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Particulate matter emissions during field application of poultry manure - The influence of moisture content and treatment

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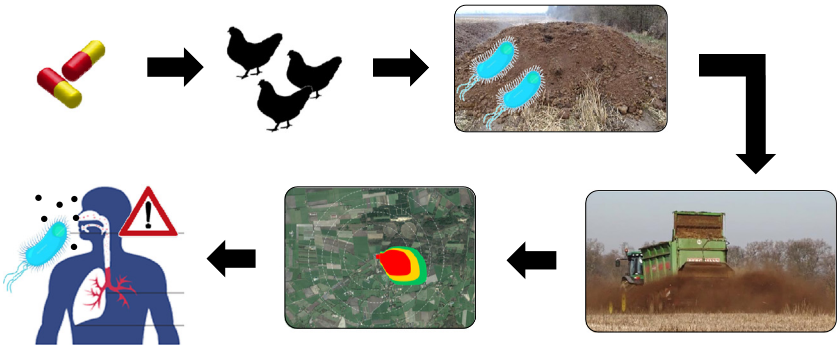
HIGHLIGHTS

- Microbial abundance decreases with increasing manure dry matter content (DM).
- High manure DM causes elevated PM emissions during field application.
- Maximum PM concentration during manure application was 10 mg PM per m^3 of air.
- Optimal DM range of poultry manure for field application is 50–70%.
- Modeling revealed a low risk for distances >400 m from the application site.

GRAPHICAL ABSTRACT



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ABSTRACT

Along with industry and transportation, agriculture is one of the main sources of primary particulate matter (PM) emissions worldwide. Bioaerosol formation and PM release during livestock manure field application and the associated threats to environmental and human health are rarely investigated. In the temperate climate zone, field fertilization with manure seasonally contributes to local PM air pollution regularly twice per year (spring and autumn). Measurements in a wind tunnel, in the field and computational fluid dynamics (CFD) simulations were performed to analyze PM aerosolization during poultry manure application and the influence of manure moisture content and treatment. A positive correlation between manure dry matter content (DM) and PM release was observed. Therefore, treatments strongly increasing the DM of poultry manure should be avoided. However, high manure DM led to reduced microbial abundance and, therefore, to a lower risk of environmental pathogen dispersion. Considering the findings of PM and microbial measurements, the optimal poultry manure DM range for field fertilization was identified as 50–70%. Maximum PM_{10} concentrations of approx. 10 mg per m^3 of air were measured during the spreading of dried manure (DM 80%), a concentration that is classified as strongly harmful. The modeling of PM aerosolization processes indicated a low health risk beyond a distance of 400 m

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from the manure application source. The detailed knowledge about PM aerosolization during manure field application was improved with this study, enabling manure management optimization for lower PM aerosolization and pathogenic release into the environment.

1. Introduction

The emergence of health issues related to the exposure to particulate matter (PM) is highly relevant for humans and the environment. PM has been ascertained to cause a variety of severe health effects by inhalation, that mostly affect the pulmonary and cardiovascular system (Anderson et al., 2012; Burnett et al., 2014; Lelieveld et al., 2015; Pope III et al., 2004). The health risks considerably depend on the inhaled PM size, concentration, composition and duration of the exposure. It is estimated that 90% of the world population is at least temporarily exposed to high PM levels, which causes approx. 4.2 million premature deaths each year (WHO, 2018). The size of airborne particulates is crucial for its transportation characteristics and significantly influences the inhalation depth into the respiratory system. Fine PM (<2.5 μm) generally penetrates into deeper sections of the lung compared to coarse particles. PM is classified according to its aerodynamic diameter mainly into the following three classes: PM_{10} (particles <10 μm), $PM_{2.5}$ (<2.5 μm) and PM_1 (<1 μm) (Lai et al., 2014). In addition to the health risk that originates from the particles themselves, PM possesses a carrier function due to its sorption capability regarding adhesive components, such as undesirable gases (odors, NH_3), bioactive chemicals (endotoxins, antibiotics) and microorganisms (bacteria, fungi, viruses) (Cambra-López et al., 2010; Mostafa et al., 2016; Schaeffer et al., 2017). Therefore, PM containing biologically active ingredients is described as bioaerosol.

There are two types of PM particles: primary particles, which are directly emitted and secondary particles, which are created indirectly by chemical reactions of different gaseous emissions within the atmosphere (Després et al., 2012; Pozzer et al., 2017). In addition to transportation, households and industry, agriculture is one of the largest anthropogenic producers of primary PM and account for approx. 20% of the global fine dust emissions, making it the largest $PM_{2.5}$ polluter in Russia, USA, Europe, and East Asia (Aarnink & Ellen, 2007; Lelieveld et al., 2015; Umweltbundesamt, 2017). Agrarian PM arises mainly from fertilizer application, livestock houses, field cultivation operations and subsequent wind erosion (Funk et al., 2008; Hoffmann and Funk, 2015; Maffia et al., 2020; Takai et al., 1998). Within livestock production, PM is mainly emitted from pig and poultry houses, contributing 30% and 50%, respectively, to the total PM emissions from agriculture in Europe (Cambra-López et al., 2010). In regions with an enhanced livestock production density, an increased PM prevalence and concentration of air suspended biological components have been observed, especially downwind from livestock farms (Lonc and Plewa, 2011; McEachran et al., 2015; Winkel et al., 2015). PM concentrations inside poultry houses can be up to 200-fold higher than concentrations in ambient air (Aarnink and Ellen, 2007; Lai et al., 2014; Winkel et al., 2015). Poultry manure is composed of feathers, feed, bedding material and excreta and has a proportion of organic matter of >90%. Excreta contribute mostly to the composition of PM in poultry housings. In detail, 30–83% of PM_{10} and 14–68% of $PM_{2.5}$ in poultry housings consist of excreta (Cambra-López et al., 2011). Since animal production buildings contribute significantly to PM and bioaerosol emissions, an increased health risk arises for people living near livestock houses (Dungan, 2010; Pozzer et al., 2017; Takai et al., 1998).

Due to the extensive use of antibiotics in livestock farming (4.7 million and 10 million kg a^{-1} in the EU and the USA, respectively), there is a high selection pressure in favor of antimicrobial-resistant (AMR) microorganisms in the intestinal tracts of farm animals (Hamscher et al., 2002; McEachran et al., 2015; Singer et al., 2016). The airborne spread of antibiotics and AMR microbes was ascertained in connection with PM measured downwind from animal houses and livestock feed yards (Cambra-López et al., 2010; Dungan, 2010; McEachran et al., 2015). Livestock manure, containing AMR microbes and antibiotic residues (Sarmah et al., 2006), is recycled as organic fertilizer on arable land to combine sustainable disposal with closing nutrient cycles. Therefore, the antibiotics contained in manure can accumulate in fertilized soil and can be transported and inhaled in agglomeration with PM (Hamscher et al., 2003; Schmitt et al., 2006; Thomas et al., 2019). While manure is a valuable fertilizer (Leip et al., 2019), care must be taken to avoid gaseous environmental pollution during manure storage and after application (Hutchings et al., 2020; Groenestein et al., 2019), which can be minimized by manure drying and composting (Amon et al., 2006; Mohankumar Sajeev et al., 2018). In Germany and the U.S., approx. 1 million and 44 million tons, respectively, of manure accrue from poultry production and are applied on agricultural land each year (Bolan et al., 2010; Siller et al., 2020; Statistisches Bundesamt, 2016). Regarding the globally increasing demand for poultry meat products and eggs (OECD, 2018; López-Mosquera et al., 2008), it can be assumed that PM emissions from poultry livestock and the associated agrarian processes using poultry manure will be of increasing relevance to air pollution in the near future.

Field spreading of manure bears a twofold health risk due to elevated PM levels and the distribution of attached, potentially harmful microorganisms around the application site. Poultry manure is characterized by an especially elevated PM emission potential due to its low density and high dry matter content (DM 50–70%) compared to those of dairy and pig manure, thus promoting aerosolization and air suspension (Hartung and Saleh, 2007; Kabelitz et al., 2020; Mostafa et al., 2016). In contrast to bioaerosol emergence from animal housings, little is known about emissions during manure field application and the associated threats to environmental and human health (Maffia et al., 2020; Münch et al., 2020; Thiel et al., 2020). Therefore, this study aims to quantify and characterize PM emissions from poultry manure field application; especially investigating the influence of manure moisture content on aerosolization rates and vertical and horizontal PM dispersion.

2. Materials and methods

2.1. Manure material

Poultry manure was collected from two broiler farms in Germany. At both farms, broilers were kept under similar conventional conditions in a floor housing system with wood pellets as bedding material and dry feed. The manure was solid, dry, and crumbly in consistency, and was mainly composed of feces, feathers, and litter (Fig. S1A). In all experiments, four broiler manure treatments were investigated to quantify the influence of manure moisture content and handling. 'Untreated' manure was freshly collected in broiler houses and had a DM of approx. 49%. Therefore, it was the treatment with the highest moisture content and the lowest age. 'Stored' manure was heaped manure with an age of circa 10 weeks and a DM of approx. 59% (Fig. S1B). 'Composted' manure was similar to stored manure but additionally mixed with an excavator once per week. It was characterized by a DM of approx. 52%. 'Dried' manure was freshly collected and immediately heated for 48 h with a hot air dryer resulting in a DM of approx. 80%.

2.2. Physical and chemical parameters

The manure DM and organic matter content (OM) were calculated by the weight difference before and after heating for 24 h at 105 °C and 5 h at 550 °C, respectively. Electric conductivity (EC) and pH value were measured with a pH meter (WTW pH 3210, Weilheim, Germany). Analyses of nitrogen (N) and ammonium-nitrogen (NH_4-N) concentrations were conducted by steam distillation according to Kjeldahl using KjelMaster K-375 (BÜCHI Labortechnik GmbH, Essen, Germany) following the manufacture's instructions. Phosphate (P) content was determined by photometric flow injection analysis (FIA) using samples obtained after Kjeldahl disintegration (ISO 15681-1). Analyses for carbon (C), sulfur (S), and hydrogen (H) were performed by combining high-temperature combustion and gas chromatography using an elemental analyzer vario EL (Elementar Analysensysteme GmbH, Langenselbold, Germany).

2.3. Storage experiments

For the storage experiments, 45 kg of untreated and composted manure were filled into 90 L covered plastic containers (air permeable) and stored for 12 weeks. Physicochemical parameters were recorded regularly: first, every 2 days (on days 0, 3, 5, 7), then weekly (on days 14, 21 and 28), and finally, every second week (on days 56, 70 and 84). The temperature was measured automatically once per hour with a MicroLite II temperature logger (FOURTEC, Rosh Ha'ayin; Israel). The only difference in treatment between the untreated and composted manure was the mixing of the latter on days 7, 14, 21, 28 and 56. The storage experiments were performed in three replicates per treatment from Nov 2017 to March 2018.

2.4. Microbiological analyses

For microbiological analysis, manure was filled into plastic bags and diluted 1:10 in liquid LB media (Carl Roth GmbH + Co. KG, Karlsruhe, Germany). The sample was homogenized for 2 min at 200 rpm with a Stomacher® (Bagmixer 400, Interscience, France) and incubated for 30 min at room temperature (approx. 20 °C). The obtained suspension was 10^1- to 10^6-fold diluted afterwards. To determine the amount of total cultivable bacteria, dilutions were plated onto Columbia blood agar and for the detection of enterococci on Kanamycin aesculin azide agar (both Oxoid Deutschland GmbH, Wesel, Germany). For *Enterobacteriaceae* and *Escherichia coli,* samples were plated on MacConkey Agar No. 3 (Oxoid), and for extended-spectrum beta-lactamase (ESBL)-producing *E. coli,* 1 mg L^{-1} cefotaxime was added. The quantification of *Clostridioides difficile (C. difficile),* methicillin-resistant *Staphylococcus aureus* (MRSA), and vancomycin-resistant *Enterococcus* (VRE) was performed on commercially available species-specific selection media (CHROMagar, Paris, France). For all microorganisms except *C. difficile*, Colony-forming units (CFUs) were counted after aerobic incubation for 24 h at 37 °C. CFUs of *C. difficile* were counted after anerobic incubation for 48 h at 37 °C.

2.5. Particle size distribution (PSD) analyses

The PSD of 100 cm^3 manure was measured by dynamic image analysis using a PartaAn 3001 L 3D analyzer (Microtrac Inc., Montgomeryville, USA). 100 pictures per s were made with an integrated high-speed camera and subsequently used to reconstruct a 3D projection of every manure particle, delivering informations about particle length, width, thickness, perimeter, and area. Particles from 3 to 100 μm, in 1 μm steps, according to their area equivalent diameter, were counted. For graphical presentation, particle counts of different sizes were summarized as indicated (Fig. 1). The total particle numbers of the 16–95 μm class were divided by 16 because the size range of this class is 16-fold higher than that of the other classes. Ten replicates were measured for every DM level.

2.6. PM emission measurements under field conditions

PM emissions released from solid manure application under practical conditions were measured with two field experiments, both performed in Brandenburg on the 27th of March and on the 31st of May 2017. Manures of the four treatments were applied with a Bergmann manure spreader TSW 6240 S (Ludwig Bergmann GmbH, Goldenstedt, Germany) or a Strautmann BE4 manure spreader (Strautmann & Söhne GmbH & Co. KG, Bad Laer, Germany). A field plot of 60 × 300 m was fertilized with approx. 15 t of each manure treatment (Fig. S1D). The resulting PM concentrations were measured with two GRIMM EDM 164 dust analyzers (GRIMM Aerosol Technik GmbH & Co. KG, Ainring, Germany) at heights of 1.5 and 3.8 m (Fig. S1E). Particles in a size range of 0.25–32 μm were counted every 6 s with an airflow rate of 1.2 L min^{-1}. Afterwards, PM concentrations for PM1, PM2.5 and PM10 were calculated in μg per m^3 of air. The distance of the PM measurement was on average 50 m downwind from the application site (yellow squares in Fig. S1D), and in the second field experiment additionally 20 and 100 m. For each manure treatment, three replicates were measured consecutively with a 10–15 min break in between. PM background levels were recorded in ambient air prior each manure application. Throughout the whole experiment, temperature, relative humidity, wind speed, and wind direction were recorded by two weather stations and anemometers (heights: 0.8 m and 2.8 m). On the 27th of March, the average weather conditions during the field experiment were as follows: wind speed: 2 m s^{-1}; wind direction: south-west; temperature: 22 °C; and air humidity: 27%. On the 31st of May, the field experiment was performed under the following average weather conditions: wind speed: 6 m s^{-1}; wind direction: north-west; temperature: 22 °C; and air humidity: 60%. The detailed weather conditions are shown in Table S1. Top view photos of visible dust generation during poultry manure application were made with a GoPro action camera installed on a drone (Fig. S1C). PM concentration measurements are shown over the time of manure application in Fig. 2B and as sum of total emissions in Fig. 3A. PM values shown in both figures were normalized to the background level.

2.7. Modeling

The PM transmission during field experiments was modeled using computational fluid dynamics (CFD). A one-way coupled Eulerian-Lagrangian solver was used to solve the flow field and the transport of PM simultaneously. For the flow field, large eddy simulations were applied with a one-equation subgrid-scale model. The model was validated in a previous study and is described in Janke et al. (2020). The transport of PM was modeled using the OpenFOAM Lagrangian particle-tracking library, which is described in Kasper et al. (2019). The computational domain had a width of 150 m, a length of 500 m, and a height of 80 m. At the inlet, a turbulent inflow was generated with the "Divergence Free Synthetic Eddy Method", described in Poletto et al. (2013). The inflow profile met the requirements of a neutral atmospheric boundary layer on a moderately rough terrain according to (Ingenier, 2000). Particles were released 50 m downstream from the inlet at a height of 1.5 m from three points at the symmetry line of the width. The size distribution and density of the particles injected into the domain were set to the values reported in Kabelitz et al. (2020). After the simulation, the mean concentrations of PM1, PM2.5 and PM10 particles in the domain were calculated.

3. Results & discussion

3.1. Physicochemical manure properties

For PM emission measurements on the field, four poultry manure treatments were investigated, representing different commercially relevant manure management strategies. The differences in the physical and chemical properties of the untreated, composted, stored and dried manure were determined (Table 1). Most measured values fall within the ranges reported in literature (López-Mosquera et al., 2008; Bolan et al., 2010). The DM of untreated, composted and stored manure was lower and the carbon (C) content was slightly higher than the reported literature values. However, the parameters strongly depend on diverse

Table 1

Physicochemical characterization of poultry manure treatments.

The physical and chemical properties of untreated, composted, stored and dried broiler manure are compared. Units are indicated in square brackets. As a measure of treatment variability, the SD [%] column shows the percent standard deviation between values. The literature values (last column) for EC were determined in a 1:5 water mixture. Therefore, they were multiplied by 5 to make them comparable to our results.

	Untreated	Composted	Stored	Dried	SD [%]	López-Mosquera et al. (2008) Bolan et al. (2010)
DM [% FW]	48.86	52.41	59.09	79.80	±23	69.4–80.5
EC [mS $*$ cm^{-1}]	49.80	59.90	44.00	52.60	±13	31.5–63
pH	7.40	5.94	6.54	6.57	±9	6.3–8.4
C [% DM]	42.87	42.19	43.15	39.38	±4	29.3–38.8
N [% DM]	4.31	4.60	4.82	3.98	±8	2.6–5.3
C/N ratio	9.94	9.17	8.95	9.89	±5	6.4–11.8
S [% DM]	0.63	0.67	0.67	0.53	±10	0.2–0.8
H [% DM]	6.44	6.42	6.51	5.91	±4	n.d.
P [g $*$ kg^{-1} FW]	5.62	4.78	5.06	7.30	±20	6.7

Abbreviations: not determined (n.d.), dry matter content (DM), fresh weight (FW), electric conductivity (EC), carbon (C), nitrogen (N), sulfur (S), hydrogen (H), and phosphate (P) content.

physiological, environmental and management factors, which can considerably differ between various broiler farms. The DM was the parameter that showed the largest standard deviation (SD 23%) between different manure treatments. It was lowest in untreated samples (49%), followed by composted (52%) and stored (59%) manure. Dried samples showed the highest DM (80%). Other parameters that revealed large variations between manure treatments were the phosphorus content (P) (SD 20%), electric conductivity (EC) (SD 13%) and the sulfur content (S) (SD 10%). There were no major differences in the hydrogen (H) content, the C content and the C/N ratio between manure treatments. Minor differences occurred for the pH and the organic nitrogen (N) content. Except for the DM, the analyzed manure treatments differed slightly in their physicochemical properties and, consequently, in their fertilizer quality. Note, no clear correlation between parameter fluctuations and DM was found.

3.2. Microbiological manure properties

Not only PM emissions themselves represent a threat to public health. PM particles can be associated with pathogenic microorganisms. Therefore, the influence of manure treatment on the prevalence and survival of microorganisms was analyzed. Here, we focused on representative antibiotic-resistant and pathogenic bacteria (Table 2). For each manure treatment, the concentrations of total bacteria (grown on blood agar), representative fecal bacteria (*Enterococcus* spp., *Enterobacteriaceae, Escherichia coli*), AMR bacteria (ESBL-producing *E. coli,* vancomycin-resistant enterococci = VRE, methicillin-resistant *Staphylococcus aureus* = MRSA), and pathogenic *Clostridioides difficile* were analyzed. The prevalence of total cultivable bacteria in untreated, fresh poultry manure was approx. 10^{10} CFU g^{-1} and in full agreement with reported literature values of 10^8–10^{11} CFU g^{-1} (Bolan et al., 2010; Jahne et al., 2015; Thiel et al., 2020). Microbial activity decreased with the DM, to as low as 10^6 CFU g^{-1} in dried manure. Enterococci and *Enterobacteriaceae* showed the same reduction tendency of three and two orders of magnitude, respectively, between untreated and dried manure. Enterobacteria were only detected in untreated and dried manure because fresh broiler litter was used for these two treatments. As shown in Fig. S3, upon excretion, *Enterobacteriaceae* in manure died within the first 2 weeks of storing. *E. coli* and *C. difficile* were detected qualitatively in all samples. We could not verify the presence of ESBL-producing *E. coli*, VRE and MRSA in any sample. In general, there was a clear correlation between the manure DM and bacterial survival. The drying of manure led to a strong bacterial reduction of several orders of magnitude. This result indicates that the higher the manure DM is, the lower the abundance of microorganisms and, thus, the potential health risk from pathogens.

Aerosolized bacteria from livestock houses were >90% gram-positive and dominated by *Staphylococcus* and *Streptococcus* species (Cambra-López et al., 2010). Depending on the farm animal species, gram-negative bacteria made up a proportion of only 0.02–5.2% and mainly contained *Pseudomonadaceae*, *Neisseriaceae*, and *Enterobacteriaceae* (Zucker et al., 2000). Several bacteria have been shown to be able to survive in manure for long time periods (Bradford et al., 2013). Hartmann et al. (2012) and Friese et al. (2013) showed that ESBL-producing *E. coli* and MRSA were transmitted from livestock houses to arable land and were able to persist for over 1 year in the soil. Land fertilization with manure increases the organic matter content of the soil, which can lead to an increased bacterial survival (Acosta-Martinez et al., 2015).

3.3. The influence of storage on manure properties

Manure storage has a strong effect on manure properties, such as moisture content, temperature, fertilizer quality (chemical composition), and microbiological activity. All these factors are influencing the PM emission potential of manure, especially of the here used untreated and composted manure material. During a three-month storage experiment in winter, temperature, microbial tenacity, and physicochemical parameters of untreated and composted manure were analyzed regularly. For the temperature, a strong correlation between manure and ambient temperature was observed (Fig. S2). Due to microbial activity, temperatures of manure were always higher than ambient temperatures and reached their maximum in the dung heap center. Both manure treatments showed a stable amount of total cultivable bacteria of approx. 10^9–

Table 2

Microbiological characterization of poultry manure treatments.

The mean frequency of indicated microorganisms is provided in colony forming units (CFUs) per g of manure.

CFU g^{-1}		Blood agar counts	Enterococci	*Enterobac-teriaceae*	*E. coli*	ESBL-producing *E. coli*	VRE	MRSA	*C. difficile*
Manure	Untreated	10^{10}	10^7	10^4	+	−	−	−	+
	Composted	10^9	10^6	10^3	+	−	−	−	+
	Stored	10^9	10^6	10^3	+	−	−	−	+
	Dried	10^6	10^4	10^2	+	−	−	−	+

Bacteria that could not be detected are labeled with a "−" and bacteria that have been detected quantitatively but not qualitatively are marked with a "+". Abbreviations: *Escherichia coli (E. coli),* extended-spectrum beta-lactamase (ESBL), vancomycin-resistant *Enterococcus* (VRE), methicillin-resistant *Staphylococcus aureus* (MRSA), *Clostridioides difficile (C. difficile)*, and not determined (n.d.).

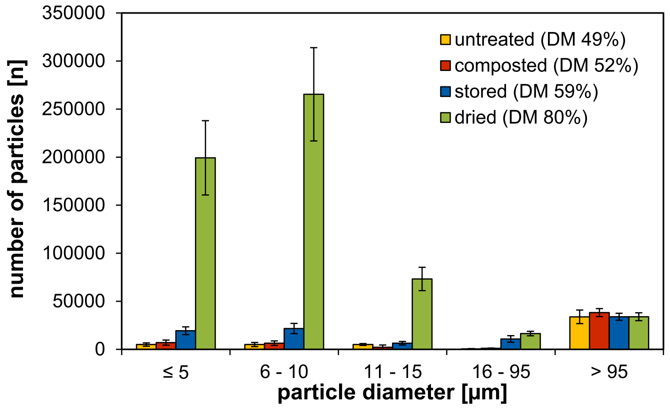


Fig. 1. Manure particle size distribution (PSD). Number of particles in indicated size classes per 100 cm^3 of used poultry manure with four dry matter contents (DM). Error bars are the standard deviation (SD) of ten replicates.

10^{10} CFU g^{-1} over the complete storage period (Fig. S3). The concentration of enterococci decreased by one log_{10}-step (from 10^7 to 10^6 CFU g^{-1}) over the three month storage. The survival of Cefotaxime-resistant Enterobacteria was approx. 10^4 CFU g^{-1} at the beginning and decreased rapidly to 10^2–10^3 CFU g^{-1} after 1 week. Enterobacteria were not viable anymore after 2 weeks of storage. Developments of diverse chemical and physical parameters during the manure storage experiment are shown in Fig. S4 and summarized in Table S2.

3.4. Manure particle size distribution (PSD)

To determine whether and how the particle size composition of solid manure differs between different treatments and DMs, the PSDs of the four manure treatments were measured. Untreated, composted, stored and dried broiler manures with a DM of 49–80% were investigated (Fig. 1). The threshold where manure dissociated into a larger part of particulate matter particles (<10 µm) than nonrespirable particles was at a DM between 60% and 80%. The amount of large particles (>95 µm) was comparable for the four different manure treatments with indicated DMs. In contrast, the highest differences were observed for small particles (<15 µm), especially in dried manure. Manures with a low DM (untreated, composted and stored) showed larger and fewer particles than the dried manure with a high DM, that was consisted of smaller and more particles. This negative correlation of DM and average particle size may be caused by increased particle coagulation, meaning that at a high moisture content small particles stick together and build larger agglomerates (Aloyan et al., 1997). With an increased manure DM the average particle size will be reduced and *vice versa*. Concluding that dry manure with a low DM has an enhanced PM aerosolization potential.

3.5. PM emissions during manure field application

The PM concentrations during the field application of untreated, composted, stored and dried manure were investigated (Fig. S1). The resulting dust cloud and manure aerosolization were already visible to the naked eye (Fig. 2A). In general, the measured PM emissions in the field were always fluctuating due to inconstant experimental conditions, such as a permanently changing wind direction and speed. A release of PM particles during manure spreading could be detected for each manure treatment and DM (Fig. 2B). The amounts of emitted PM particles, measured approx. 50 m downwind from the application site, were nearly identical during the spreading of untreated, composted and stored manure (DM 49–59%). The average concentrations of PM_{10}, $PM_{2.5}$ and PM_1 were approx. 150, 20 and 8 µg per m^3 of air, respectively. In contrast, the PM aerosolization was much higher when dried manure (80% DM) was applied. Here, the maximum PM_{10}, $PM_{2.5}$ and PM_1 concentrations

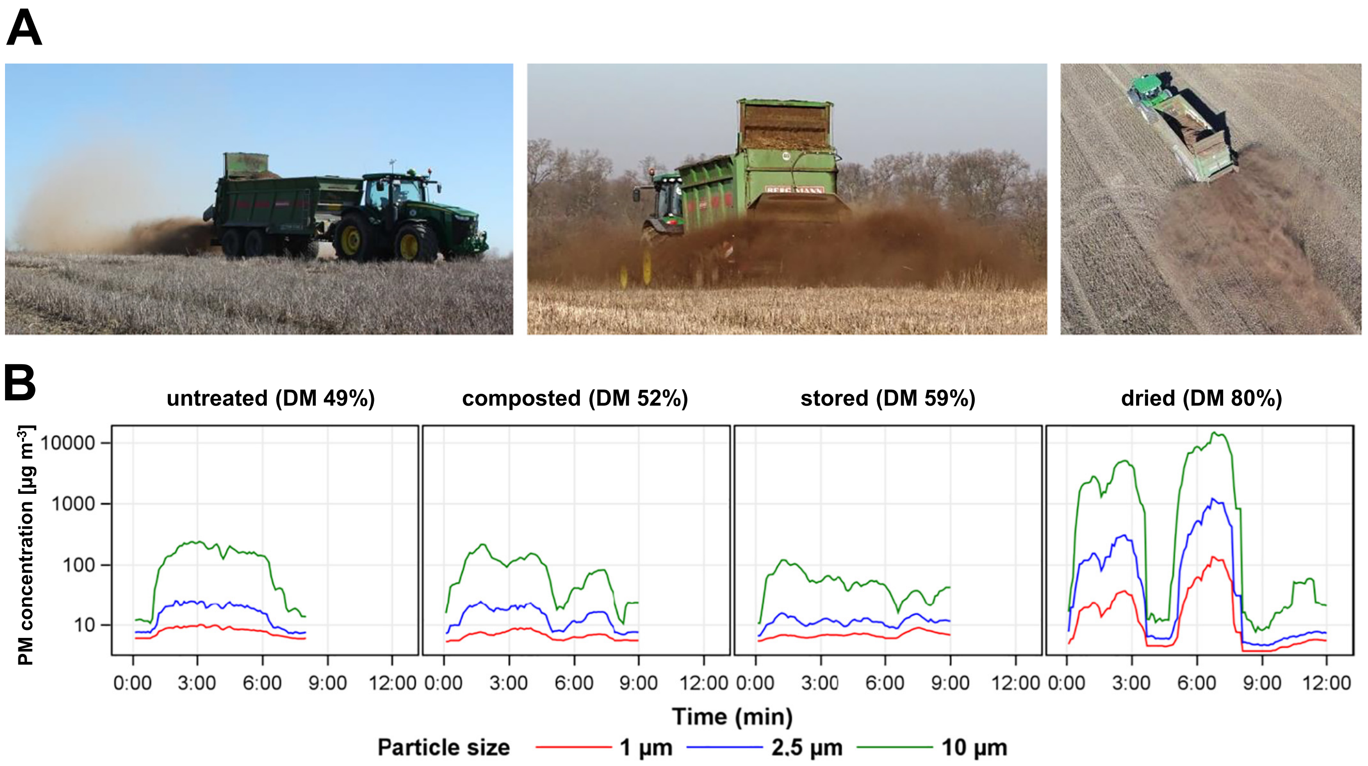


Fig. 2. PM emissions during poultry manure application. (A) Side, back and top view photo of visible dust cloud generation during poultry manure field application. (B) Means of PM concentrations (in µg of PM per m^3 of air) for class PM_1 (red), $PM_{2.5}$ (blue) and PM_{10} (green) from three replicates of four poultry manure treatments over the application time. PM concentrations shown, are averaged from measurements at 1.5 m and 3.8 m height.

were approx. 10,000, 600 and 50 μg m^{-3} air, respectively. Based on these concentrations, Münch et al. (2020) calculated PM emission factors, using Gaussian dispersion models, of 0.05–0.15 kg PM_{10} per ha for untreated, stored and composted manure; and 8.37 kg ha^{-1} for dried manure. Thus, the emission factor for dried manure is 55- to 167-fold higher. In conclusion, manure treatments that cause strong DM changes seemed to have a strong effect on the emitted PM during application. Our findings identified a strong positive correlation between the manure DM and PM release. This result is in full agreement with the observations of the PSD analyses (Fig. 1), where a large amount of small particles (<10 μm) was found in dried manure. In 2005, governmental guidelines determined annual and daily PM threshold values to protect environment and public health. According to these, the daily average concentration for PM_{10} should not exceed 50 μg m^{-3} of air (Aarnink & Ellen, 2007; Winkel et al., 2015). The annual PM average concentration limits for PM_{10} and $PM_{2.5}$ were defined with 40 and 25 μg m^{-3}, respectively. According to professional evaluation and medical knowledge, concentrations of $PM_{10}/PM_{2.5}$ up to 50/12 μg m^{-3} are of a low health risk, concentrations from 50-150/12-35 μg m^{-3} pose a moderate risk, concentrations from 150-400/35–250 μg m^{-3} are unhealthy, and concentrations >400/250 μg m^{-3} are hazardous. Thus, the PM concentrations measured during the field application of untreated, composted and stored manure were of moderate risk, but the threat level during dried manure application was hazardous. However, manure application occurs on average only twice per year, and the high measured PM concentrations were close to the application site (50 m distance). Health risks are probably limited to farmers spreading the manure and people living near the fertilized field. It can be assumed that PM concentrations will significantly decrease with distance from the emission source (Thiel et al., 2020). Therefore, we conclude that the relevance of manure application to annual PM air pollution is probably low but that it contributes to local and seasonal peaks.

As described before (Section 3.2), a high manure DM leads to a low microbial activity and increased PM emissions and *vice versa*. Consequently, bacterial survival and PM generation are in opposite relationship to each other. The optimal manure DM is a tradeoff between microbial abundance and PM emission potential. In a previous publication, we defined the optimal DM range for poultry manure under standardized wind tunnel conditions between 55 and 70% DM (Kabelitz et al., 2020). The present study confirms that the same optimal DM range is true for field conditions. Similarly, Bolan et al. (2010) found the optimal DM range of poultry manure for composting to be at 45–55%.

Manure has been shown to harbor diverse antibiotic-resistant and pathogenic bacteria (Blaustein et al., 2015; Bradford et al., 2013), which can be aerosolized and transported by PM emissions during manure application (Dungan, 2010; Siller et al., 2021; Thiel et al., 2020). It is estimated that >25% of atmospheric particles are composed of organic matter and microorganisms (Jones and Harrison, 2004). The bacterial abundance in ambient air was determined to be 10^4 CFU m^{-3} on average (Burrows et al., 2009a; Després et al., 2012). After manure spreading, the concentration of aerial bacteria was 10^5–10^8 CFU m^{-3} and thus one to four orders of magnitude higher (Boutin et al., 1988; Hobbs et al., 2004; Jahne et al., 2015; Münch et al., 2020; Thiel et al., 2020), strongly arguing for a microbial aerosolization during manure application. Microorganisms naturally survive better as parts of aggregates due to higher protection against damaging environmental influences (heat, UV radiation, and dryness). They were detected particularly often in aggregates with particle sizes of 3–12 μm (Després et al., 2012; Jones and Harrison, 2004; Madsen et al., 2018), which means that coarse PM (2.5–10 μm) is likely the particle size class with the highest microbial health risk. Aggregation with coarse PM enable microbes aerial residence times for several weeks and airborne transport over thousands of kilometers (Burrows et al., 2009b; Griffin, 2007; Hervàs et al., 2009; Maki et al., 2019). This can expose a huge number of people to increased levels of livestock-associated PM and pathogens, especially in the area around the application site (Schultz et al., 2019).

3.6. Horizontal and vertical PM profiles

Poultry manure with 70% DM was used to determine the horizontal (20, 50 and 100 m distance from the manure application source) and

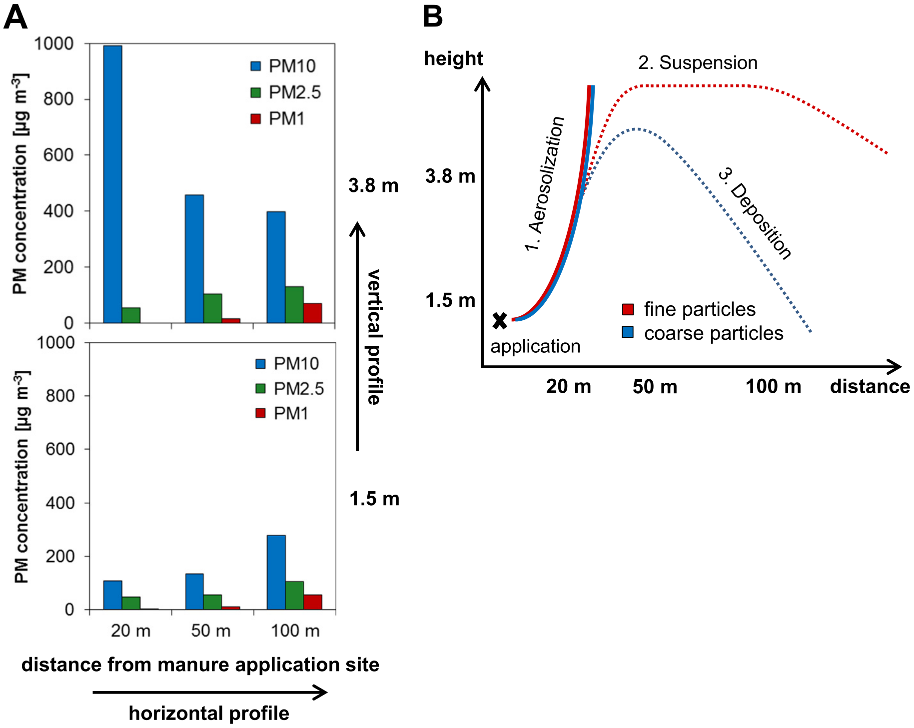


Fig. 3. PM emission profiles. (A) Total PM concentration (sum of all measured concentrations during the application normalized to the background, in μg of PM per m^3 of air) with increasing horizontal and vertical distance from the manure application site for class PM_1 (red), $PM_{2.5}$ (green) and PM_{10} (blue). (B) Schematic model for different flight characteristics of fine (red) and coarse (blue) particles after aerosolization. The manure application source is symbolized by a cross.

vertical (1.5 and 3.8 m height) profile of airborne PM concentrations during spreading (Fig. 3A).

3.6.1. Vertical profile

All analyzed particle classes, showed a higher concentration at 3.8 m, compared to 1.5 m. This result can be explained by the aerosolization process, where particles are elevated by wind forces. Wind speed and particulate matter concentration profile have a logarithmic relationship. It is assumed, that ground-level PM emissions are mixed up at heights above 2m due to air turbulences (Öttl and Funk, 2007). The particle concentrations of PM_{10}, $PM_{2.5}$ and PM_1 at the higher measuring position were 9.2-, 1.9- and 1.25-fold increased compared to the lower one.

3.6.2. Horizontal profile

For all PM size classes except the PM_{10} particles measured at 3.8 m, the particle concentration increased with the distance from the source. There are three phases of airborne particle behavior (Fig. 3B). In phase I, fine and coarse particles are aerosolized the same way. Due to gravitation and connected sedimentation, small and light particles are retained longer in an air suspension (phase II) than large and heavy particles, which deposit faster (phase III). The sedimentation rate is approx. 2 mm s^{-1} for organic PM_{10} and even slower for smaller PM particles (Zanke, 1982). Except PM_{10} at 3.8 m, all analyzed PM particles were found in higher concentrations with increasing horizontal distance from the application site. Therefore, fine dust particles are nearly unaffected by deposition, and turbulent wind dispersion is the dominant effect acting on PM. The observations of this study confirmed that the airborne state of PM particles can be maintained over longer distances and can represent a health risk not exclusively close to the PM generation site. Particles aerosolized during this field experiment had the potential to be transported >1000 km (Thiel et al., 2020).

Akbar-Khanzadeh et al. (2012) studied horizontal and vertical PM profiles during biosolids field application and further agricultural activities. A total of 94% of the airborne PM was respirable. The particle concentration decreased with increasing height from the ground and was significantly highest at 0.5 m. For the horizontal profile, PM emissions slightly decreased with increasing distance from the application site. In particular, coarse particle concentrations declined rapidly due to faster deposition compared to that of finer particles.

3.7. Comparison of manure PM emissions under lab and field conditions

A comparison of PM concentrations measured under practical conditions on the field (Fig. 2) and under controlled conditions in a wind tunnel (Kabelitz et al., 2020) shows that only small amounts of PM were emitted when broiler manure had a DM <60%. Under controlled and field conditions, broiler manure with a DM >60% DM began to aerosolize high PM concentrations in a linear manner (Fig. 4). The higher the manure DM was, the more PM was measured. Thus, there is a strong positive correlation between the manure DM and PM release. However, the slope and amount of PM concentration measured were higher in the wind tunnel than on the field. A possible explanation for this variation are the different wind forces acting on the PM particles in both settings. On the field, PM particles are distributed in all directions around the emission source through wind fluctuations and turbulences. In the wind tunnel, all particles are forced to flow in the same direction and under constant wind conditions. Therefore, PM concentrations at the same distance for the PM emission source are supposed to be lower in the field than in the wind tunnel.

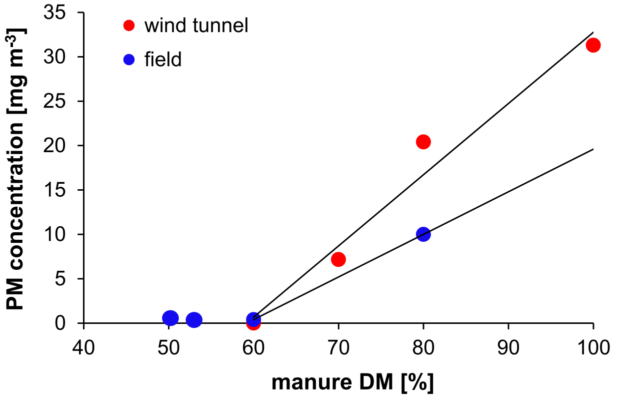


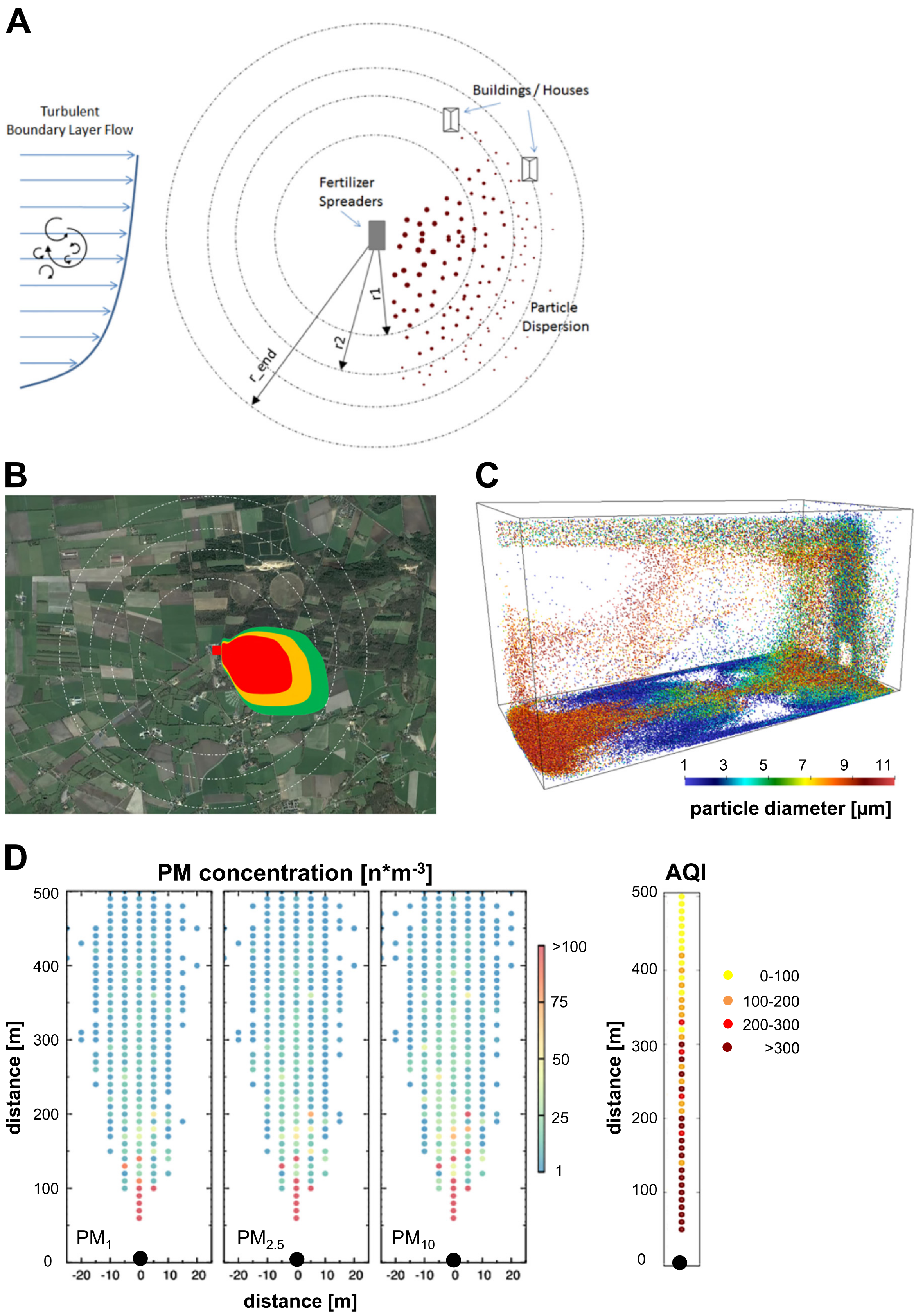
Fig. 4. Comparison of PM_{10} emission measurements from standardized and practical conditions. PM_{10} concentrations (in mg of PM per m^3 of air) measured under standardized (in a wind tunnel, red, data from Kabelitz et al., 2020) and practical (at the field, blue, data from Fig. 2) conditions from poultry manure with different dry matter contents (DM) are shown.

Untreated, composted and stored poultry manure had a DM of 49%, 52%, and 59% respectively (Table 1). According to our field experimental results, the PM_{10} concentrations of untreated, composted and stored broiler manure were comparable and relatively low (<1 mg m^{-3}). Note that in winter storing and composting do not lead to high moisture losses in manure. The situation is very different in summer, when manure DM increases rapidly because of elevated evaporation due to high temperatures. We attempted to represent manure under summer conditions with our dried treatment (approx. 80% DM). Here, much higher PM_{10} concentrations of approx. 10 mg m^{-3} were detected. Concluding that longer storage of poultry manure at cold and wet weather conditions is not problematic, but should be avoided in summer because of the potential elevated PM aerosolization during field application.

3.8. Modeling of PM emissions during manure application

To estimate PM pollution and the environmental risk in the surroundings of a manure application site, a computational fluid dynamic (CFD) model was developed. The model simulated aerial PM dispersion and was trained with measurements obtained in this study and from Kabelitz et al. (2020). A detailed description and analysis of the modeling will be published elsewhere. For the modeling, a turbulent boundary layer atmosphere and a stationary fertilizer spreader with a defined particle release were assumed (Fig. 5A). With the help of the model, risk zones for high, medium and low particle concentrations can be defined (Fig. 5B). Airborne dispersion of different particle size classes in a model room was calculated (Fig. 5C). Here, particles with smaller diameters (blue) dispersed farther than larger particles (red). Diverse input parameters (e.g., wind speed and direction, turbulence intensity of the inflow, particle DM, particle density and composition at the source) can be varied and simulated. The model was run with the input parameters measured during the application of dried poultry manure in the first field experiment and particle number concentrations of PM_1, $PM_{2.5}$ and PM_{10} per m^3 of air were simulated (Fig. 5D). The environmental and health risk estimates were based on the Air Quality Index (AQI), resulting in the following risk zones: I) up to 150 m from the source, >100 particles m^{-3} of air were calculated, resulting in an AQI >300 and thus a “hazardous” PM level; II) 150–300 m from the source, 25–75 particles m^{-3} were predicted, amounting to an AQI of 200–300 and an “very unhealthy” PM concentration; III) 300–400 m from the source, 1–25 particles m^{-3} were simulated, which corresponds to an AQI of 100–200 and an “unhealthy” PM level; and IV) >400 m from the PM emission site, <1 particle m^{-3} was calculated, resulting in an AQI <100 and a “moderate” to “good” air quality.

It can be concluded, that there might be an increased risk for humans who are directly exposed to the dust plume during fertilizer application, e.g., people living next to the field or agricultural workers. However, according to the simulations, there is probably a low risk to health and the environment beyond a distance of 400 m from the manure application site. Of course, the risk estimate and simulations essentially depend on the wind speed and direction, which are permanently fluctuating on the field. A recent study recommended a minimum distance of 160 m between manure application sites and crop fields or buildings, to avoid contamination with pathogens from manure (Jahne et al., 2016). Dungan (2012) detected an aerosolized *E. coli* marker strain 125 m downwind from a field fertilized with swine slurry, but not at 250 and 500 m.



4. Conclusions

In conclusion, organic fertilizer from livestock origin represents a currently underresearched source of PM aerosolization during and after application. In addition to the health effects of PM itself, gaseous emissions, odors and harmful microorganisms might be co-transported. Due to expanding cities, the need for increasing agricultural production, longer drought periods and the associated PM formation, the exposure to bioaerosols will further increase in the future. Thus, this study attempted to characterize PM production during poultry manure field application, as well as the influence of manure moisture content and treatment. As shown, solid manure application represents a relevant dust emission source with a high PM emission potential. Furthermore, manure PM acts as a vector for microbial distribution. Until now, research focused mainly on fertilized soil associated dissemination of AMR microbes, but disregarded PM as a potential carrier of the airborne spread of resistances. Our study identified the optimal DM of poultry manure to reduce PM aerosolization and microbial dispersion to be 50–70%, which corresponds to the natural DM of fresh poultry manure. Therefore, fresh poultry manure can be used for organic fertilization without further processing. However, longer storing in summer or drying without pelleting should be avoided. It was shown that the manure treatment strongly influence PM aerosolization rates. As an ideal treatment to minimize PM aerosolization and microbial risk, we recommend manure composting, where the DM should not exceed 70%. During composting, aerobic manure fermentation with temperatures around 60 °C induces inactivation of microorganisms and hence a lower health threat. From the results of our modeling and previous literature, we advise a distance of at least 150 m and optimally >400 m from the PM emission source to minimize the risk for public health and the environment. For the development of holistic mitigation measures of livestock associated PM emissions, more research is needed, especially regarding the aerosol parametrization and improved transmission studies. It remains unclear how the tenacity of pathogens is influenced by PM interactions, which particle sizes are typically associated with pathogenic microbes, and what the influences of physical particle composition and atmospheric environment are.

CRediT authorship contribution statement

Tina Kabelitz: Conceptualization, Data curation, Investigation, Methodology, Project administration, Resources, Supervision, Validation, Visualization, Writing – original draft, Writing – review & editing. **Oliver Biniasch:** Conceptualization, Data curation, Investigation, Methodology, Resources. **Christian Ammon:** Data curation, Formal analysis, Investigation, Resources, Software, Validation, Visualization, Writing – original draft. **Ulrich Nübel:** Conceptualization, Funding acquisition, Supervision, Writing – original draft, Writing – review & editing. **Nadine Thiel:** Investigation, Data curation, Methodology, Resources. **David Janke:** Software, Validation, Supervision, Visualization, Writing – original draft. **Senthilathiban Swaminathan:** Investigation, Software, Validation, Visualization. **Roger Funk:** Conceptualization, Funding acquisition, Supervision, Writing – original draft. **Steffen Münch:** Investigation, Methodology, Resources, Writing – original draft. **Uwe Rösler:** Conceptualization, Funding acquisition, Supervision. **Paul Siller:** Investigation, Methodology, Resources, Writing – original draft, Writing – review & editing. **Barbara Amon:** Conceptualization, Writing – original draft. **André J.A. Aarnink:** Conceptualization, Writing – original draft. **Thomas Amon:** Conceptualization, Funding acquisition, Project administration, Resources, Supervision, Writing – original draft.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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Fig. 5. Modeling of PM emissions during poultry manure application. (A) Graphical visualization of modeling assumptions: a stationary fertilizer spreader located near buildings is generating PM emissions. Coarse PM particles are dispersed over shorter distances (r1), whereas finer PM particles are spread over longer (r2) and maximum ranges (r_end). All particles are permanently subjected to a turbulent boundary layer flow. (B) The goal of the modeling was to predict minimum (green), medium (yellow) and high (red) risk zones for PM abundance under definable weather conditions. (C) Numerical simulation and particle dispersion model for PM particles with different diameters (fine = blue, coarse = red). (D) Particle concentrations and risk estimate levels related to the distance of the particle emission source (black dot). Particle concentrations for PM_1, $PM_{2.5}$ and PM_{10} (in particle number per m^{-3} of air) in the range of 1 (blue) to >100 (red) are colour scaled. Risk estimate (on the right) is based on PM_{10} concentrations using the Air Quality Index (AQI) according the four categories: 0–100 (yellow), 100–200 (orange), 200–300 (red) and > 300 (dark red).

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