

# An emission inventory of livestock-related bioaerosols for Lower Saxony, Germany

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## Abstract

Detailed livestock-related emission inventories are now available for gases but not for bioaerosols, which are emitted in significant amounts and in varying compositions. In view of the environmental importance of bioaerosols, a model for their calculation is proposed here. The basic formula multiplies emission factors by the number of farm animals, but the model is extended by a factor which considers provisionally the influence of production cycles of various types of livestock on the estimated emissions. Despite several uncertainty factors, emissions factors are calculated for dust (inhalable, respirable), endotoxins (inhalable, respirable) and microorganisms (total mesophilic bacteria, *Enterobacteriaceae*, fungi) from ventilated livestock buildings.

The calculation model and the emission factors are the basis for a simple geographical information system designed to display the calculated emission potencies of livestock-related bioaerosols for the year 1999 in the 46 districts and autonomous cities in Lower Saxony, Germany. The three highest emissions of inhalable dust were determined for the three animal-dense districts of Grafschaft Bentheim ( $485.3 \text{ kg a}^{-1} \text{ km}^{-2}$ ), Cloppenburg ( $648.8 \text{ kg a}^{-1} \text{ km}^{-2}$ ) and Vechta ( $1203.4 \text{ kg a}^{-1} \text{ km}^{-2}$ ). On the other hand, the lowest bioaerosol emissions were found for the cities of Salzgitter ( $9.6 \text{ kg a}^{-1} \text{ km}^{-2}$ ), Braunschweig ( $10.6 \text{ kg a}^{-1} \text{ km}^{-2}$ ) and Wolfenbüttel ( $12.2 \text{ kg a}^{-1} \text{ km}^{-2}$ ) due to their more urban, non-agricultural setting.

With the aid of the agricultural census data, the percentages of temporal emission variations were assessed between 1996 and 1999, and found to have changed distinctly due to fluctuations in animal numbers in the districts. The following changes were noted in the three districts with the greatest increase or decrease of emitted particulate matter from 1996 to 1999: more inhalable dust was emitted in the rural districts of Stade (+9.6%), Cloppenburg (+14.9%) and Emsland (+18.2%), while there were clear declines in Oldenburg City (−24.1%), the district Helmstedt (−15.1%) and Braunschweig City (−14.4%).

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## 1. Introduction

Modern farm animal production is increasingly regarded as a source of solid, liquid and gaseous emissions which can be both a nuisance and environmentally harmful (Hartung and Wathes, 2001). The

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most important aerial pollutants within animal houses are the so-called bioaerosols, which are emitted in considerable quantities into the environment by ventilation systems. Bioaerosols in livestock buildings consist of a complex mixture of organic dust (e.g. proteins, polycarbohydrates), biologically active components (e.g. endotoxins, glucans) and microorganisms (e.g. bacteria, fungi). Sources of indoor releases are feed, bedding material, the animals themselves and their faeces with relative contributions to the total airborne dust of 80%–90%, 55%–68%, 2%–12% and 2%–8%, respectively. Mechanical forces (such as scratching on the pen wall) are also a minor source of dust originating from the equipment within the buildings. Small amounts of atmosphere-related particles are also transported into the livestock building via the ventilation system (Seedorf and Hartung, 2002a). The particle size of bioaerosols varies between 0.5 and 100  $\mu\text{m}$ , and bigger particles that have already settled can become airborne again by reentrainment forces (e.g. animal activity). Typically, bioaerosols are characterised by a range of biological properties which include infectivity, allergenicity, toxicity and pharmacological or similar effects (Hirst, 1995). Because of these impacts, bioaerosols are recognised as important health hazards at least for workers in livestock operations (Donham, 1993; Seedorf and Hartung, 2002a).

As is the case with other airborne pollutants, particulate matter such as bioaerosols are expected to be characterised in emission inventories. Emission inventories are helpful tools for the description of released amounts of specific airborne pollutants in a spatial and temporal context and they serve as a prospective planning basis for justified reduction and abatement measures. In this context, well-known inventory-related publications are the *EMEP/CORINAIR Atmospheric Emission Inventory Guidebook* (2001) and the document AP-42 of the US Environmental Protection Agency (EPA, 1995), which provide both methodologies for inventories and emission factors. Even though these guidebooks contain an agriculture-related chapter, they provide neither a defined methodology nor any agreed emission factors for livestock-related particulate matters. Alternative sources of information are available from the CEPMEIP (Co-ordinated European Programme on Particulate Matter Emission Inventories, Projections and Guidance) project at the Dutch research and technology organisation TNO (TNO, 2001), in which emission factors for livestock-related particulate matter are listed, but not clearly linked to references; nor have these factors been subject to independent review. The US Committee on Air Emissions from Animal Feeding Operations, 2002, 2003 have published two other reports with discussions of basic methodologies, proposals for methodological improvements and critical evaluations of available emission factors. The opinion

was that there is a lack of sufficient and validated data, which makes it impossible to recommend emission factors for particulate matter from livestock buildings.

It has also been argued that no currently accepted standard methods are available for bioaerosol measurements and that the importance of released bioaerosols for rural health demands is more or less speculative. On the other hand, it is known that bioaerosols contribute to the overall amount of atmospheric  $\text{PM}_{10}$  (particles  $< 10 \mu\text{m}$ ), which potentially increase the relative risk for morbidity and mortality (Lippman, et al., 2000; WHO, 1999). In a regional context, there is increasing concern that bioaerosol emissions may also be noxious agents that affect people and farm animals living in the vicinity of animal enterprises, because it was shown that abiotic and biotic particles may travel over relatively great distances (Gloster et al., 2003; Hartung et al., 1998; Müller and Wieser, 1987; Wilson, 2002). Airborne particles are also known to scatter radiation, contribute to climate change and attenuate visible radiation. The resulting impaired visibility and reduced solar radiation may also have ecological impacts such as restricting photosynthetic capabilities of plants (Grantz et al., 2003). Despite these complexities, an attempt has been made to cover the whole range of existing emission qualities due to agricultural operations in a first proposal by Phillips et al. (2002) of an applicable methodology for the calculation of agricultural  $\text{PM}_{10}$  emissions using livestock-related emission factors originally published by Takai et al. (1998). In the agricultural sector, which was responsible for 8.6% of all  $\text{PM}_{10}$  emissions in the EU in 2000, the major sources of particulate matter are poultry and pig production facilities, which are responsible for 57% and 32% of  $\text{PM}_{10}$  emissions (Klimont and Amann, 2002).

Despite all obstacles, progress toward creating an emission inventory is to be expected, as the UNECE Task Force on Emission Inventories decided in 1999 to incorporate a chapter on particulates in the *Atmospheric Emission Inventory Guidebook*. In this context, the aim of this paper is to propose a calculation procedure which will make it possible to establish an emission inventory for bioaerosols released from housed farm animals via the ventilation system and to introduce a simple geographical information system for Lower Saxony, Germany, for the spatial and temporal visualisation of emission potencies.

## 2. Material and methods

### 2.1. Measurement of indoor concentrations

A set of versatile and robust methods for quantifying gas and bioaerosol emissions was originally developed for a common field survey of livestock buildings in

northern Europe (England, The Netherlands, Denmark, Germany). Wathes et al. (1998) give an overview of this multinational project. The measurement devices and strategies used for German livestock buildings were identical to those used by all other project partners. Overall descriptions of these methods have recently been given by Phillips et al. (1998) and Seedorf et al. (1998). The present paper is using an updated German data base (Seedorf, 2003). The basic methods are briefly summarized below.

An air quality survey was carried out in all seasons in 82 commercially operated livestock buildings located in Lower Saxony, Germany. The livestock operations included dairy cow housing ( $n = 8$ ), beef cattle ( $n = 10$ ), calves ( $n = 16$ ), sows ( $n = 16$ ), weaners ( $n = 8$ ), fattening pigs ( $n = 8$ ), laying hens ( $n = 8$ ) and broilers ( $n = 8$ ). The determination of airborne pollutants was carried out by means of a fully mobile laboratory including most of the analytical instrumentation and auxiliary sampling devices mounted in a trailer which was parked adjacent to the wall of the respective animal house. Six sampling points were distributed on a vertical cross-section close to the centre of the respective animal house with three points at animal level and three points at a higher level. A seventh sampling location was individually installed close to a ventilation exhaust to monitor the emission loads released via the outlet system of the investigated livestock buildings.

Continuous hourly measurements of carbon dioxide ( $\text{CO}_2$ ) were made at one outdoor reference sampling point and at all seven indoor sampling sites. The  $\text{CO}_2$  analyser used was an infrared absorptiometer. Inhalable, respirable dust (50% cut-off at an aerodynamic particle diameter of  $5\text{ }\mu\text{m}$ ) and endotoxins in the dust fractions were collected at all seven sampling sites. The dust fractions and thus the endotoxins were sampled continuously on glass fibre filters via the filtration method. Samples of inhalable dust were collected on IOM (Institute of Occupational Medicine, Edinburgh) dust samplers (Mark and Vincent, 1986). Particle-size selective sampling of respirable dust was achieved with cyclone samplers. After gravimetric analysis, the dust results were expressed as  $\text{mg dust per m}^3$  air. For dust emission calculations only the seventh sampling location was used to determine released particulate loads. Pooled samples from all indoor filters were analysed to determine endotoxin concentrations in the dust samples with a turbidimetric-kinetic Limulus-Amebocyte-Lysate (LAL) gelation test. The airborne yields of endotoxins were expressed as  $\text{ng endotoxin per m}^3$  air. Pumps in the trailer and connecting tubes provided the necessary vacuum for the dust sampling devices, and the indoor and outdoor air samples were transported by a bundle of insulated tubes to the  $\text{CO}_2$  analyser inside the trailer. Every measuring cycle in each of the animal houses comprised a 24-h period always starting at 06.00 h.

Sampling of airborne microorganisms was carried out with an automated bacteria sampler (ABS), which has been described elsewhere (Pahl et al., 1997). The principle of the ABS is based on a slit sampler, which sucks air through a  $360^\circ$  rotating slit and impacts airborne microorganisms directly onto the surface of agar plates beneath the slit. The total amount of incubated and cultivable mesophilic bacteria, *Enterobacteriaceae* and fungi were determined by the ABS on Plate-Count-Agar, Violet-Red-Bile-Agar and Sabouraud-Agar plates, respectively, usually mounted in the centre of the animal house and exposed at 10.00 h and at 02.00 h the following day. After aerobic cultivation at between 20 and  $37^\circ\text{C}$  the grown colonies were counted and the concentrations expressed as colony-forming units (CFU) per  $\text{m}^3$ .

## 2.2. Determination of emission factors

The emission factors were estimated by multiplying the daily mean concentrations of bioaerosols and the daily mean ventilation rate ( $\text{m}^3\text{ h}^{-1}$ ), which was determined with the aid of the  $\text{CO}_2$  balance method (CIGR, 1984; DIN 18910, 1992; van Ouwerkerk, 1994; van Ouwerkerk and Pedersen, 1994). The original emission factors are expressed per livestock unit (LU, equivalent to 500 kg body weight). Relating the emission factors to a standardised body weight is advantageous, because emission calculations can more easily be scaled up or down with the varying weights of animals of the same species (Lacey et al., 2003). Furthermore, it is quite difficult to make a direct comparison of emission factors among farm animals with naturally different body sizes (such as broilers and dairy cows) and can possibly give misleading information about the emission potency of an individual animal of the various farm animal species. Therefore, the LU relation was chosen to extrapolate the individual emission factors per animal depending on the assumed body weight (see next chapter). The calculated emission factors represent median values per livestock class (Table 1).

## 2.3. Methodology of emission calculations

Here, a calculation model for regional bioaerosol emissions is described according to the preliminary proposal of Seedorf and Hartung (2001). Basically, emission amounts can be calculated by multiplying compound-specific emission factors and the number of animals per a specified space. These data are regularly recorded by the official national agricultural census. The mathematical equation is:

$$E_{B(x,t,s)} = n_{T(t,s)} \text{EF}_{(x,t)}, \quad (1)$$

where  $E_{B(x,t,s)}$  is the emission strength of a specific bioaerosol component  $x$  for a livestock class  $t$  in a

Table 1

Calculated emission factors for livestock-related bioaerosols in Lower Saxony, Germany (Seedorf, 2003)

| Per LU <sup>a</sup><br>livestock class (t) | Inh. <sup>c</sup> dust<br>(g h <sup>-1</sup> ) | Resp. <sup>d</sup><br>dust<br>(g h <sup>-1</sup> ) | Inh. etox <sup>e</sup><br>(μg h <sup>-1</sup> ) | Resp.<br>etox<br>(μg h <sup>-1</sup> ) | mBact. <sup>f</sup><br>(CFU <sup>i</sup> h <sup>-1</sup> ) | Enterobact. <sup>g</sup><br>(CFU h <sup>-1</sup> ) | Fungi <sup>h</sup><br>(CFU h <sup>-1</sup> ) |
|--|--|--|---|--|--|--|--|
| Dairy cows                                 | 0.216  | 0.018  | 0.877   | 0.023                                  | 1.823E+06  | 1.000E+04  | 1.073E+06                                    |
| Beef                                       | 0.131  | 0.009  | 2.082   | 0.075                                  | 2.480E+06  | 1.000E+04  | 6.130E+05                                    |
| Calves                                     | 0.216  | 0.038  | 4.082   | 0.220                                  | 6.815E+06  | 2.750E+04  | 2.285E+06                                    |
| Sows                                       | 0.235  | 0.029  | 4.216   | 2.257                                  | 5.720E+07  | 2.800E+05  | 1.829E+06                                    |
| Weaners <sup>b</sup>                       | 0.625  | 0.058  | 4.806   | 1.160                                  | 1.653E+07  | 7.342E+06  | 5.625E+05                                    |
| Fattening pigs                             | 0.678  | 0.045  | 2.917   | 0.470                                  | 3.073E+07  | 1.446E+06  | 6.630E+05                                    |
| Laying hens                                | 0.676  | 0.027  | 5.624   | 0.260                                  | 8.273E+06  | 2.610E+05  | 1.013E+06                                    |
| Broilers                                   | 2.988  | 0.477  | 88.875  | 19.971                                 | 3.435E+09  | 1.414E+06  | 3.628E+07                                    |

<sup>a</sup>LU is equivalent to 500 kg body weight.<sup>b</sup>Emission factors for microorganisms from Seedorf et al. (1998).<sup>c</sup>Inh. = inhalable.<sup>d</sup>Resp. = respirable.<sup>e</sup>etox = endotoxins.<sup>f</sup>mBact. = mesophilic bacteria.<sup>g</sup>Enterobact. = *Enterobacteriaceae*.<sup>h</sup>Mesophilic fungi.<sup>i</sup>CFU = colony-forming units.

geographically defined region  $s$  (i.e. district),  $n_{T(t,s)}$  is the number of farm animals, and  $EF_{(x,t)}$  indicates the emission factor expressed as mass per animal and year and CFU per animal and year. Due to the availability of emission factors, which are related to LU, it is necessary to make a conversion to the animal level, for which purpose a species-related conversion factor  $f_{(t)}$  is introduced (Table 2) to take into account the assumed specific representative body weight of housed farm animals. Because of the requirement for annual emission quantities, an extrapolation of the hourly based emission factors must be performed by adding a factor of 8760 (24 h × 365 days). All components together lead to the following equation:

$$EF_{(x,t)} = f_{G(t)} EF_{500(x,t)} \times 8760, \quad (2)$$

where  $EF_{500(x,t)}$  represents the species- and pollutant-related emission factors in Table 1. The livestock classes do not correspond to the animal type classifications published by the national office of statistics of Lower Saxony in the agricultural census. Appropriate adjustments of these categories are indicated in Table 2. It is currently not possible to include turkeys, ducks, geese, horses, sheep and goats because there are no reliable emission data available for these species.

The proposed emission factors have to be used in relation to the average number of animals in a certain year. As already mentioned, this number of animals is obtained from the agricultural census, and the total emission loads are related to 1 yr. However, due to typical animal production cycles (all-in/all-out system

with cleaning and disinfection periods; grazing periods), livestock buildings are not occupied for a whole year. As a consequence, an animal-free livestock building releases no emissions and the theoretical yearly average number of animals deviates from the real mean number of animals. These practical circumstances were recently recognised by Klimont and Amann (2002), who have supplemented the emission factors with data about the length of the housing period; however, it remains unclear how they calculated the housing periods. To supply this missing link, the following procedure was used as follows here. A fattening pig unit of 1000 animal places with a fattening period of 125 days and a service period of 7 days needs a total of 132 days for one complete production cycle. This means that there are 2.8 fattening periods per year, but that the livestock building is animal free nearly 20 days per year. As a consequence, this corresponds to a production capacity of only 96%. Therefore, the real average number of animals per year would be 960. This number is then linked to the emission factor to calculate the annual average emission quantities. The specific production circumstances of each animal type have to be known to take the dynamic production cycle into consideration.

The animal-specific length of the production cycle  $P_{(t)}$ , expressed as number of days, is the sum of the number of days that animals are kept within the production facility ( $A_{(t)}$ ) and the period of time when the livestock house is empty ( $L_{(t)}$ ):

$$P_{(t)} = A_{(t)} + L_{(t)}. \quad (3)$$

Table 2

Animal types according to the agricultural census data; definition of animal type categories and their related conversion  $f_{G(t)}$  to estimate the number of animals per livestock unit (LU)

| Animal type and the integrated subgroups  | Definition of animal type categories for the emission inventory and the estimated body weight (BW) in kg (in brackets) | Conversion factor <sup>a</sup> $f_{G(t)}$ |
|---|--|---|
| <i>Cattle:</i>  |  |   |
| ● 2 yr old and older <ul style="list-style-type: none"> <li>○ Female for slaughtering</li> <li>○ Female for breeding</li> <li>○ Dairy cows</li> <li>○ Extensive kept cattles (e.g. suckler cows)</li> <li>○ Fattening female cattle</li> </ul>  | Dairy cows (500)   | 1.0                                       |
| ● Juvenile cattle <ul style="list-style-type: none"> <li>○ 6 months to less than 1 yr old (female, male)</li> <li>○ 1 to less than 2 yr old <ul style="list-style-type: none"> <li>● male</li> <li>● Female for slaughtering</li> <li>● Female for breeding</li> <li>● 2 yr and older (male)</li> </ul> </li> </ul> | Beef (350)   | 0.7                                       |
| ● Calves less than 6 months or with BW < 220 kg   | Calves (150)   | 0.3                                       |
| <i>Pigs:</i>  |  |   |
| ● Pigs for breeding with BW > 50 kg <ul style="list-style-type: none"> <li>○ Young sows in 1. pregnancy</li> <li>○ Young sows not pregnant</li> <li>○ All adult sows pregnant and not pregnant</li> </ul>   | Sows (150)   | 0.3                                       |
| ● Boars   |  |   |
| ● Piglets <ul style="list-style-type: none"> <li>○ less than 20 kg and up to 50 kg BW</li> </ul>  | Weaners (20)   | 0.04 <sup>b</sup>                         |
| ● Fattening pigs <ul style="list-style-type: none"> <li>○ 50–110 kg BW</li> </ul>   | Fattening pigs (80)  | 0.16                                      |
| <i>Chickens:</i>  |  |   |
| ● Laying hens <ul style="list-style-type: none"> <li>○ 6 months and older</li> <li>○ Chicks and hens younger than 6 months</li> </ul>   | Laying hens (2)  | 0.004                                     |
| ● Broiler   | Broiler (2)  | 0.004                                     |

<sup>a</sup>Federal Ministry of Consumer Protection, Food and Agriculture (2000).

<sup>b</sup>Mean of conversion factors from weaners (0.02) and from young pigs with less than 50 kg body weight (0.06).

With a 365-day year the number of production cycles  $Z_{J(t)}$  is then:

$$Z_{J(t)} = \frac{365}{P_{(t)}}. \quad (4)$$

Combining the information from Eqs. (3) and (4), a correction factor  $f_{J(t)}$  can be calculated to adjust the official results of the agricultural census to arrive at a figure about the probable mean number of animals per year:

$$f_{J(t)} = \frac{(P_{(t)} - L_{(t)} - O_{(t)})Z_{J(t)}}{365}, \quad (5)$$

where  $O_{(t)}$  is the period when active ventilation does not take place or is negligible, for example in broiler production. Table 3 contains the assumed livestock

production data for the use of Eqs. (3)–(5). Taking all relevant factors together, Eq. (1) can now be modified in the following way:

$$E_{B(x,t,s)} = f_{J(t)} n_{T(t,s)} E_{F(x,t)}. \quad (6)$$

In conclusion, the corresponding emission loads of all contributing animal production facilities can be calculated for each bioaerosol component as follows:

$$E_{B(\text{total})} = \sum_{i=1}^n E_{B(x,t,s)_i}. \quad (7)$$

Eqs. (1)–(7) were used to establish a comprehensive spatial emission inventory for bioaerosols. Apart from local emissions, temporal variations in released bioaerosols can also be calculated according to the numbers of

Table 3

Assumed data of production cycles and calculated correction factors by Eqs. (3)–(6)

| Livestock class ( <i>t</i> ) | $A^d$ | $L^e$ | $P$   | $O$  | $Z_J$ | $f_J$ |
|------------------------------|-------|-------|-------|------|-------|-------|
| Dairy cows <sup>a</sup>      | 182.5 | 182.5 | 365.0 | 0.0  | 1.0   | 0.50  |
| Beef                         | 365.0 | 0.0   | 365.0 | 0.0  | 1.0   | 1.00  |
| Calves                       | 182.5 | 7.0   | 189.5 | 0.0  | 1.9   | 0.95  |
| Sows <sup>b</sup>            | 365.0 | 0.0   | 365.0 | 0.0  | 1.0   | 1.00  |
| Weaners                      | 49.0  | 7.0   | 56.0  | 0.0  | 6.5   | 0.87  |
| Fattening pigs               | 125.0 | 7.0   | 132.0 | 0.0  | 2.8   | 0.96  |
| Laying hens                  | 365.0 | 0.0   | 365.0 | 0.0  | 1.0   | 1.00  |
| Broilers <sup>c</sup>        | 33.0  | 14.0  | 47.0  | 10.0 | 7.8   | 0.49  |

<sup>a</sup>Dairy cows only housed for 6 months during the winter.<sup>b</sup>Continuous housing of sows assumed.<sup>c</sup>Shortest fattening period defined for broilers.<sup>d</sup>Production cycles > 1 yr (beef, laying hens) were set to 365 days.<sup>e</sup>Cleaning, disinfection and drying-up period; grazing period in case of dairy cows.

animals recorded regularly during the agricultural census. The accuracy of each inter-annual emission calculation is highly dependent on the availability of a complete official data set. However, the authorities are not required automatically to provide complete data sets of animal numbers, and the absence of data can be due to data protection regulations; therefore, data sets have to be checked for completeness. The relative difference in emission quantities between two years is then expressed as follows:

$$\Delta E_B = \frac{E_{B(y1)} - E_{B(y2)}}{E_{B(y2)}} \times 100, \quad (8)$$

where  $\Delta E_B$  is the relative increase or decrease of bioaerosol emissions in percent, for one year and km<sup>2</sup>. The terms  $E_{B(y1)}$  and  $E_{B(y2)}$  indicate the total bioaerosol emission amounts for 1999 and 1996, respectively.

The calculated bioaerosol emissions for 1999 are displayed in terms of a simple geographic information system (GIS), which included either lists of numerical data in tables or as graphics. The area within the borders of each district is marked according to a colour legend for the total yearly emissions and the relative deviation of emissions between 1996 and 1999, for an area of 1 km<sup>2</sup> calculated from the individual surface area per district. All districts and autonomous cities of Lower Saxony comprise a total surface area of approximately 47,600 km<sup>2</sup> (NLS, 1998).

### 3. Results

A simple GIS was applied to represent an emission inventory for livestock-related inhalable dust. Fig. 1 shows the emission conditions in Lower Saxony, Germany, during 1999. Three different regions can be distinguished in Lower Saxony with clearly different

emission strengths per km<sup>2</sup>. The highest dust releases were calculated for the central part of northwest Lower Saxony, followed by the more coastal districts in the north. A third group of districts with low emissions are located in northeastern and southern Lower Saxony.

The relative proportion of emission-causing livestock species is also integrated in Fig. 1. The distribution of livestock species over Lower Saxony is also remarkable, because the different livestock types cause different magnitudes of emissions. For example, the grassland coastal regions are mainly occupied by cattle, and 96% and 90% of the dust emitted per year and km<sup>2</sup> in the districts Wesermarsch and Leer, respectively, was caused by cattle livestock buildings. In the remaining districts, pigs represent a large group of emitters, which generally caused the highest relative emissions of all considered farm species both in districts with high and low annual release quantities per km<sup>2</sup>. For example, the pig herds in the districts of Hildesheim (low dust load area) and Cloppenburg (high dust load area) contributed 71% and 63% of the district-specific total inhalable dust emissions, respectively. The districts with the highest emission strength in the northwestern part of Lower Saxony are characterised by a relatively high amount of housed chickens. The three districts with more than one-third of the dust emissions caused by chickens are Grafschaft Bentheim (46%), Vechta (37%) and Emsland (36%).

The released bioaerosol amounts for 1999 are listed in Table 4 for all districts and cities in Lower Saxony in terms of absolute emission quantities. A rank order of increasing emissions was additionally included in Table 4 to show which districts caused the lowest and highest atmospheric particle loads per km<sup>2</sup>. Comparison of the first and the last rank positions shows that livestock buildings within the borders of the city of Salzgitter caused only 9.6 kg a<sup>-1</sup> km<sup>-2</sup> as opposed to



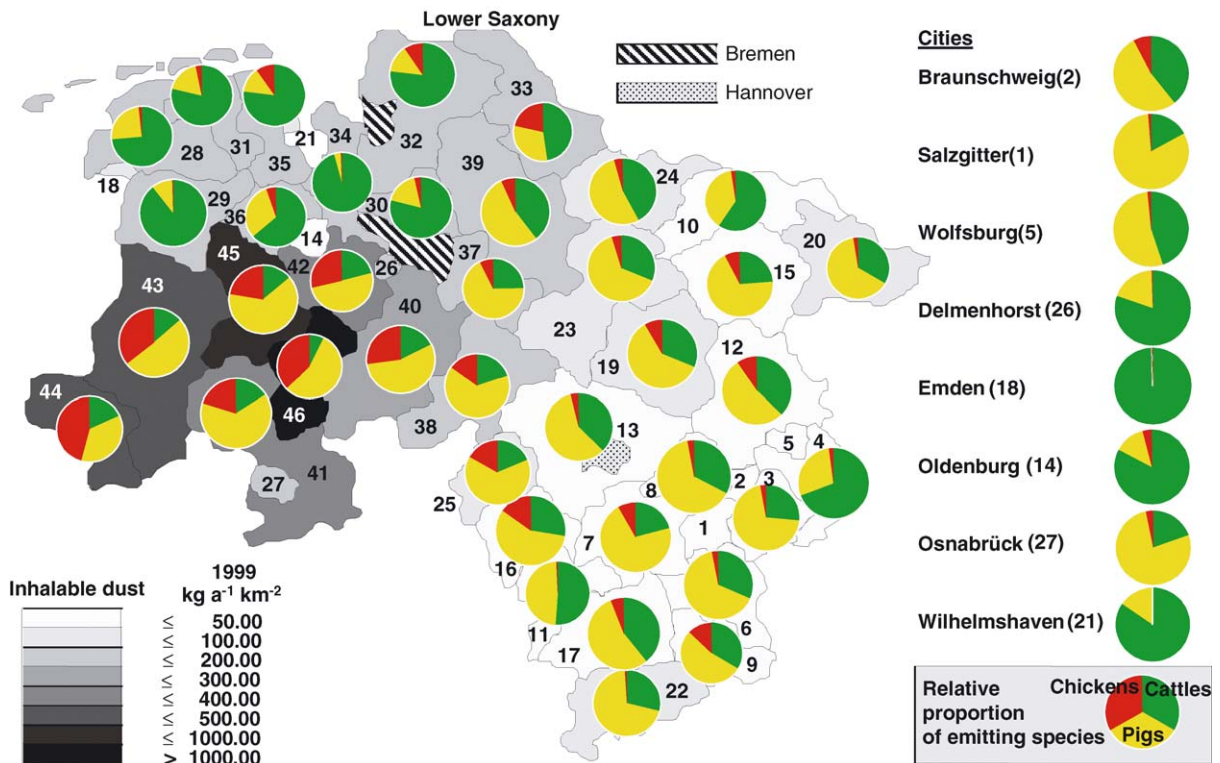


Fig. 1. Spatial distribution of emitted inhalable dust quantities from livestock operations in Lower Saxony, Germany, and the relative proportion of emitting species for 1999. Numbers indicate names of the districts and cities according to Table 4, first column. The cities Hannover and Bremen (Bremen does not belong to Lower Saxony) were additionally marked.

1203.4 kg a<sup>-1</sup> km<sup>-2</sup> in the district of Vechta, which is equivalent to a difference of factor 125. But animal production facilities were also responsible for the highest emissions of respirable dust, inhalable endotoxins, Enterobacteriaceae, and fungi in Vechta both on the yearly and area-related bases. The district Grafschaft Bentheim had the highest emissions of respirable endotoxins and total mesophilic bacteria, although the relative difference to Vechta was only 0.34% for respirable endotoxins, but it was 14.8% for bacteria. Despite the rank order for inhalable dust, it is possible to rule out a similar hierarchy for the other airborne components with a few exceptions. This observation is a strong indication of the agglomeration of dust, dust components and microorganisms covered by the terminus bioaerosol.

The yearly comparison of emitted livestock-related bioaerosols allows the assessment of temporal variations in a spatial context, as was done for 1996 and 1999 (Fig. 2). The coloured map identifies the geographical zones with increased and decreased inhalable dust amounts immediately; these zones reflect the dynamic range of dust emissions in Lower Saxony. Especially in the western part of Lower Saxony, in the districts of Emsland and Cloppenburg, livestock operations have

caused marked increases of 18.2% and 14.9%, respectively. The district of Vechta was responsible for only a marginal increase of 5.8%, although this region showed the highest dust emissions in 1999. Coastal districts like Stade, Friesland and Cuxhaven have also shown increases of 9.6%, 6.2% and 5.3%, respectively. Small increases of ≤2% were observed in the districts of Wesermarsch, Harburg, Hildesheim, Gifhorn and Hannover. In the south of Lower Saxony, the district of Hameln-Pyrmont showed the largest increase of dust emissions in that region (9.0%). Within three years, there was a reduction of emitted inhalable dust in only 16 of 46 districts (Table 5). Since the same calculation schemes were applied for emission quantities in the years 1996 and 1999, the deviations found can be attributed to fluctuations in numbers of animals recorded during the agricultural census.

#### 4. Discussion

Emission inventories enable authorities to record and to monitor atmospheric pollution caused by a wide variety of emitting facilities. Furthermore, these inventories are important decision-making tools for

Table 4

Emission inventory for livestock-related bioaerosols in Lower Saxony, Germany, for the year 1999; rank order according to increasing inhalable dust emissions

| 1999<br>rank | District                      | Inh. dust<br>(kg a <sup>-1</sup> km <sup>-2</sup> ) | Resp. dust<br>(kg a <sup>-1</sup> km <sup>-2</sup> ) | Inh. etox<br>(g a <sup>-1</sup> km <sup>-2</sup> ) | Resp. etox<br>(g a <sup>-1</sup> km <sup>-2</sup> ) | mBact.<br>(KBE a <sup>-1</sup> km <sup>-2</sup> ) | Enterobact.<br>(KBE a <sup>-1</sup> km <sup>-2</sup> ) | Fungi<br>(KBE a <sup>-1</sup> km <sup>-2</sup> ) |
|--------------|-------------------------------|---|--|--|---|---|--|--|
| 1            | Salzgitter, City <sup>a</sup> | 9.6   | 0.7  | 0.06   | 0.010   | 4.3E+11   | 2.5E+10  | 1.8E+10  |
| 2            | Braunschweig, City            | 10.6  | 0.8  | 0.09   | 0.015   | 4.8E+11   | 2.1E+10  | 3.5E+10  |
| 3            | Wolfenbüttel                  | 12.2  | 1.0  | 0.09   | 0.018   | 6.3E+11   | 3.2E+10  | 3.3E+10  |
| 4            | Helmstedt                     | 12.6  | 1.1  | 0.11   | 0.013   | 4.3E+11   | 1.5E+10  | 5.6E+10  |
| 5            | Wolfsburg, City               | 15.8  | 1.2  | 0.11   | 0.010   | 5.2E+11   | 2.2E+10  | 4.8E+10  |
| 6            | Goslar                        | 20.4  | 1.6  | 0.15   | 0.026   | 9.3E+11   | 5.0E+10  | 5.7E+10  |
| 7            | Hildesheim                    | 28.2  | 2.3  | 0.24   | 0.049   | 3.2E+12   | 7.2E+10  | 8.5E+10  |
| 8            | Peine                         | 31.3  | 2.4  | 0.23   | 0.039   | 1.5E+12   | 7.2E+10  | 8.9E+10  |
| 9            | Osterode am Harz              | 33.4  | 2.6  | 0.28   | 0.037   | 3.2E+12   | 5.4E+10  | 1.1E+11  |
| 10           | Lüneburg                      | 36.5  | 3.1  | 0.33   | 0.041   | 1.4E+12   | 5.7E+10  | 1.5E+11  |
| 11           | Holzminde                     | 41.5  | 3.4  | 0.34   | 0.045   | 1.6E+12   | 7.6E+10  | 1.5E+11  |
| 12           | Gifhorn                       | 43.2  | 3.6  | 0.39   | 0.061   | 4.6E+12   | 8.8E+10  | 1.5E+11  |
| 13           | Hannover <sup>b</sup>         | 43.8  | 3.7  | 0.38   | 0.072   | 3.1E+12   | 1.1E+11  | 1.5E+11  |
| 14           | Oldenburg, City               | 44.0  | 3.6  | 0.46   | 0.028   | 1.0E+12   | 2.5E+10  | 2.1E+11  |
| 15           | Uelzen                        | 46.2  | 3.5  | 0.34   | 0.061   | 2.2E+12   | 1.1E+11  | 1.2E+11  |
| 16           | Hameln-Pyrmont                | 48.6  | 4.5  | 0.53   | 0.107   | 1.0E+13   | 1.1E+11  | 2.1E+11  |
| 17           | Northeim                      | 49.4  | 4.1  | 0.43   | 0.076   | 4.1E+12   | 1.0E+11  | 1.8E+11  |
| 18           | Emden, City                   | 51.1  | 4.6  | 0.52   | 0.020   | 8.0E+11   | 3.9E+09  | 2.8E+11  |
| 19           | Celle                         | 51.8  | 4.0  | 0.40   | 0.063   | 2.2E+12   | 1.2E+11  | 1.4E+11  |
| 20           | Lüchow-Dannenberg             | 59.5  | 4.8  | 0.48   | 0.087   | 2.9E+12   | 1.5E+11  | 1.8E+11  |
| 21           | Wilhelmshaven, City           | 59.6  | 5.1  | 0.56   | 0.032   | 1.3E+12   | 3.4E+10  | 2.9E+11  |
| 22           | Göttingen                     | 59.7  | 4.7  | 0.43   | 0.081   | 2.9E+12   | 1.5E+11  | 1.6E+11  |
| 23           | Soltau-Fallingbostel          | 68.1  | 5.6  | 0.59   | 0.110   | 5.6E+12   | 1.8E+11  | 2.2E+11  |
| 24           | Harburg                       | 73.5  | 5.7  | 0.57   | 0.071   | 2.8E+12   | 1.4E+11  | 2.3E+11  |
| 25           | Schaumburg                    | 92.7  | 7.5  | 0.83   | 0.158   | 1.3E+13   | 2.1E+11  | 2.9E+11  |
| 26           | Delmenhorst, City             | 108.6   | 9.3  | 1.16   | 0.100   | 3.1E+12   | 1.1E+11  | 5.2E+11  |
| 27           | Osnabrück, City               | 109.3   | 8.2  | 0.76   | 0.135   | 5.3E+12   | 2.7E+11  | 2.4E+11  |
| 28           | Aurich                        | 115.7   | 10.1   | 1.12   | 0.126   | 5.0E+12   | 1.3E+11  | 5.5E+11  |
| 29           | Leer                          | 124.1   | 10.7   | 1.14   | 0.064   | 2.5E+12   | 4.9E+10  | 6.3E+11  |
| 30           | Osterholz                     | 129.6   | 11.2   | 1.39   | 0.123   | 6.7E+12   | 1.1E+11  | 6.4E+11  |
| 31           | Wittmund                      | 140.6   | 12.6   | 1.47   | 0.147   | 7.6E+12   | 1.3E+11  | 7.2E+11  |
| 32           | Cuxhaven                      | 144.8   | 12.8   | 1.63   | 0.154   | 1.3E+13   | 9.8E+10  | 7.6E+11  |
| 33           | Stade                         | 148.5   | 14.4   | 1.96   | 0.320   | 3.8E+13   | 1.9E+11  | 8.2E+11  |
| 34           | Wesermarsch                   | 150.1   | 13.2   | 1.52   | 0.070   | 2.6E+12   | 3.2E+10  | 8.0E+11  |
| 35           | Friesland                     | 155.3   | 14.3   | 1.76   | 0.175   | 1.8E+13   | 9.7E+10  | 8.5E+11  |
| 36           | Ammerland                     | 164.6   | 14.5   | 1.77   | 0.210   | 1.5E+13   | 2.1E+11  | 7.7E+11  |
| 37           | Verden                        | 180.5   | 15.0   | 1.55   | 0.284   | 2.2E+13   | 4.5E+11  | 5.7E+11  |
| 38           | Nienburg upon Weser           | 185.4   | 15.5   | 1.76   | 0.348   | 2.8E+13   | 4.5E+11  | 6.2E+11  |
| 39           | Rotenburg upon Wümme          | 192.8   | 16.1   | 1.76   | 0.254   | 2.0E+13   | 3.7E+11  | 7.0E+11  |
| 40           | Diepholz                      | 276.9   | 27.1   | 3.58   | 0.748   | 9.0E+13   | 5.8E+11  | 1.4E+12  |
| 41           | Osnabrück                     | 330.9   | 26.7   | 3.12   | 0.617   | 4.5E+13   | 8.0E+11  | 1.0E+12  |
| 42           | Oldenburg                     | 331.4   | 31.4   | 4.25   | 0.809   | 9.9E+13   | 6.1E+11  | 1.6E+12  |
| 43           | Emsland                       | 405.1   | 43.5   | 6.30   | 1.397   | 1.7E+14   | 8.5E+11  | 2.4E+12  |
| 44           | Grafschaft Bentheim           | 485.3   | 58.6   | 9.16   | 2.065   | 2.7E+14   | 9.1E+11  | 3.6E+12  |
| 45           | Cloppenburg                   | 648.8   | 59.7   | 7.39   | 1.449   | 1.7E+14   | 1.4E+12  | 2.7E+12  |
| 46           | Vechta                        | 1203.4  | 94.9   | 12.05  | 2.058   | 2.3E+14   | 2.3E+12  | 3.8E+12  |

For abbreviations, see Table 1.

<sup>a</sup>The additions 'City' mean autonomous city.

<sup>b</sup>Hannover includes city and surrounding district.

plans to prevent atmospheric pollution regionally and globally. For this reason inventories should reflect the calculated emissions as well as possible. But inventories contain several uncertainty factors that can influence the

quality of such emission estimates. In the following, selected critical points are pointed out and discussed as to their particular relevance to the bioaerosol emission inventory presented here.



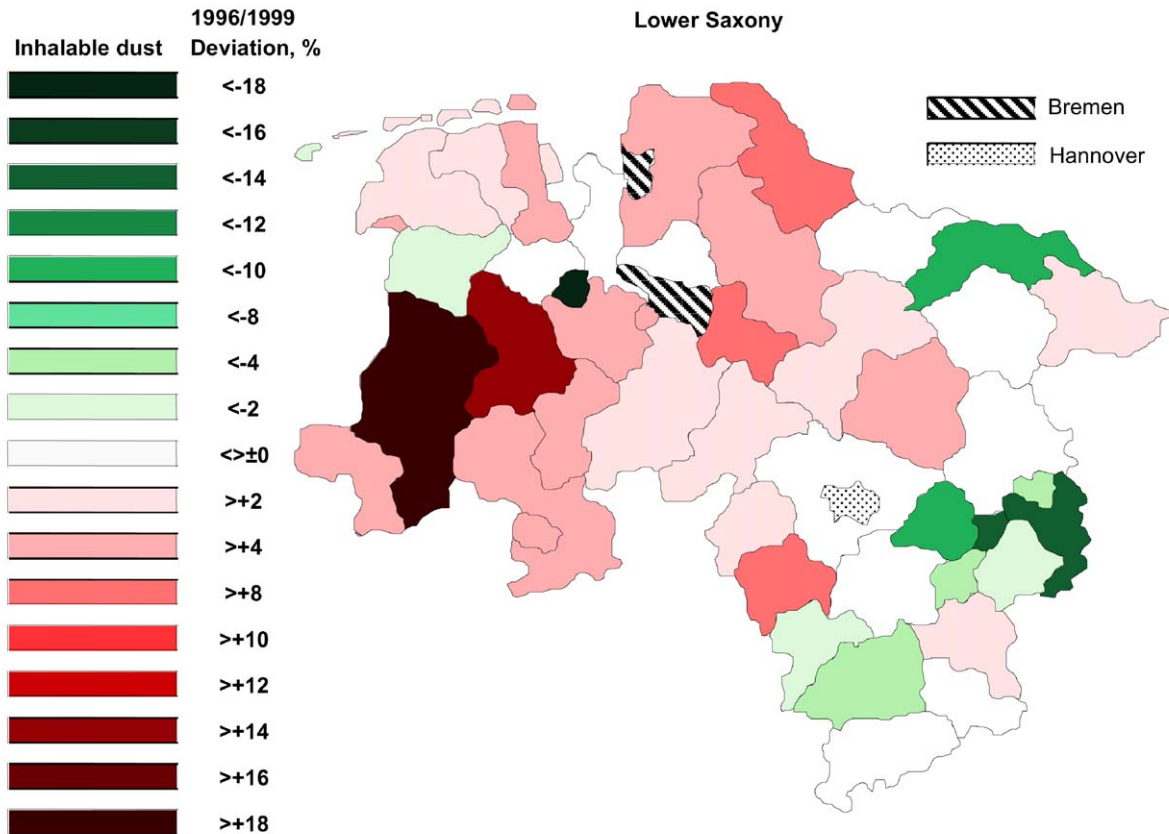


Fig. 2. Relative inter-annual increase (+ %) and decrease (– %) of emitted inhalable dust from livestock buildings in Lower Saxony, Germany, between 1996 and 1999.

#### 4.1. Determination of the emission factors

The quality of the applied methods for deriving emission factors is important for the reliability of an inventory. As emission factors are a combination of measured concentrations of the airborne pollutant and the corresponding ventilation rates, the quality of the measurement techniques has to be assured. While sampling and analysis methods for particulate matters, microorganisms and endotoxins have been comprehensively described elsewhere (Douwes et al., 1995; Henningson and Ahlberg, 1994; Li et al., 2000; Taylor and Reynolds, 2001; Thorne et al., 1997), the derivation of valid ventilation rates needs some explanation. Despite the use of a representative multi-point measurement strategy with seven indoor sampling points, the indirect determination of the ventilation rate via the CO<sub>2</sub> balance method may be a critical point, because the balance principle is founded on the suggestion that most of the released CO<sub>2</sub> is produced by the animals themselves as a steady state. But individual and chronobiological variations of the metabolism (the

animal's age, performance, pregnancy, diseases, activity etc.) cause a more dynamic CO<sub>2</sub> production. This is one reason why Hinz et al. (1995) suggested that the determination of animal-related CO<sub>2</sub> production is not accurately predictable, and Scholtens and van't Klooster (1994) stated that the balance method may cause an error of 20%. To reduce this error, van Ouwwerkerk and Pedersen (1994) integrated the so-called respiratory quotient (RQ) in their balance model by which the amount of produced CO<sub>2</sub> is indirectly defined as an indicator for varying metabolic activities. The RQ therefore improves the accuracy of calculated ventilation rates. In the present study, the emission factors were calculated with the aid of RQ.

In addition to CO<sub>2</sub>-expiring animals, there are other potential CO<sub>2</sub> sources that cannot be fully excluded. Decomposition processes in manure contribute to CO<sub>2</sub> yields in the air in livestock buildings (Bieber, 1975); this is a factor difficult to estimate (Müller, 2001). However, Hilliger (1983) estimated the manure-related portions of CO<sub>2</sub> as between 5% and 10% of that expired by the animals. Other investigators found similar, relatively

Table 5

Comparison of bioaerosol emissions from livestock buildings between 1996 and 1999 in Lower Saxony, Germany

| Rank | District                        | Differences in bioaerosol emissions between 1996 and 1999 (%) |            |           |            |        |             |       |
|------|---------------------------------|---|------------|-----------|------------|--------|-------------|-------|
|      |                                 | Inh. dust   | Resp. dust | Inh. etox | Resp. etox | mBact. | Enterobact. | Fungi |
| 1    | Oldenburg, City                 | −24.1   | −21.9      | −19.0     | −23.7      | −33.0  | −41.8       | −15.5 |
| 2    | Helmstedt                       | −15.1   | −13.4      | −13.7     | −16.4      | −17.6  | −30.0       | −9.5  |
| 3    | Braunschweig, City <sup>a</sup> | −14.4   | −13.3      | −8.5      | −10.5      | −16.2  | −13.5       | −10.1 |
| 4    | Lüneburg                        | −10.8   | −8.6       | −9.5      | −10.6      | −12.3  | −16.4       | −5.3  |
| 5    | Peine                           | −10.7   | −10.7      | −12.8     | −12.3      | −7.9   | −12.5       | −12.2 |
| 6    | Wolfsburg, City                 | −5.1  | −7.2       | −13.9     | −6.9       | 1.6    | 12.0        | −16.7 |
| 7    | Salzgitter, City                | −4.6  | −5.1       | −6.0      | −4.3       | −2.8   | −8.3        | −5.7  |
| 8    | Northheim                       | −4.4  | −5.0       | −7.6      | −1.5       | −2.3   | −3.7        | −7.6  |
| 9    | Holz Minden                     | −3.8  | −4.0       | −4.4      | −1.0       | 0.9    | −4.6        | −4.9  |
| 10   | Wolfenbüttel                    | −2.7  | −2.5       | −7.3      | −1.1       | −2.2   | −2.6        | −5.6  |
| 11   | Leer                            | −2.1  | −1.5       | −6.6      | −20.8      | −56.5  | 2.9         | −3.4  |
| 12   | Ammerland                       | −1.9  | −1.2       | −2.3      | 2.5        | 1.1    | 8.1         | −2.8  |
| 13   | Göttingen                       | −1.6  | −1.9       | −3.5      | −1.1       | 0.5    | −0.8        | −4.7  |
| 14   | Uelzen                          | −1.5  | −1.6       | −4.0      | 2.0        | 4.4    | 2.2         | −6.1  |
| 15   | Osterholz                       | −1.0  | 0.0        | 1.5       | 7.2        | 46.4   | 0.0         | 1.4   |
| 16   | Osterode am Harz                | −0.8  | −2.3       | −6.1      | −3.0       | 1.2    | −8.1        | −7.2  |
| 17   | Hannover <sup>b</sup>           | 0.6   | 3.2        | 4.3       | 11.7       | 47.0   | 5.7         | 4.3   |
| 18   | Gifhorn                         | 0.8   | 6.4        | 13.9      | 23.7       | 161.3  | −1.1        | 13.7  |
| 19   | Hildesheim                      | 1.3   | 0.3        | −2.6      | 1.5        | 4.8    | −0.8        | −4.1  |
| 20   | Harburg                         | 1.7   | 1.1        | −1.6      | 2.2        | 6.3    | 2.7         | −2.6  |
| 21   | Wesermarsch                     | 2.0   | 3.0        | 1.5       | 3.7        | 4.0    | 2.5         | 2.8   |
| 22   | Goslar                          | 2.3   | 1.8        | −3.2      | 1.8        | 3.7    | 11.9        | −6.7  |
| 23   | Wilhelmshaven, City             | 2.4   | 4.8        | 4.0       | 10.3       | 13.1   | 22.4        | 1.7   |
| 24   | Lüchow-Dannenberg               | 2.5   | 2.0        | −2.2      | 0.3        | 4.0    | 5.0         | −3.2  |
| 25   | Schaumburg                      | 2.5   | 6.9        | 10.9      | 16.5       | 49.4   | 0.2         | 13.3  |
| 26   | Wittmund                        | 2.8   | 7.4        | 7.9       | 27.0       | 105.0  | 23.4        | 6.4   |
| 27   | Nienburg upon Weser             | 3.2   | 5.8        | 5.0       | 10.0       | 23.6   | 3.9         | 5.8   |
| 28   | Aurich                          | 3.2   | 5.3        | 4.6       | 14.0       | 42.6   | 15.6        | 3.4   |
| 29   | Diepholz                        | 3.8   | 3.0        | 2.0       | 3.1        | 3.6    | 5.0         | 1.1   |
| 30   | Soltau-Fallingb.ostel           | 3.9   | 6.9        | 7.5       | 16.4       | 47.6   | 10.9        | 6.8   |
| 31   | Grafschaft Bentheim             | 4.1   | 2.8        | 1.6       | 2.2        | 1.6    | 8.9         | 1.0   |
| 32   | Celle                           | 4.1   | 3.3        | 0.0       | 8.6        | 8.7    | 10.9        | −3.0  |
| 33   | Emden, City                     | 4.8   | 10.9       | 14.4      | 19.4       | 15.1   | 5.5         | 11.0  |
| 34   | Cuxhaven                        | 5.3   | 4.9        | 7.3       | 18.9       | 32.3   | 18.6        | 5.0   |
| 35   | Delmenhorst, City               | 5.4   | 5.6        | 4.7       | −2.2       | −0.1   | 6.1         | 4.7   |
| 36   | Vechta                          | 5.8   | 3.2        | −1.0      | −1.1       | −4.5   | 6.8         | −3.0  |
| 37   | Friesland                       | 6.2   | 13.8       | 18.1      | 49.0       | 120.1  | 5.6         | 17.0  |
| 38   | Osnabrück, City                 | 6.7   | 4.6        | 2.0       | 1.5        | 6.4    | 3.2         | −0.9  |
| 39   | Oldenburg                       | 6.9   | 10.5       | 13.4      | 17.7       | 24.0   | 2.7         | 14.5  |
| 40   | Rotenburg upon Wümme            | 7.0   | 5.7        | 4.1       | 8.7        | 10.9   | 11.0        | 1.9   |
| 41   | Osnabrück                       | 7.5   | 7.8        | 6.1       | 7.9        | 14.8   | 8.6         | 5.2   |
| 42   | Verden                          | 8.6   | 7.9        | 5.7       | 8.9        | 7.9    | 17.6        | 2.7   |
| 43   | Hameln-Pyrmont                  | 9.0   | 15.3       | 22.4      | 34.1       | 64.4   | 7.0         | 22.0  |
| 44   | Stade                           | 9.6   | 12.9       | 16.6      | 28.9       | 33.8   | 17.2        | 14.7  |
| 45   | Cloppenburg                     | 14.9  | 15.8       | 16.7      | 20.9       | 26.3   | 14.2        | 16.1  |
| 46   | Emsland                         | 18.2  | 22.0       | 25.5      | 27.1       | 37.4   | 12.2        | 25.8  |

Differences are expressed in percent (%). Negative values indicate decreased emissions from 1996 to 1999 and positive values represent increased emissions. Rank order according to the relative increase of inhalable dust emissions.

For abbreviations, see Table 1.

<sup>a</sup>The additions 'City' mean autonomous city.

<sup>b</sup>Hannover includes city and surrounding district.

small percentages of CO<sub>2</sub> due to manure of between 4% (Aarnink et al., 1992) and 8.5% (Curtis, 1983). Therefore, here 4% of manure-related CO<sub>2</sub> was included to adjust the calculated ventilation rates and ultimately the emission factors. This procedure was confirmed by van Ouwerkerk and Pedersen (1994).

Additional useful information on the reliability of estimated ventilation rates can be derived from the comparison of ventilation rates derived by the CO<sub>2</sub> balance method and by direct measurements with fan anemometers as a reference method. Investigations of Hinz and Linke (1998a) have shown deviations between the two methods of  $\pm 20\%$ , on the basis of 24-h average values, but it must be recalled that fan anemometry itself has an error of  $\pm 5\%$  (Hinz and Linke, 1998b). Interestingly, the balance-related ventilation rates have shown a pattern of increasing and decreasing ventilation rates quite similar to those of the direct measurements (Hinz and Linke, 1998a), which indicates the sensitivity of the CO<sub>2</sub> balance method, at least for a 24-h period. This observation is important because the proposed emission factors were originally based on 24-h averages.

#### 4.2. Methodology

Not only must the algorithm used in an emission inventory be defined, the factors integrated into that algorithm also have to be evaluated for their reliability. Therefore, it is necessary to evaluate uncertainties for selected factors which may influence the accuracy of the proposed calculation procedure. Selected uncertainty factors are discussed below and summarised in Table 6.

Unlike the usual expression of emission factors as mass per animal and year, expressing emissions in relation to livestock units has the advantage that

emission factors can be extrapolated for the animal's body weight by conversion factors (see Table 2). In this way, emission factors can be applied more flexibly, because the theoretical emission potency can be calculated for each body weight. Unfortunately, such conversion factors are not always standardised. This is important, because emission inventories are a snapshot so to speak of a given number of farm animals counted during the annual agricultural census. No information about body weights in the herds is recorded in great detail or is at least not published. For example, two current official conversion factors of 0.004 and 0.0034 for laying hens result in body weights of 2.0 and 1.7 kg, respectively, which is equivalent to a 15% difference in emission potency. Therefore, it would be useful to have information about the course of weight development within a production cycle. Lacey et al. (2003) have recently shown how a broiler growth equation can be used to calculate a mean PM<sub>10</sub> mass release per bird during a whole fattening period. From this, a mean animal body mass could be computationally determined for the entire production cycle and substituted for the current conversion factors for body weights as shown in Table 2.

The temporal differentiation between occupied and unoccupied housing periods is also a relevant factor. For example, the proportional split into equal housing and grazing periods of 182.5 days each was chosen for dairy cows (see Table 3), but a grazing period of 172 days can be assumed in Lower Saxony. This deviation may be negligible, because the grazing period can easily be extended with favourable weather conditions and a sufficient supply of pasture grass. It also has to be kept in mind that the term 'dairy cows' includes further cattle types which might have still longer grazing periods (cf. Table 2). For example, grazing periods for cattle kept

Table 6  
Summary of uncertainty factors influencing the estimation of bioaerosol emissions

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|  |
|--|
| Quality of measurement techniques for bioaerosols (mechanical or optical, cultivable or non-cultivable determination methods).   |
| Physical density and particle size distribution of bioaerosols.  |
| Type of feeding system (dry or wet, manual or automatic).  |
| Kind of floor (partly or fully slatted, with/without bedding material, quality of bedding material, housed or free-range animals).   |
| Manure disposal technique (liquid or solid, removal and storage, manure drying on conveyor belt systems).  |
| Activity of animals (species, age, circadian rhythm, laying hens: cages or aviaries).  |
| Ventilation rate (season, mechanically or naturally ventilated, method of determination).  |
| Geometry and location of inlets and outlets (re-entrainment of particles caused by turbulences).   |
| Indoor microclimate (temperature, relative humidity).  |
| Duration of stay within the livestock building (whole year or seasonal housing, grazing period, all-in/all-out operation or continuous rearing system, length of service periods).                                   |
| Varying mutual and intermittent keeping of animals among livestock buildings located in one region (not all livestock buildings are equally occupied at a certain time, possible overestimation of total emissions). |
| Secondary sources (exhaust fumes of tractors or fuel-operated generators, secondary aerosol formation, i.e., via ammonia).   |
| Abatement techniques (oil sprinkling, waste air purification).   |

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Examples of dependencies for the determination of emissions are given in brackets (Seedorf and Hartung, 2001; modified).

extensively (e.g. suckler cows) can be as long as 257 days in Lower Saxony (Lüttich et al., 2003). Therefore, a housing period of 182.5 days seems to be an acceptable compromise.

A further adjustable, or tuneable, factor concerns broiler housing conditions, where there is scarcely any active ventilation during the first 10 days; therefore the expectable emissions are probably insignificant, whereas the high emission loads are particularly associated with the end of the fattening period. This was confirmed by Redwine et al. (2002), who observed very low emission quantities of particulate matter at the beginning of a production period, when the emission loads were only approximately 1–4% of those determined at the end of the fattening period. This observation is important, because the emission factors for broilers used in this study were determined close to the end of the fattening periods. The application of the broiler-related emission factor over the whole fattening period would therefore probably overestimate the emissions for particulate matter in the inventory.

There are two ways in which the methodology could and should be refined. First, there is no distinction made between housing types with slurry- or litter-based systems, although differences in emissions could be expected, because bedding material is a potential particle-releasing material. It would make sense to introduce separate emission factors for different housing types if this would make it possible to assign species and numbers of animals to specific slurry- or litter-based systems. But this information is obviously not available if the categories of animal types in Table 2 have to be used. The emission factors for beef cattle, calves and sows represent values determined in houses with and without litter, whereas most of the dairy cows, all weaners and fattening pigs were kept on slurry-based systems. Laying hens were obligatorily housed in cages with conveyor-belt systems for manure drying and broilers were kept on the floor with litter.

A further factor to be included in the methodology would be the effects of abatement techniques. Due to the European Directive on Integrated Pollution Prevention and Control (IPPC), abatement techniques are becoming more and more important. Biological waste air purification devices (e.g. biofilters) are already quite common in animal housing to permit siting of livestock buildings in areas with high animal densities. But more knowledge is necessary for the specification of their spatial distribution and reduction efficiency before such additional terms can be integrated in the methodology of emission inventories (Lükewille et al., 2001). This is especially relevant for bioaerosols, because their heterogeneous composition can lead to agent-specific reduction patterns or even to an enrichment within the clean exhaust gas (Seedorf et al., 1999; Seedorf and Hartung, 2002b).

#### 4.3. Relating the data to other emission factors

It is difficult to compare the emission factors proposed here with other sources of pertinent data. An emission factor of 1.3 g PM<sub>10</sub> per bird was determined per fattening period of a broiler flock in tunnel-ventilated broiler buildings in the southern US (Lacey et al., 2003). Total particulate load from US swine production has been estimated at nearly 1.6 kg per pig and year (Hatfield et al., 2002), but this is more than twice the averaged and extrapolated emission factors for sows, weaners and fattening pigs in Lower Saxony (see Table 1). It is obvious that there are differences in the definitions of emission factors (i.e. g time<sup>-1</sup> animal<sup>-1</sup> as opposed to g animal<sup>-1</sup>). In some cases, emission factors refer only to a species without any distinction among different livestock classes (e.g. pigs or sows, weaners or fattening pigs). Another important question is the transferability of the data to other regions. For example, the use of North American data for European purposes and vice versa is problematic. Among the confounding factors that place serious doubt on transferability are significant differences in climatic conditions, length of fattening period, housed breed, type of ventilation systems, quality of bedding material, farm management practices, and cleaning and disinfection regimes. The obviously higher emission factor for pigs from Hatfield et al. (2002) may be an indication of such non-transferability. This shows that verification and harmonisation of emission factors are highly desirable if emission factors are to be used in a broader spatial context.

At present, the emission factors for dust and endotoxins based on the studies of Takai et al. (1998) and Seedorf et al. (1998) seem to be the most comprehensive. This evaluation has recently been confirmed in an interagency emission factor report (TWP, 2001), in which it is pointed out that other studies under review lacked detailed supporting information and analysis of important variables that were considered in the emission factors. It was also pointed out that, due to the difficulty and variability of quantifying endotoxins, one should not place a great deal of weight on this kind of emission factor. Furthermore, the fact has to be taken into account that common endotoxin measurements are based on bioassays, in which non-estimable interfering reactions may influence the accuracy or alter the representativeness of analyses (Jacobs, 1997). Despite these limitations the TWP report selected the endotoxin emission factors of Seedorf et al. (1998) as the most representative available data. The emission factors presented here were originally linked to the 1998 studies of Takai and Seedorf. Therefore, the strengths and weaknesses of the data in both these publications will also be balanced in the German data pool presented here.

#### 4.4. Particle size and conversion factors

In addition to many other relevant airborne pollutants, agricultural primary particulate emissions in northwestern Europe have been calculated on the basis of statistics for animal populations in European countries and on the basis of emission factors for different animals. Furthermore, the spatial distribution of the emissions has been visualised as a map (Schaap et al., 2002). For that purpose most publications referred to the emission factors of inhalable and respirable dust according to Takai et al. (1998), because no specific  $PM_{10}$  and  $PM_{2.5}$  emission factors are yet available for all livestock classes. Nonetheless, occasionally inhalable and respirable particles are commonly referred to as  $PM_{10}$  and  $PM_{2.5}$ , respectively (Chetner and Sasaki, 2001). Such a definition may overestimate the emitted particle quantities, because Takai's dust measurement devices have sampled inhalable and respirable particles up to a 50% cut-off diameter ( $d_{50}$ ) of 100 and 5  $\mu m$ , respectively, while inhalable dust is also often used as equivalent to total suspended particulate matter (TSP).

To fulfil the requirements of the guideline IPPC (1996), environmental dust measurements have to take into account  $PM_{10}$  and  $PM_{2.5}$  to make the harmonisation of the relationships between emissions and receptor concentrations possible. However, according to temporary agreements allowing for national regulations it will still be legal until 31 December 2004 to use a conversion factor of 1.2 to transform amounts of  $PM_{10}$  yields into total dust concentrations (22. BImSchV, 2002). Consequently, a vice-versa procedure should also be possible by which conversion factors are used to transform any kind of dust emission data into  $PM_{10}$  and  $PM_{2.5}$  emission factors instead of real undertaken  $PM_{10}$  and  $PM_{2.5}$  measurements. This was the procedure used by Phillips et al. (2002), who derived a ratio of  $PM_{10}$  to total dust of 0.45 and a ratio of  $PM_{2.5}$  to total dust of 0.08 for pig buildings. Both conversion factors were based on the publication of Louhelainen et al. (1987). Recently, additional ratios were cited in a governmental report (MAFF, 2000), according to which 40% of the dust in pig buildings is  $PM_{10}$ , as is 70% in chicken buildings. The CEPMEIP database has proposed emission factors for TSP,  $PM_{10}$  and  $PM_{2.5}$ , according to which the ratios of  $PM_{10}$  to TSP and  $PM_{2.5}$  to TSP are 0.45 and 0.10, for poultry, cattle and pig livestock buildings, respectively (TNO, 2001). Other investigations have found ratios of approximately 56% between dust masses with particle sizes of  $\leq 9 \mu m$  and total dust in houses for rearing pigs, and ratios of nearly 39% in broiler livestock houses (Aarnink et al., 1999), while measured particle size distributions in a small-scale experimental turkey house were shown to be only 25%  $PM_{10}$  (Hinz et al., 1999). Redwine et al. (2002) found a very low proportion of  $PM_{10}$  in TSP samples from

broiler houses and determined a mean mass fraction of 5.94%  $PM_{10}$  in the TSP samples. Seedorf and Hartung (2001) have determined a conversion factor of 0.46 for cattle livestock houses. It is obvious that these conversion factors are quite heterogeneous, and the listed ratios indicate that the majority of studies did not directly measure amounts of emitted  $PM_{10}$  or  $PM_{2.5}$ . Nevertheless, theoretical emission factors for  $PM_{10}$  and  $PM_{2.5}$  have been proposed on the basis of conversion factors and are used in a regional air pollution information and simulation model that addresses present and future emissions of fine particulates in Europe. For this purpose, the  $PM_{10}$  emission factors in kg per animal and year are 0.0473 for poultry, 0.4376 for pigs and 0.4336 for dairy and other cattle. These factors correspond to 0.0105, 0.0778 and 0.0964  $PM_{2.5}$ , respectively (Klimont et al., 2002).

The application of conversion factors leads to virtual estimates of emissions with specific PM sizes, but in reality appropriate size distribution factors are not well-defined and are different for varying dust qualities, especially for different sampling and measurement methods. The latter point is quite crucial, because dual measurements of  $PM_{10}$  and TSP with an instrument with  $d_{50} = 100 \mu m$  will for example ultimately produce significantly lower conversion factors than a device with  $d_{50} = 35 \mu m$  under the same sampling conditions. But these are not the only circumstances which will clearly decrease the accuracy of predicted  $PM_{10}$  or  $PM_{2.5}$  emission inventories; there are also many other considerable uncertainty factors that can negatively affect the quality of an inventory (see Table 6). In consideration of these issues, it was decided to design the emission inventory for particulates on the basis of original available data for inhalable and respirable dust to ensure the best possible reliability.

#### 4.5. Environmental aspects

The significance of atmospheric dust in environmental hygiene is the fact that increased incidences of human mortality and morbidity are associated with elevated levels of particulate air pollution. These effects are mainly due to the physical properties of particulates, but their chemical and microbiological composition may also be of environmental concern. This is especially valid for the organic livestock-related dust and its function as a vector for a huge range of components (endotoxin, mycotoxin, allergen, etc.) with biological effects (Seedorf and Hartung, 2002a).

First indications of such effects were reported by Monn and Becker (1999), who suggested that endotoxins and/or gram-negative bacteria (e.g. *Enterobacteriaceae*) in outdoor dust cause proinflammatory effects on macrophages in the respiratory tract. These results would support the hypothesis that endotoxin and



microorganism-rich livestock dust may contribute to the overall atmospheric dust burden, which was generally confirmed by observations of  $PM_{10}$  from swine buildings in central Iowa, where there was an increase above ambient levels but not above air quality standards (Hatfield et al., 2002). On the other hand, it is assumed that the level of environmental exposure to endotoxins and other bacterial wall components may be an important protective determinant against the development of atopic diseases in childhood (e.g. von Mutius et al., 2000). Furthermore, it remains unclear whether the emitted non-viable and viable dust amounts from livestock buildings are masked by the much greater dust releases supplemented with biogenic material from industry and traffic. Due to atmospheric transportation and mixed dust qualities, it will not necessarily be possible to distinguish clearly between the most potent emission sources and their specific impact on any environmentally relevant target. As mentioned above, detailed dust analyses may isolate key components and/or composition patterns of compounds indicating the contribution of livestock operations to primary particulates in the ambient air.

Fungi, which comprise a considerable part of livestock-related bioaerosols, are ubiquitous in nature. Dose–response relationships for specific fungi can be used for the evaluation of their allergic and toxic potency and can serve as warning measures in regions where exposure to a particular fungus is high (Downs et al., 2001). But it is difficult to decide whether naturally occurring fungi or anthropogenic fungal emissions are responsible for alterations in public health. From a qualitative point of view, a further question remains open as to whether human-related emission activities can alter the ratios of individual species in the environment (Fischer and Dott, 2003). In one scenario, interspecies shifts are conceivable involving increased fungal parasitic affinities to plants or a changed environmental allergen spectrum, which would affect allergic individuals. Such speculations can only be ruled out if the results of comprehensive chemical and microbiological analysis of dust samples in a spatial context are available in future to fill these gaps in our knowledge.

In addition to research on typical dust components like microorganisms and endotoxins, there is also a current focus on antibiotics in livestock dust as a new entrance route into the environment (as well as the known route of soil fertilisation with liquid manure contaminated with antibiotics) (Hamscher et al., 2003). The entry of antibiotics into the environment enhances the spread of antibiotic resistance (Witte, 1998) and may provoke long-term ecologically adverse effects. In this context, there is a need for the study of the role of microorganisms carrying resistant genes and cross-species spread of resistance by plasmid transfer.

## 5. Conclusions

For many decades, agriculture-related gases such as ammonia have been considered to be important atmospheric and terrestrial pollutants. Enhanced environmental protection regulations also led to the discussion of the impact of emitted bioaerosols on ecological and human systems and what their contribution to atmospheric pollution may be. Despite existing data gaps and lack of reliable calculation models, the methodology proposed here for an emission inventory of livestock-related bioaerosols is a first step toward an overview of emitted bioaerosol loads which will hopefully initiate the creation of further alternative, extended and improved calculation models. Despite several uncertainty factors, the calculation procedure presented here gives at least a provisional idea of how spatially emission loads can be estimated if livestock class-related production cycles are considered. Future work should focus on a higher spatial resolution, verification of the emission factors (i.e.  $PM_{10}$ ,  $PM_{2.5}$ ) and upgrading of the calculation models in terms of necessary farm-related and process-based factors. With the knowledge of farm localisations and the related animal production conditions, it should also be possible to calculate a mesoscale and a macroscale distribution pattern of gridded primary bioaerosol emissions. Furthermore, monitoring campaigns are urgently needed for the whole production cycle and the inclusion of emission activities relevant to the outdoors, because intensified animal welfare laws and the trend toward organic farming are having the effect of increasingly shifting the source of total emissions from livestock buildings to the free-range areas where animals are kept. Additionally, it is highly desirable that emission factors for other farm animals (turkeys, ducks, geese, horses, sheep, goats) be available to make it possible to close the gaps in prospective inventories. Despite the missing emissions factors, the emission inventory presented here provides a conservative estimation of bioaerosols released into the atmosphere of Lower Saxony, Germany, which may help to assess the environmental risks of livestock farming.

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