performance of the research project.

Keywords
Secondary particles, dust emission, ammonia emission

Abstract
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Inorganic secondary particles can be formed in different ways; livestock production is an important source. Their environmental effect encompasses atmosphere, people, animals and plants.

Ammonia (NH_3) is the only basic trace gas present in significant amounts in the atmosphere. In association with acid air components, especially sulphuric acid (H_2SO_4) and nitric acid (HNO_3), airborne ammonia reacts to the salts ammonium sulphate ($(NH_4)_2SO_4$) and ammonium nitrate (NH_4NO_3).

As secondary inorganic aerosols these salts can contribute to a considerable extent to regional dust concentrations. According to measurements [1] up to 40 % of the particulate mass in rural areas can be through this secondary particle formation. The proportion of ammonia in total dust is up to 10 %.

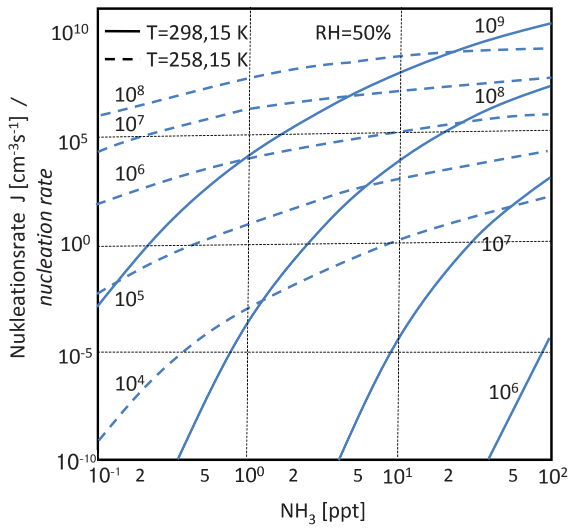
Figure 1 underlines the importance of ammonia for nucleation, in that the nucleation rate increases steeply in line with ammonia concentration [2]. Also apparent here is the temperature dependence of the nucleation rate. The different lines give the sulphuric acid concentrations per cm^3 of air. A typical ammonia concentration of 5–20 µg/m^3 in areas with intensive livestock production [3] calculates out at a concentration of approximately 10 000 ppt. With a medium sulphuric acid concentration in the atmosphere of 10^7/cm^3 (equal to 3.7 ppt) [2] are therefore produced according to **figure 1** at least 10^{11} secondary particles per day and m^3. The products created in the identified chemical reactions form thermodynamic stable clusters (consisting of about 20 molecules) through homogeneous nucleation. Through condensation of further organic and inorganic molecules present in the atmosphere and coagulations (**figure 2**) these clusters then grow into aerosol particles [4].

During the transmission of the particles not only an increase in particle concentration can be observed but also an elimination of particles from the atmosphere. For particles with a diameter smaller than 100 nm this latter action takes place mainly through diffusion onto the ground and through coagulation. For particles with a diameter between 100 nm and 1 µm the moisture deposition (i.e. elution through rain, fog, etc.) is the dominant process. Particles with a diameter greater than 1 µm leave the atmosphere largely through sedimentation.

Up to 90 % of ammonia present comes from agriculture with livestock production responsible for 82 % of this, main sources being cattle (49 %) followed by pigs (22 %) and poultry (7 %). Emissions from the production chain occur mainly from housing (37 %), during storage and spreading of manure (59 %) and whilst stock are grazing (4 %) [5].

The acids involved are mainly formed from precursors produced by burning of fossil fuels in transport, industry and households. The oxidation of sulphur dioxide (SO_2) leads to the formation of H_2SO_4; HNO_3 is formed from the oxidation of nitrogen oxides. Additionally, volatile organic compounds (VOCs) can participate in particle formation. Investigations in a dairy

Fig. 1



Nucleation rate as a function of ammonia mixing ratio [2]

cow barn [6] identified over 70 different VOCs, mainly emitted from silage. In that only VOCs with chain lengths of more then 10 C-atoms have significant influence in aerosol formation, and that the major proportion of VOCs from livestock production units are short chain, it can be assumed that these have only a very limited importance regarding particulate mass [7].

During investigation of secondary aerosols the focus has been mainly on particle number concentration and mass density, chemical composition and optical and aerodynamic characteristics [8].

Their small size means that secondary particles have only a low mass (**figure 3**). It can thus be assumed that the emission burden in the housing exhaust air does not significantly increase and that its mass proportion in the immediate vicinity of the housing is negligibly low. On the other hand, because larger and heavier dust particles sediment-out relatively rapidly, the mass proportion of secondary aerosols in dust during longer distance transport can be markedly higher.

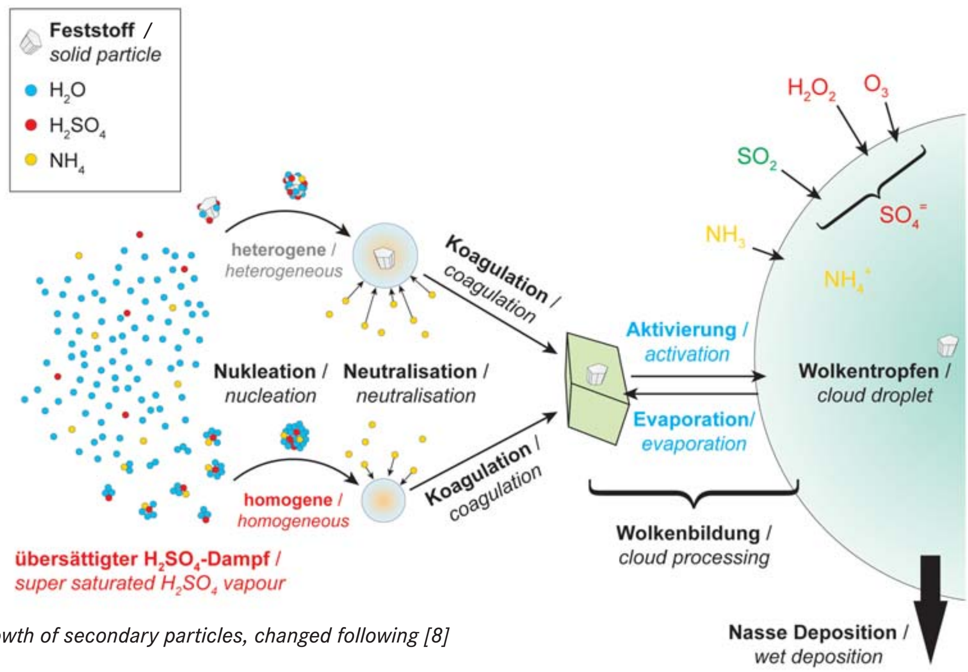
One of the major effects of secondary particles is based on the optical characteristics of aerosol mixes in the atmosphere. Through the ability to reflect and to scatter light, secondary particles are in the position to influence radiation. It can be demonstrated that particles have the short-term ability of scattering solar radiation back into space (indirect effect). However, they also encourage formation of clouds through setting-up condensation nuclei (direct effect) and thus contribute to cooling of the atmosphere of $-2\ W/m^2$ [10]. To a limited degree this process can combat global warming. Chemical reactions on the surfaces of the aerosols in the stratosphere are, however, also under suspicion of being partly to blame for the establishment of the ozone hole [8].

Because of their extremely small particle size, secondary aerosols are able to penetrate very deeply into the lungs of humans and animals. This has led to repeated discussions about possibilities of health damage through long-term exposure. The secondary particles are also blamed for a negative effect on vegetation. If the produced salts land on the surface of plants they then act as desiccators, drawing moisture out of the plant via their hygroscopic characteristics [11]. This effect can have wide-reaching consequences, particularly in regions of low moisture availability.

Project aim

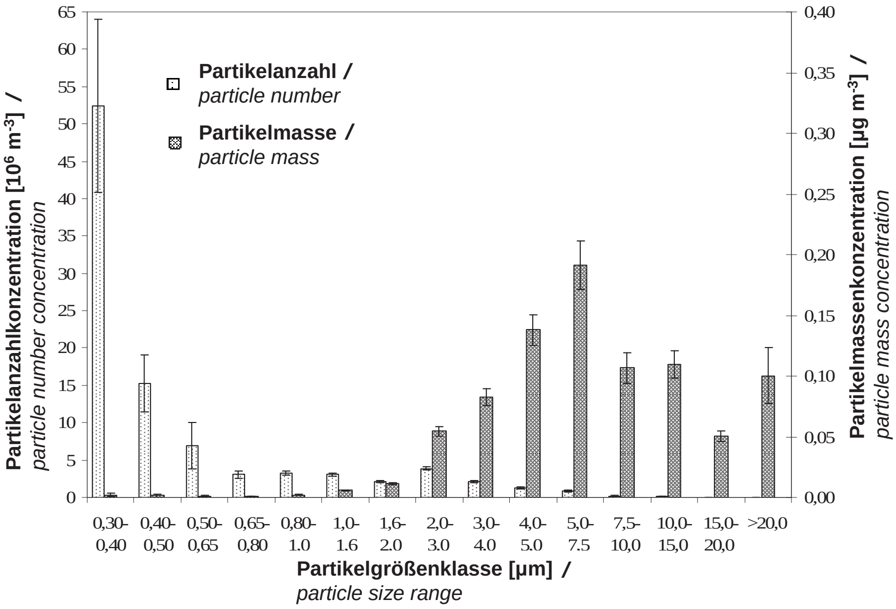
On the basis of the level of knowledge described above the aim of the research project covers the following points:

Fig. 2



Formation and growth of secondary particles, changed following [8]

Fig. 3



Particle number- and particle mass distribution in the outlet air of a deep litter system [9]

- Which processes lead to formation of secondary particles?
- What influences the formation (original gases, UV light, housing ventilation influence)?
- Where do the formation processes take place? In the immediate vicinity of the point of emission or only following increasing distribution and dilution?
- What importance has agricultural livestock production for the general particle load in North Rhine Westphalia?
- Which possibilities for reducing the load might be available?

Materials and method/experiment plan

Through analysis of the literature it became apparent that the level of knowledge concerning NH_3 emission factors in modern naturally ventilated dairy cow housing is particularly limited. There is, therefore, a need for more data in this subject area. The first recordings were made in two dairy cow barns beginning of August 2010.

Focus of the observations were recording parameters air exchange rate, ammonia concentration and mass flow as well as particle mass and size distribution outside the housing. Additionally the meteorological conditions applying (temperature, relative air moisture content, wind velocity and direction) were determined via a weather station. Based on this climate data the effect of the meteorological conditions in the immediate vicinity of the housing on air volume flow should be looked into more closely in further investigations and, with that, also the emission potential for ammonia more precisely defined.

Alongside the described dairy cow housing there should be investigations featuring other production systems and types of livestock. If possible, particle concentration and chemical composition should be measured alongside ammonia recording.

Conclusions

From the detailed study of the current literature regarding formation of inorganic aerosols and their effects on regional particulate pollution it can be concluded that ammonia from livestock production units plays a key role in the formation of secondary particles. The contribution of livestock production to reduction of secondary particle formation should, therefore, take place through reduction of NH_3 emissions. At the same time, reduction of nitrogen oxide and sulphur is necessary for effective and sustainable reduction of secondary particle pollution.

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