

# PM10 concentrations and emissions of six naturally ventilated dairy housings with cubicles and an outdoor exercise area

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To improve the database for emission inventories, PM10 emissions for the most common dairy housing situation in Switzerland were determined. Measurements were taken on six naturally ventilated dairy loose housings with cubicles, solid floors and an outdoor exercise area in two out of three seasons (summer, transitional season, winter) per farm. PM10 was collected cumulatively over 72 h using impactors at 9 to 14 measuring points in the housing area, outdoor exercise area, and in the background. Emissions were determined by a tracer ratio method with two tracer gases ( $\text{SF}_6$ ,  $\text{SF}_5\text{CF}_3$ ). PM10 concentrations in the animal area were usually just above or within the range of the background concentration. The PM10 emissions varied between 0.02 and 2.1 g LU<sup>-1</sup> d<sup>-1</sup> across all farms. With the present amount of data, there was no recognisable relationship with the considered influencing variables. At 0.64 g cow<sup>-1</sup> d<sup>-1</sup> the derived PM10 emission factor is considerably lower than the emission factors used in inventories to date.

## Keywords

PM10 emission, PM10 concentrations, Natural ventilation, Dairy loose housing, Outdoor exercise area

The total PM10 emissions for Switzerland in 2010 are estimated at around 20'000 t (FOEN 2013). According to the FOEN calculations, 27 % of these PM10 emissions are attributed to the agriculture and forestry sector. Around 34 % of these are derived from animal husbandry. Dairy farming plays a major role here, since in 2012 it accounted for around 45 % – the largest contribution – of all Switzerland's livestock units (LU) (SCHWEIZERISCHER BAUERNVERBAND 2012).

## State of knowledge

There is a lack of data for PM10 emissions from naturally ventilated dairy loose housings. Up to now, particulate matter from livestock farming has primarily been investigated in view of animal health and occupational safety (HINZ 2002). In many cases, only the concentrations of dust fractions or bioaerosols in specific individual situations such as feed distribution or strawing were measured when considering adverse effects on human and animal health (HANHELA et al. 1995, LOUHELAINEN et al. 1987, LOUHELAINEN et al. 1997). PURDY et al. (2009) investigated dust emissions from different areas (e.g. milking parlour, loafing pen, commodity barn, compost field) of four dairy facilities in the USA. However, Results of American cattle feed lots measurements (e.g. GONZALES et al. 2011, HUANG et al. 2013, RAZOTE et al. 2004, SWEETEN et al. 1988, SWEETEN et al. 1998) are not transferable to dairy housings or outdoor exercise areas under Swiss housing conditions. PM10 emission values from TAKAI et al. (1998) range from 0.12 to 4.05 g LU<sup>-1</sup> d<sup>-1</sup>. These values were derived from PM2.5 and

PM100 measurements in dairy loose housings in the Netherlands, Great Britain, Germany and Denmark. These data form the basis for PM10 emission factors in cattle housing in several inventories (e.g. DÖHLER et al. 2002, EUROPEAN ENVIRONMENT AGENCY 2013, HAENEL et al. 2014).

SEEDORF (2004) also measured inhalable and respirable dust fractions as well as bioaerosols (inhalable endotoxins, respirable endotoxins, mesophilic bacteria, Enterobacteriaceae, fungi) in eight dairy housings in Northern Germany. But PM10 emissions were not derived from these measurements because of uncertainties in the conversion factors.

An overview of PM10 concentration and emission measurements in dairy housings from literature is given in Table 1. Differences in the PM10 concentration determination method, the calculation of emission, measurement concept, housing system, season, measurement duration and reference variables make it difficult to compare these published investigations. KAASIK and MAASIKMETS (2013) measured concentrations for the particle size fractions TSP, PM10, PM2.5 and PM1.0 in nine cubicle housings for dairy cattle in Estonia. However, these are only short-time measurements lasting 1.2 to 2.5 hours. SCHMIDT et al. (2002), by contrast, carried out 24-hour measurements per season over ten days in only one dairy farm in the United States.

PM10 concentrations vary from  $4 \mu\text{g m}^{-3}$  (HENSELER-PASSMANN 2010) to  $370 \mu\text{g m}^{-3}$  (SCHMIDT et al. 2002). In the German investigation on three dairy farms, PM10 concentrations in a deep litter loose housing were higher than in two cubicle loose housings (HENSELER-PASSMANN 2010). In a deep litter loose dairy housing in a Czech study, PM10 concentrations measured in summer were clearly lower with daily averages ranging from  $42$  to  $132 \mu\text{g m}^{-3}$  (DOLEJŠ et al. 2006) than in the deep litter housing in Germany with  $198 \mu\text{g m}^{-3}$  found by HENSELER-PASSMANN (2010). A temperature effect on PM10 concentrations was shown in Joo et al. (2013). In this study, PM10 concentrations increased with a rise in temperature (Joo et al. 2013). Dust measurements of feedlots showed an effect of surface water content and watering on PM10. PM10 concentration decreased with increasing pen surface water content (GONZALES et al. 2011). Rainfall events or sprinkling caused a significant reduction of PM10 concentration (BONIFACIO et al. 2011).

PM10 concentration and emission values of dairy housings from the literature show a wide distribution; there is a large range both within and between the studies. PM10 emissions from a cubicle loose housing system for dairy cattle in the Netherlands ranged from  $0.08$  to  $0.41 \text{ g animal place}^{-1} \text{ d}^{-1}$  (MOSQUERA et al. 2012). Higher emission values resulted for cubicle loose housing systems at  $0.10$  to  $1.44 \text{ g animal place}^{-1} \text{ d}^{-1}$  in Germany (HEIDENREICH et al. 2008) and in the USA of  $0.38$  to  $1.99 \text{ g LU}^{-1} \text{ d}^{-1}$  (SCHMIDT et al. 2002). Further measurements taken in two naturally ventilated dairy housings with and without an open floor pen in the USA revealed markedly higher PM10 emissions of  $11.9$  to  $15.0 \text{ g cow}^{-1} \text{ d}^{-1}$  (Joo et al. 2013).

Table 1: Literature overview: Concentration and emission of PM10 from dairy housing (Su = summer; Tr = transition period; Wi = winter; LU = livestock unit, 1 LU = 500 kg live weight)

Housing system	PM10 concentration in $\mu\text{g m}^{-3}$	PM10 emission	Methods	Duration of measurements	Region	Reference
Deep litter loose housing	Su: 42-132	Not depicted	DustTrak aerosol monitors	3 days	Czech Republic	DOLEJŠ et al. (2006)
Cubicle loose housing with straw mattress	Su: 14.4; 49.9 Tr: 35.5; 63.6; 107.3 Wi: 20.5; 42.2	Su: 0.96 g animal place <sup>-1</sup> d <sup>-1</sup> Tr: 1.44 g animal place <sup>-1</sup> d <sup>-1</sup> Wi: 0.24 g animal place <sup>-1</sup> d <sup>-1</sup>	Aerosol spectrometer, vane anemometer, sonic anemometer, tracer gas decay (Kr <sup>85</sup> )	2 days each season and housing	Germany	HEIDENREICH et al. (2008)
Cubicle loose housing with rubber mat	Wi: 13.7; 25.5	Wi: 0.10 g animal place <sup>-1</sup> d <sup>-1</sup>				
Deep litter loose housing	Su: 198 Tr: 170 Wi: 206	Su: 2.06 g LU <sup>-1</sup> d <sup>-1</sup> ; Tr: 2.35 g LU <sup>-1</sup> d <sup>-1</sup> Wi: 2.78 g LU <sup>-1</sup> d <sup>-1</sup>	Aerosol spectrometer, tracer gas decay (SF <sub>6</sub> )	Several days each season and housing	Germany	HENSELER-PASSMANN (2010)
Cubicle loose housing	Su: 15 Tr: 13 Wi: 4	Not depicted				
Cubicle loose housing	Tr: 11 Wi: 8	Not depicted				
Tie stall, forced ventilated	Not depicted	0.19 g cow <sup>-1</sup> d <sup>-1</sup>	Isokinetic sampling, gravimetric methods anemometer	Several days in July and November	Poland	HINZ et al. (2007)
Cubicle loose housings with and without an open-floor pen (2 housings)	Su and Tr: 64-240 Wi: 22-29	11.9-15.0 g cow <sup>-1</sup> d <sup>-1</sup>	TEOM (Tapered element oscillating microbalance), 3D sonic anemometers	More than a year	USA	JOO et al. (2013)
Cubicle loose housings (9 farms)	27-123	Not determined	Aerosol spectrometer	Short-time: 1.2-2.5 h	Estonia	KAASIK and MAASIKMETS (2013)
Cubicle loose housing, year-round indoor housing	Su: 31; 41 Tr: 11; 23 Wi: 25; 29	Su: 0.27; 0.29 g animal place <sup>-1</sup> d <sup>-1</sup> Tr: 0.31; 0.41 g animal place <sup>-1</sup> d <sup>-1</sup> Wi: 0.08; 0.21 g animal place <sup>-1</sup> d <sup>-1</sup>	Gravimetric cyclone separator, carbon dioxide balance	6 single days over the year	The Netherlands	MOSQUERA et al. (2012)
Cubicle loose housing with mattresses and sawdust bedding	Su: 370 Wi: 60	Su: 0.12; 0.38 g LU <sup>-1</sup> d <sup>-1</sup> Wi: 0.70; 1.99 g LU <sup>-1</sup> d <sup>-1</sup>	Portable air sampler, carbon dioxide balance	10 days each season	USA	SCHMIDT et al. (2002)

Emission data for PM10 have heretofore not been available for dairy housing systems with cubicles and an outdoor exercise area common in Switzerland, which means, inter alia, larger activity areas. The aims of this study were to determine the PM10 emissions and to derive a PM10 emission factor for the naturally ventilated loose housing system with cubicles and an outdoor exercise area common in Swiss dairy farming, thereby making a contribution to emissions inventories.

## Materials and Methods

### Farms

We selected the most common dairy loose-housing system with cubicles and an outdoor exercise area in Switzerland. It consists of naturally ventilated single-building loose-housings with cubicles, no thermal insulation, solid floor surfaces and an outdoor exercise area arranged lengthways to the housing (SCHRADE et al. 2011). We investigated two outdoor exercise area concepts, each on three farms: i) outdoor exercise area (OEA) spatially separated from the housing, ii) combined cubicle access aisle/outdoor exercise area (CAA/OEA) on three farms (Figure 1). Cubicles were designed as deep-bedded cubicles with long straw, chopped straw or a combination of straw and sawdust (Table 2). The feeding aisle, cubicle access area or combined cubicle access area/outdoor exercise area were mucked out three to four times daily with stationary scrapers, whilst the outdoor exercise area was mucked out with portable equipment every three days or so.

Herd sizes ranged between 20 and 74 animals. Besides dairy cows and young calves, on farm 3 female offspring and on farms 2 and 4 additionally breeding bulls were kept in the housing. The average milk yield during the measuring periods varied from 19 to 31 kg cow<sup>-1</sup> d<sup>-1</sup>. On farms 1, 2 and 3, the feed ration basically consisted of silage, hay and concentrate. Farm 4 fed no silage, and farms 5 and 6 provided a total mixed ration. The animals were not grazed during measurements, and a preceding three-day acclimatisation period was set before starting the measurements. The farms are described in more detail in SCHRADE et al. (2012) and SCHRADE (2009).

### Measuring concept, analysis, tracer ratio method, and emission calculation

It is not possible to transfer emission data from measurements on one single farm to an entire housing system (SEIPELT 1999). Only measurements from a housing system on several farms can give reliable values (AARNINK and OGINK 2006; Groot Koerkamp et al. 1998). Several measurements spread throughout the year are also essential to take account of the climatic variations in housing affected by external weather conditions (AARNINK and OGINK 2006; GROOT KOERKAMP et al. 1998). Therefore, we carried out measurements on six farms in two out of three seasons (summer, transition period, winter) from August 2007 to August 2008. Each combination of seasons occurred on two farms (SCHRADE et al. 2011; SCHRADE et al. 2012).

Particulates of the particle-size fraction PM<sub>10</sub> was sampled cumulatively gravimetric with impactors (PEM-200-4-10, MSP Corp., USA). These impactors were developed for workplace hygiene applications and were additionally validated for low air speeds (LAI and CHEN 2000). They were used to determine PM<sub>10</sub> concentrations in previous studies in pig housings (BERRY et al. 2005). Controlled diaphragm pumps (GilAir 5, Sensidyne, USA) enabled an even volumetric flow rate of 4 l min<sup>-1</sup>. The pumps were located at a maximum distance of 2 m from the impactor in a box protecting them from damp and dust. The impactors themselves were protected from coarse dust, insects, rain or high air speeds by aluminium caps. These had circular openings that allowed sufficient airflow independently of the wind direction, in analogy to DIN EN 12 341 (2014). The aerosol-containing sample is strongly accelerated with a defined volumetric flow rate in a nozzle and then deflected. Due to their inertia, particles larger than 10 µm bounce a lubricated baffle plate and is deposited there. Smaller particles (≤ PM<sub>10</sub>) remain in the gas stream and retain on the subsequent filter (S&S, GF 10 HY, Ø 37 mm; Whatman membrane filter, PTFE supported, 5.0 µm, Ø 37 mm). The PM<sub>10</sub> mass on the filter was gravimetrically determined in the laboratory. Both before and after measurement, the filters were

conditioned for 24 hours at 22 °C and 50% relative humidity. Based on previous studies we assumed a detection limit of  $10 \mu\text{g m}^{-3}$  (BERRY et al. 2005). To accumulate sufficient dust, the filters were exposed for 72 h. This exposure time was derived from preliminary experiments on naturally ventilated dairy housing with an outdoor exercise area, where varied exposure durations were tested systematically.

To obtain a representative sampling inside the housings and at the outdoor exercise areas, three to five impactors evenly distributed along each measuring axis at a height of approx. 3 m were operated simultaneously. This yielded a total of 9 to 14 measuring points (Figure 1), depending on the housing dimensions. To determine the background concentration, two impactors were exposed at a location not influenced by the housing.

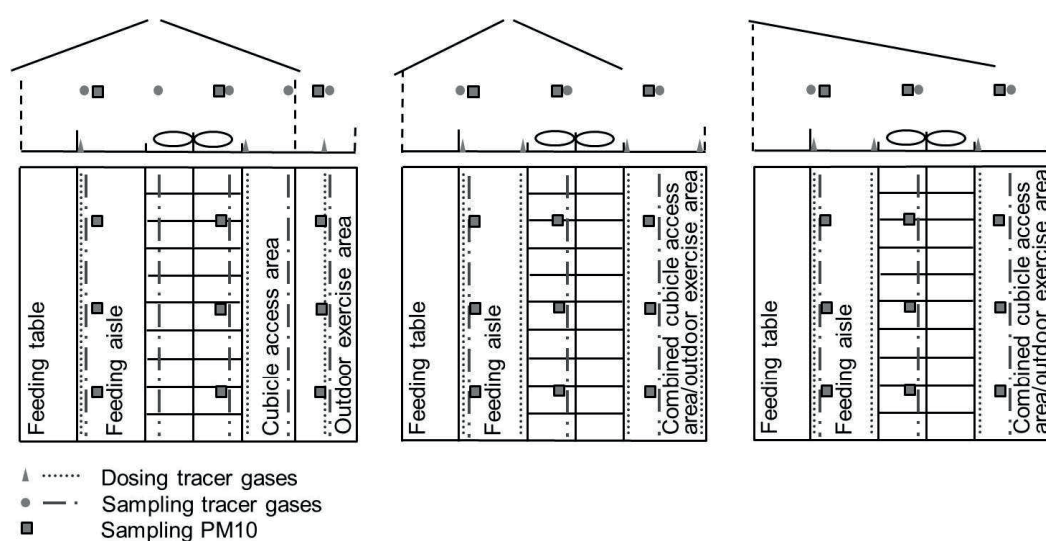


Figure 1: Schematic diagrams showing layout and section of both commercial farm housing concepts with dosing and sampling axes: outdoor exercise area separate from housing (left); combined cubicle access aisle/outdoor exercise area (middle and right) (dashed lines in the section drawing stand for open or semi-open façades)

A tracer-ratio method with two tracer gases was developed in order to determine the emissions for natural ventilation and from diffuse sources. In addition to the already-established tracer gas sulphur hexafluoride,  $\text{SF}_6$ , for emission measurements in naturally ventilated livestock housings (e.g. BERRY et al. 2005; HENSELER-PASSMANN 2010; MÜLLER et al. 2006; NANNEN et al. 2006; NIEBAUM 2001; SCHIEFLER 2013; SEIPELT 1999), trifluoromethyl sulphur pentafluoride,  $\text{SF}_5\text{CF}_3$  (Ho et al. 2008; STURGES et al. 2000), was used as a second tracer. The diluted tracer gases were continuously supplied next to the floor surfaces (Figure 1) via a metal tube system ( $\text{Ø}i$  4.53 mm; Interally, Switzerland) with steel critical capillaries ( $\text{Ø}i$  30  $\mu\text{m}$ ; Lenox Laser, Glen Arn, USA). Within each longitudinal axis the distances between the dosing critical capillaries were 3 or 6 m. The tracer gases were diluted with compressed air. At the dosed concentrations (600–800 ppm each of  $\text{SF}_6$  and  $\text{SF}_5\text{CF}_3$ ) the density of the tracer gases differs by less than 1% from ambient air and hence does not hinder mixing with the air in the housing.

An air-collection system consisting of Teflon tubes ( $\text{Ø}i$  6 mm) and glass critical capillaries ( $\text{Ø}i$  250  $\mu\text{m}$ ; Thermo-Instruments, Germany and Louwers, The Netherlands) every 3 m enabled us to take a representative sampling of the tracer gases in the spacious housings. A similar system was used by NIEBAUM (2001) and SEIPELT (1999). The sampling lines on farms 2, 3, 4, 5, and 6 were fitted

in longitudinal axes at a height of 3 m (Figure 1) next to the PM10 impactors. On farm 1, the air-collecting system was positioned in the housing openings (gate, roof ridge, windows) as well as on the central axis of the outdoor exercise area at a height of 3 m. The two tracer gases were analysed simultaneously by means of gas chromatography with electron-capture detector (GC-ECD, 3400Cx Series, Varian, USA). More detailed information on the tracer-ratio method is given in SCHRADE et al. (2012) and ZEYER et al. (2012).

The basis for calculating PM10 emissions were the PM10 concentrations determined over a 3-day measurement timeframe. The median was formed from the PM10 concentrations of the single measuring locations in the animal area (housing area and outdoor exercise area or cubicle access aisle/outdoor exercise area). Then the PM10 emissions were calculated based on the 72 h averages tracer gas concentrations and mass flows. The calculation of the emission by the tracer ratio method is based on the assumption that the tracer gas ( $T$ ) behaves in the same way as the emitted particle-size fraction PM10 ( $PM10$ ) and thus mimics the emitting source. The ratio of the concentration ( $c$ ) of both substances then corresponds to the ratio of their mass flow ( $\dot{m}$ ).

$$\frac{\dot{m}_{PM10}}{\dot{m}_T} = \frac{c_{PM10}}{c_T} \quad (\text{Eq. 1})$$

and thus

$$\dot{m}_{PM10} = \frac{\dot{m}_T \cdot c_{PM10}}{c_T} \quad (\text{Eq. 2})$$

This tracer-ratio-method is described in detail in SCHRADE (2009) and SCHRADE et al. (2012).

### Accompanying parameters

In addition to the descriptive farm data (q.v. table 2), the soiling of floor surfaces, and the use of different areas by the animals and a variety of climate parameters served to describe the measuring situation in each case, to determine the plausibility of measuring data, to act as reference values, and to aid in determining variables with a significant influence on emissions:

- Outdoor climate ( $1 \text{ min}^{-1}$ ): A weather station recorded air temperature (NTC, Testo, Germany), relative atmospheric humidity (capacitive thin-film sensor, Testo, Germany) wind speed, and wind direction (2-axis ultrasonic anemometer WindObserverTM, GILL, United Kingdom) at a height of approx. 2.5 m at a distance of 100–200 m from the housing. Sensors for pressure (absolute pressure sensor, thick film ceramic), global radiation (pyranometer) and precipitation (self-emptying rocker with pulse sensor) were positioned approx. 15 m away from the housing at a height of approx. 2 m.
- Climate in the housing and in the outdoor exercise area or combined cubicle access aisle/outdoor exercise area ( $1 \text{ min}^{-1}$ ): Air temperature (NTC, Testo, Germany), relative atmospheric humidity (capacitive thin-film sensor, Testo, Germany) and wind speed (Hot-wire anemometer, Schmidt Technology, Germany) were recorded in the individual areas (feeding aisle, cubicle access aisle, combined cubicle access aisle/outdoor exercise area and outdoor exercise area) both close to the floor (50 cm above the ground) and at a sampling height of 3 m above ground. Additionally wind speed and wind direction (3-axis ultrasonic anemometer WindMasterTM, GILL, United Kingdom)



were measured either between housing and outdoor exercise area (separated outdoor exercise area) or inside the housing (combined cubicle access aisle/outdoor exercise area).

In SCHRADE et al. (2012) accompanying parameters are described in more detail.

Table 2: Description of farms and measuring periods: herd, feed, arrangement of outdoor exercise area, façade design, bedding, dosing and sampling, climate (FA = feeding aisle; CAA = cubicle access area; CAA/OEA = cubicle access aisle/outdoor exercise area; OEA = outdoor exercise area; Su = summer; Tr = transition period; Wi = winter; LU = livestock unit, 1 LU = 500 kg live weight; DM = dry matter; TMR = total mixed ration)

Parameter	Farm 1		Farm 2		Farm 3		Farm 4		Farm 5		Farm 6	
	Su	Tr	Su	Wi	Tr	Wi	Tr	Wi	Su	Wi	Su	Tr
Herd	Dairy cows		Dairy cows, breeding bulls		Dairy cows, female offspring		Dairy cows, breeding bull		Dairy cows		Dairy cows	
Number of animals	20	20	40	40	74	71	27/28	28	47	46	50/53	50
Number of LU	28	28	58	70	94	97	39/40	41	77	78	85/90	83
Feed components	Grass silage, hay, concentrate		Grass silage, maize silage, hay, concentrate, Su: green forage		Grass silage, maize silage, hay, concentrate		Hay, concentrate		TMR: Grass silage, rapeseed cake maize grain silage, extracted soybean meal; Su: urea Wi: potato, maize silage, alfalfa silage, alfalfa hay, sugar beet pulp silage		TMR: Grass silage, maize grain silage, alfalfa hay, extracted soybean meal, extracted rapeseed meal, maize gluten, corn cob mix, concentrate	
Bedding (deep-bedded cubicles)	Straw and sawdust		Long straw		Long straw		Chopped straw		Long straw		Long straw	
Arrangement of housing and OEA	OEA separate		OEA separate		CAA = OEA		OEA separate		CAA = OEA		CAA = OEA	
Design of façade towards outdoor exercise area	Wall and open windows		Timber wall, open at top		open		Wall and curtains (open)		open		open	
Total area in m <sup>2</sup>	289		575		858		412		529		568	
of which traffic area	215		440		624		295		377		388	
of which OEA; CAA/OEA	82		197		360		99		168		180	
Tracer gas dosing, number of axes and gas												
FA	1 x SF <sub>6</sub>		1 x SF <sub>6</sub>		1 x SF <sub>5</sub> CF <sub>3</sub>		2 x SF <sub>6</sub>		2 x SF <sub>6</sub>		-	
CAA	1 x SF <sub>6</sub>		1 x SF <sub>6</sub>		-		2 x SF <sub>6</sub>		-		2 x SF <sub>6</sub>	
CAA/OEA	-		-		2 x SF <sub>6</sub>		-		2 x SF <sub>5</sub> CF <sub>3</sub>		-	
separate OEA	1 x SF <sub>5</sub> CF <sub>3</sub>		1 x SF <sub>5</sub> CF <sub>3</sub>		-		1 x SF <sub>5</sub> CF <sub>3</sub>		-		2 x SF <sub>5</sub> CF <sub>3</sub>	
Tracer gas sampling location (number of axes)	Gate, roof ridge, windows, OEA (3)		Feeding barrier, 2 x cubicles, OEA (4)		FA, cubicles, CAA/OEA (3)		FA, CAA, OEA (3)		FA, cubicles, CAA/OEA (3)		FA, cubicles, CAA/OEA (3)	
PM10 sampling, number of impactors												
housing	9		10		8		6		8		8	
CAA/OEA, separate OEA	5		3		5		3		4		4	
background	2		2		2		2		2		2	
Climate: 3-day measurement timeframe 1 and 2												
Mean air temperature (background) in °C	14	15	15	5	8	4	8	0	19	1	18	13
	17	13	18				10	1				
Mean wind speed (housing) in m s <sup>-1</sup>	0.2	0.2	0.5	0.5	0.5	0.4	0.3	0.2	0.5	0.3	0.3	0.4
	0.2	0.2	0.4				0.4	0.3				
Mean relative atmos. humidity (housing) in %	88	85	83	90	60	87	91	80	72	71	57	89
	85	94	77				71	79				
Rainfall accumulated per 3 days in l m <sup>-2</sup>	0.5	6.4	0.1	2.0	2.3	3.0	62.7	8.9	9.3	0	3.3	2.8
	3.0	2.0	0				0.4	0				

### Statistical analysis

Statistical analysis was carried out using the S-Plus ® Version 7.0 for Windows statistics program on the three-day measurement timeframe level. Average values over the relevant measurement timeframe were determined for the climate data of high temporal resolution (e.g. air temperature, wind speed, relative atmospheric humidity, air pressure, global radiation, precipitation).

Variance analysis was then used to find out how the PM10 concentrations of the individual measurement points in the animal area (housing and outdoor exercise area or cubicle access aisle/outdoor exercise area) differed from the background values. This took into account a hierarchically nested effect of multiple measurements  $b_{ijkl}$  in the measurement timeframe  $b_{ijk}$  in the measuring period  $b_j$  on farm  $b_i$ . The area  $\beta_1 A$  (animal area versus background) was entered as a fixed effect. The target variable PM10 concentration  $c_{ijklm}$  ( $\mu\text{g m}^{-3}$ ) was converted to a logarithmic value. The variance inhomogeneity was also corrected:

$$c_{ijklm} = \mu + b_i + b_j + b_{ijk} + b_{ijkl} + \beta_1 B + \varepsilon_{ijklm} \quad (\text{Eq. 3})$$

The influence of season  $\beta_1 JZ$ , outside temperature  $\beta_2 AT$  ( $^{\circ}\text{C}$ ) and relative atmospheric humidity  $\beta_3 RF$  (%) on PM10 emission  $E_{ij}$  ( $\text{g LU}^{-1} \text{d}^{-1}$ ) was checked using a linear mixed-effects model which took the farm into account as a random effect:

$$E_{ij} = \mu + b_i + \beta_1 JZ + \beta_2 AT + \beta_3 RF + \varepsilon_{ij} \quad (\text{Eq. 4})$$

The target variable  $E_{ij}$  was logarithmically transformed. A graphical residuals analysis was used to check the model assumptions. The significance level was set at 5%.

### PM10 emission factor calculation and derived Swiss emissions for dairy farming

The emission factor was calculated as the arithmetic mean of PM10 emissions of all 3-day measuring periods (reference variable: livestock unit LU). Only the transition measurement period at farm 6 was not included because of a power failure which limited the measurement to 24 h.

Data from the “Swiss Farmers’ Union Statistical Surveys and Estimates of Agriculture and Nutrition” (SCHWEIZERISCHER BAUERNVERBAND 2012) were used to derive the PM10 emissions from Swiss dairy farming and illustrate the trend between 1999 and 2012. It is currently not possible to differentiate between the various dairy housing systems as there are no PM10 emission factors for tied housing in the international emission inventory. Because of this, to calculate PM10 emissions for the total Swiss dairy cattle population, we used the PM10 emission factor based on our measurements.



## Results

### Climate

On a minute level, the temperature in all measuring periods ranged from  $-8$  to  $37$  °C (SCHRADE et al. 2012). The background air temperature varied between  $14$  and  $19$  °C in summer, between  $8$  and  $15$  °C in the transition period, and between  $1$  and  $5$  °C in winter (Table 2). There were only slight temperature differences between the individual measuring points of background, outdoor exercise area or cubicle access aisle/outdoor exercise area and housing area: in winter the average air temperature in the housing area was in part up to  $2$  K above the background, in summer these were virtually identical (SCHRADE et al. 2012). The mean wind speed (mean minute values) was lowest inside the housing, followed by the outdoor exercise area or cubicle access aisle/outdoor exercise area and the background measurement site (SCHRADE et al. 2012). The mean relative atmospheric humidity in the housing per 3-day measurement timeframe ranged from  $57$  to  $94$ %. In ten of the 17 3-day measurement timeframes, the mean relative atmospheric humidity in the housing was above  $80$ %. In the case of relative atmospheric humidity the differences between the individual areas of background, outdoor exercise area or cubicle access aisle/outdoor exercise area and housing area were very slight (SCHRADE 2009). Three of the 3-day measurement timeframes were precipitation-free (summer farm 2; winter farms 4 and 5). The greatest amount of precipitation,  $63$  l m<sup>-2</sup> within three days, fell on farm 4 in the transition period.

### PM10 concentrations

The PM10 concentrations of the 17 3-day measurement timeframes over 12 measuring periods, separated according to animal area (housing area, outdoor exercise area or cubicle access aisle/outdoor exercise area) and background are shown in Figure 2. The PM10 concentrations over all measurements varied in the background between the detection limit ( $< 10 \mu\text{g m}^{-3}$ ) and  $40 \mu\text{g m}^{-3}$ , and ranged in the animal area (outdoor exercise area or cubicle access aisle/outdoor exercise area and housing area) from the detection limit to  $69 \mu\text{g m}^{-3}$ . Surprisingly, the measured background concentrations for the summer measurement of farm 5 were markedly higher than the values in the animal area. The reason for this could be the cereal harvest on fields in the surrounding area, or an increased amount of dust from a gravelled service road approx. 20 m apart from the background measuring location. Hence, the PM10 measurements of a suitable air pollution monitoring site (Winterthur Obertor) were used as a background value for this measuring period (OSTLUFT 2008). For farm 6, only one 24-hour measurement could be conducted in the transitional season because of a power outage caused by a storm. The wide variation of these values can be attributed to the short exposure duration of the filters, and to the resulting measurement uncertainty.

PM10 concentrations in the housing area were in the majority of the measuring periods slightly higher than in the outdoor exercise area or cubicle access aisle/outdoor exercise area (SCHRADE et al. 2014).

The variance analysis shows that the background concentration differed significantly from the animal-area concentration across all 3-day measurement timeframes ( $F_{1,17} = 14.62$ ;  $p = 0.001$ ), with the arithmetic means of background concentration at  $17 \mu\text{g m}^{-3}$  and animal area concentration at  $26 \mu\text{g m}^{-3}$ .

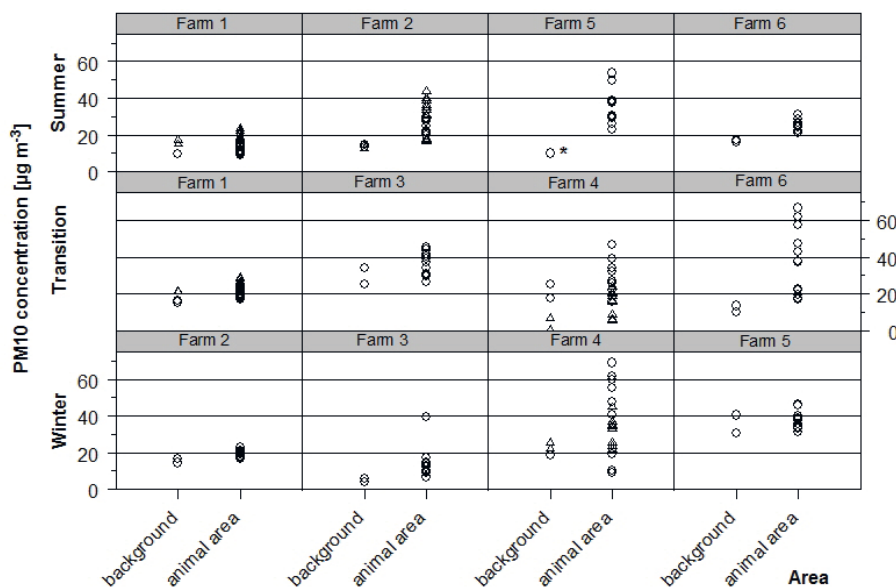


Figure 2: PM10 concentrations in  $\mu\text{g m}^{-3}$  in the animal area and background per farm and season, shown within the measuring periods as 3-day measurement timeframes (O = 3-day measurement 1;  $\Delta$  = 3-day measurement 2); \* measurements at Ostluft monitoring site Winterthur Obertor (OSTLUFT 2008)

### PM10 emissions

Over all farms and seasons, the PM10 emissions ranged between 0.02 and 2.1 g LU<sup>-1</sup> d<sup>-1</sup> or 0.03 and 2.8 g animal<sup>-1</sup> d<sup>-1</sup> (Figure 3). Within the farms, differences in emission levels between seasons, measuring periods and 3-day measurement timeframes were in some case cognoscible. Seasonal effects are not systematically identifiable, however. On farm 5, for example, at 1.27 g LU<sup>-1</sup> d<sup>-1</sup> the emissions in summer were significantly higher than the 0.07 g LU<sup>-1</sup> d<sup>-1</sup> in winter, whereas on farm 4 the emissions in one winter measurement, 1.42 g LU<sup>-1</sup> d<sup>-1</sup>, were significantly above those of the transition period (0.32 g LU<sup>-1</sup> d<sup>-1</sup>; 0.52 g LU<sup>-1</sup> d<sup>-1</sup>). On farm 1 and 3 the differences in PM10 emissions between the seasons are negligible. The high value in the transition period on farm 6 may be due to the wide variation of the concentrations measured in the shorter 24-h measurement because of a power outage caused by a storm.

This inconsistent picture is also reflected by the statistical analysis of the influencing variables. According to a linear mixed-effects model which takes account of the farm as a random effect, none of the investigated influencing variables (season, outside temperature, relative humidity in the housing) showed a significant influence on the PM10 emissions. This may be due to the fact that the PM10 concentrations in the animal area were often only slightly above or within the range of the background measurements, which leads to a high level of relative uncertainty in the resulting emissions.

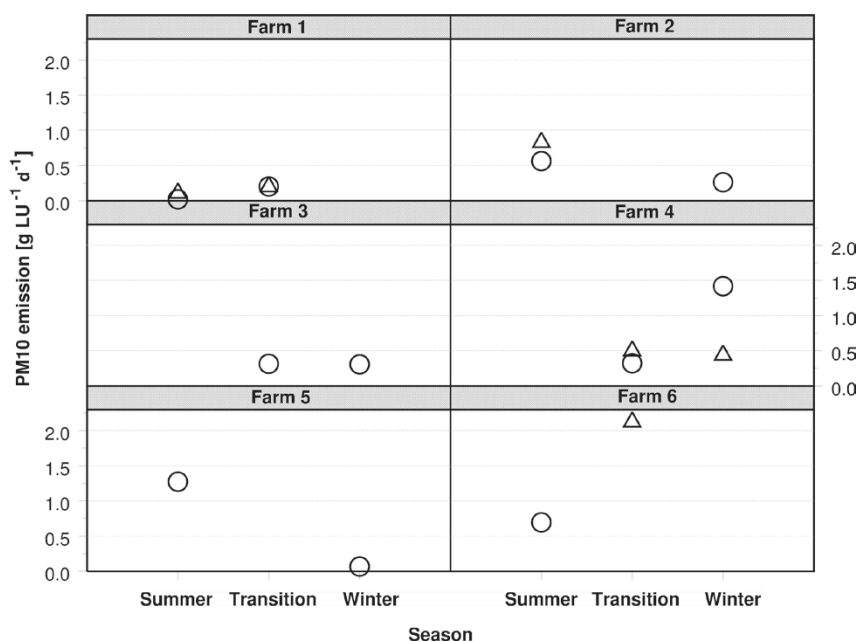


Figure 3: PM10 emissions in g LU<sup>-1</sup> d<sup>-1</sup> according to farm and season, given per 3-day measuring period, calculated on the basis of the median of the PM10 concentration from the animal area (housing area, outdoor exercise area or cubicle access aisle/outdoor exercise area) and the tracer gas measurements, (O = 3-day measurement 1; Δ = 3-day measurement 2)

### PM10 emission factor and extrapolation

Over all measuring periods the PM10 emissions varied between 0.07 and 1.27 g LU<sup>-1</sup> d<sup>-1</sup>. The arithmetic mean calculated from this, and hence the PM10 emission factor, is 0.48 g LU<sup>-1</sup> d<sup>-1</sup> (0.18 kg LU<sup>-1</sup> a<sup>-1</sup>) and 0.64 g animal<sup>-1</sup> d<sup>-1</sup> (0.23 kg animal<sup>-1</sup> a<sup>-1</sup>). The conversion was carried out using the KTBL live-stock unit calculator (KTBL 2014).

Between 1999 and 2004 the number of dairy cows in Switzerland fell from approx. 684'000 to around 620'000 animals, in subsequent years fluctuated around the 2004 level, increased slightly again in 2008 (around 630'000) and between 2010 and 2012 amounted to some 590'000 animals (SCHWEIZERISCHER BAUERNVERBAND 2000–2012).

Since we have no means to emission factors that differentiate between the housing systems, the PM10 emissions scale linearly with the dairy cattle population. The calculated PM10 emissions of Swiss dairy farming based on the emission factor derived from our own measurements decreased from 160 t a<sup>-1</sup> in 1999 to 138 t a<sup>-1</sup> in 2012 (Figure 4). The extrapolated PM10 emissions based on our own PM10 emission factor derived from measurements are significantly lower than those based on emission factors currently used in the inventories (CEPMEIP 2014, EUROPEAN ENVIRONMENT AGENCY 2013). Grazing and alpine pasturing could not be included in the Swiss PM10 emission calculation due to inadequate base.

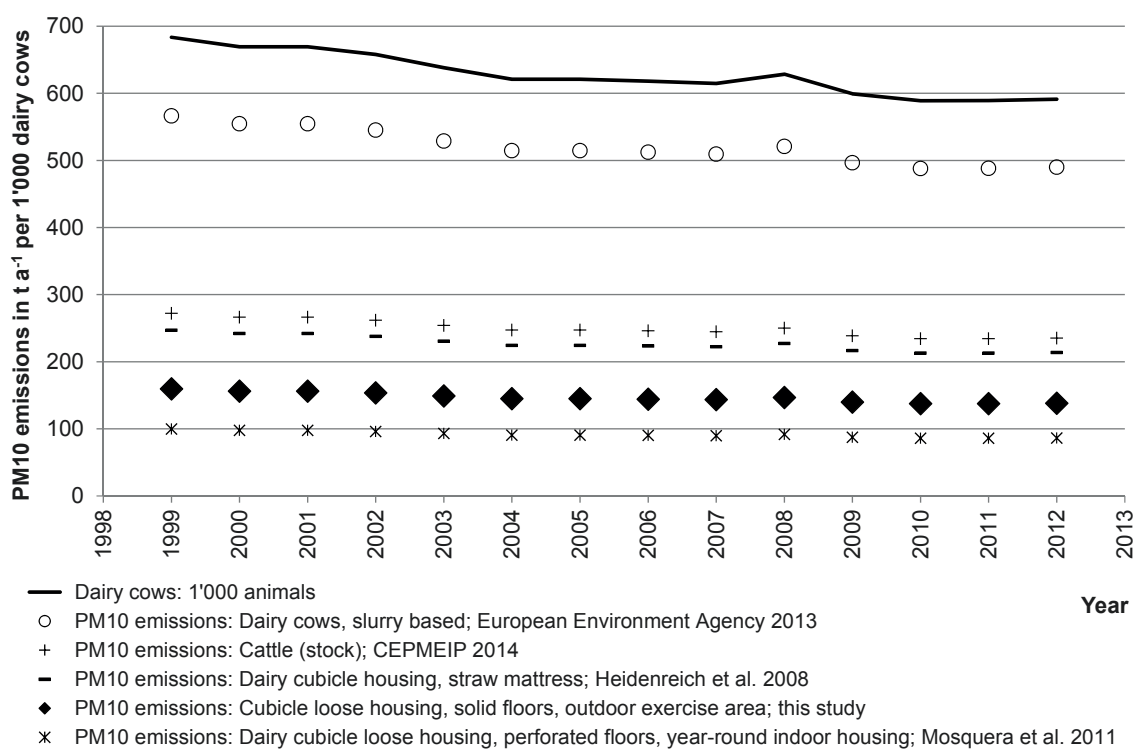


Figure 4: Dairy cattle population and PM10 emission in t a<sup>-1</sup> trends in Switzerland between 1999 and 2012. PM10 emissions for Swiss dairy farming are depicted calculation based on emission factor derived from our own measurements as well as using various emission factors (CEPMEIP 2014; EUROPEAN ENVIRONMENT AGENCY 2013; HEIDENREICH et al. 2008; MOSQUERA et al. 2011).

## Discussion

### Measuring concept and methods

With systematic emission measurements in cubicle loose housing with solid floors and an outdoor exercise area on six commercial farms in altogether twelve measuring periods it was possible in this study to model farm effects for a housing system and to ensure measurement conditions commonly found in practice. The six farms selected differed in respect of feed, management, farming method, herd performance, size, and structural details such solid floor and façade design. This study, therefore, covers a broad spectrum of the variety found in practice.

With measurements in two out of three seasons per farm it was possible to record two different climate situations within each of the farms and to cover the climate over the course of a year throughout all the farms. The temperature range in this study was very wide, with outside temperatures of between  $-8$  and  $37$  °C.

The tracer-ratio method developed for these studies with constant dosing, air collection samples and online analysis was successfully employed in loose housing with outdoor exercise areas.  $SF_5CF_3$  proved to be a suitable tracer gas in addition to the already established  $SF_6$  (ZEYER et al. 2012).

At the same time, the gravimetric determination of PM10 with impactors allowed spatially highly resolved sampling in the generously proportioned housing and outdoor exercise areas. As in measurements taken in pig housings, it proved effective to use controlled pumps and aluminium caps to protect the impactors (BERRY et al. 2005). The PM10 concentration of the investigated dairy farms was significantly lower by comparison with the measurements of BERRY et al. (2005) in pig housings, sometimes close to the  $10 \mu\text{g m}^{-3}$  detection limit. The difference between concentrations found in the animal area and the background was often very small, leading to large uncertainties in the calculation of emission factors. This could be somewhat improved by reducing the analytical accuracy through a longer exposure time or the use of high-volume samplers. This, however would reduce the amount of available data either in time or space, resulting in less information value of the available data.

## Results

### PM10 concentrations

The PM10 concentrations for the animal area from our own measurements ranged from  $< 10 \mu\text{g m}^{-3}$  (detection limit) up to  $69 \mu\text{g m}^{-3}$  (mean:  $26 \mu\text{g m}^{-3}$ ). The PM10 concentrations of a dairy loose housing with cubicles in the Netherlands lie in the similar range with values from  $11$  to  $41 \mu\text{g m}^{-3}$  (MOSQUERA et al. 2012). In contrast, the PM10 concentrations of the measurements taken by Kaasik and Maasikmets (2013) in nine non-thermally-insulated cubicle housings for dairy cows in Estonia were significantly higher; here, average monthly PM10 concentrations ranged from  $27$  to  $123 \mu\text{g m}^{-3}$  (mean:  $65 \mu\text{g m}^{-3}$ ). In studies carried out by HENSELER-PASSMANN (2010) the PM10 concentrations in both cubicle loose housing systems tended to be lower than our own values at  $4$  to  $15 \mu\text{g m}^{-3}$ , whereas the concentrations of a deep litter loose housing were many times higher at  $170$  to  $206 \mu\text{g m}^{-3}$ . While the PM10 concentrations of our own data and those of HEIDENREICH et al. (2008) and MOSQUERA et al. (2012) showed no clear seasonal effects, the PM10 concentrations in studies by Joo et al. (2013) and SCHMIDT et al. (2002) in the warm season were significantly above those in winter. Differences in concentration could be attributable to the size, strength and particle composition of the sources as well as to temperature, relative humidity, wind speed and air exchange rate. While rainfall events have shown a significant PM10 concentration reduction lasting for a few days in dust measurements of a beef cattle

feedlot (BONIFACIO et al. 2011), in our own measurements no effect of rainfall on PM10 concentration was detectable.

No negative emissions occurred, as the mean values of the background PM10 concentrations were lower than each of the corresponding medians in the animal area. This indicates that the analytical procedures were relatively robust and sufficiently precise.

### PM10 emissions

There were pronounced differences in PM10 emissions throughout farms, seasons and 3-day measurement timeframes. At 0.03 to 2.8 g animal<sup>-1</sup> d<sup>-1</sup> the values were within a similar range or slightly higher to the values in the literature for cubicle loose housing by HEIDENREICH et al. (2008) at 0.10 to 1.44 g animal place<sup>-1</sup> d<sup>-1</sup> and SCHMIDT et al. (2002) at 0.12 to 1.99 g LU<sup>-1</sup> d<sup>-1</sup>. MOSQUERA et al. (2012) measured PM10 emission values from 0.08 to 0.41 g animal place<sup>-1</sup> d<sup>-1</sup> on six independent measuring days spread throughout the year in one cubicle housing system in the Netherlands. Somewhat higher PM10 emissions, 2.06 to 2.78 g LU<sup>-1</sup> d<sup>-1</sup>, were measured in a deep litter loose housing system in Germany (HENSELER-PASSMANN 2010). Significantly higher mean PM10 emissions were given by measurements in two naturally ventilated dairy housings in the USA at 11.9 to 15.0 g cow<sup>-1</sup> d<sup>-1</sup> (Joo et al. 2013). In these studies, the PM10 emissions increased as the temperature rose (Joo et al. 2013). In our own measurements in dairy housing with an outdoor exercise area, however, temperature had no significant effect on PM10 emission. PM10 emissions in Swiss pig housings with an exercise yard were significantly higher in summer than in winter (BERRY et al 2005). Consistent with our own measurements, those taken in three seasons by HEIDENREICH et al. (2008), HENSELER-PASSMANN (2010) and MOSQUERA et al. (2012) showed no systematic seasonal effect. Emission data by SCHMIDT et al. (2002) in cubicle loose housing and by HENSELER-PASSMANN (2010) in deep litter loose housing even showed higher PM10 emissions in winter than in the warmer season.

The fact that with the linear mixed-effects model no significant connection was found between the influencing variables investigated, i.e. season, outside temperature and/or relative humidity in the housing, could be due to i) the high uncertainty in emission factors due the small increase in the animal area compared to background concentration, ii) the use of accumulated PM10 readings over each three-day period, with the variation of climatic parameters within the three day period not being shown, or iii) the comparatively small sample size per farm. On the six commercial farms in this study, ammonia was recorded with high temporal resolution in the same measuring segments. A linear mixed-effects model gave outside temperature, wind speed in the housing and milk urea content as significant influencing variables on ammonia emission (SCHRADE et al. 2012).

### PM10 emission factors and extrapolation

The underlying data for our PM10 emission factor are based on a more sizeable number of farms as PM10 emission factors from the literature (Table 3). PM10 measurements on six commercial farms in two out of three seasons form the database for the PM10 emission factor for the loose housing system with solid floors, cubicles and an outdoor exercise area most common in Switzerland. The PM10 emission factors cited by GOODRICH et al. (2006) for free-stall dairy housing and an open-pen area were calculated on the basis of concentration measurements and dispersion modelling on one farm. In the emission inventory of the EUROPEAN ENVIRONMENT AGENCY (2013) PM10 emission factors are differentiated by “dairy cows solid manure” and “dairy cows, slurry based”. These values are based



on studies by TAKAI et al. (1998) in which the PM10 emissions were not measured, but derived from the total suspended particles (TSP) fraction. The emission factors from the Netherlands for cubicle loose housing with perforated floors with or without grazing are based on measurements from four farms (MOSQUERA et al. 2011).

Table 3: Comparison of PM10 emission factors from dairy housing derived from this study with the literature (AAP Average Animal Population; CEPMEIP Co-ordinated European Programme on Particulate Matter Emission Inventories; EEA European Environment Agency; FOEN Federal Office for the Environment; LU livestock unit, 1 LU = 500 kg live weight)

Particulars of housing system	PM10-emission factor	Region	Data basis	Reference
Dairy cows, solid manure (straw based)	1.18 g AAP <sup>-1</sup> d <sup>-1</sup>	Europe	Based on TSP measurements from TAKAI et al. (1998)	EEA (2013)
Dairy cows, slurry based	2.27 g AAP <sup>-1</sup> d <sup>-1</sup>			
Cattle (stock)	1.09 g head <sup>-1</sup> d <sup>-1</sup>	Europa	Not depicted	CEPMEIP (2014)
Dairy cattle (for the year 2012)	1.07 g animal <sup>-1</sup> d <sup>-1</sup>	Switzerland	Not depicted	FOEN (2014)
Free-stall dairy housing	5.0 g LU <sup>-1</sup> d <sup>-1</sup>	Texas, USA	Concentration measurements and dispersion modelling, 1 farm	GOODRICH et al. (2006)
Open pen area of the dairy	11.3 g LU <sup>1</sup> d <sup>-1</sup>			
Dairy cubicle housing with straw mattress	0.99 g animal place <sup>-1</sup> d <sup>-1</sup>	Germany	Emission measurements, 1 farm Emission measurements, 2 farms	HEIDENREICH et al. (2008)
Dairy cubicle housing with rubber mat	0.58 g animal place <sup>1</sup> d <sup>-1</sup>			
Deep litter dairy loose housing with grazing (May to October)	1.48 g LU <sup>-1</sup> d <sup>-1</sup>	Germany	Emissionsmessungen, 3 Betriebe	HENSELER-PASSMANN (2010)
Dairy cubicle loose housing with grazing (May to October)	0.16 g LU <sup>-1</sup> d <sup>-1</sup>			
Dairy cubicle loose housing with perforated floors with grazing	0.32 g animal place <sup>-1</sup> d <sup>-1</sup>	The Netherlands	Emissionsmessungen, 4 Betriebe	MOSQUERA et al. (2011)
Dairy cubicle loose housing with perforated floors (year-round indoor housing)	0.40 g animal place <sup>-1</sup> d <sup>-1</sup>			
Cubicle dairy loose housing with solid floors and outdoor exercise area	0.64 g animal <sup>-1</sup> d <sup>-1</sup> 1) or 0.48 g GV <sup>-1</sup> d <sup>-1</sup>	Switzerland	Emissionsmessungen, 6 Betriebe	This study

1) Conversion using KTBL Livestock Calculator (2014)

At 0.64 g animal<sup>-1</sup> d<sup>-1</sup> the PM10 emission factor derived from our own measurements is significantly lower than the values used to date in Swiss and European inventories at 1.07 g head<sup>-1</sup> d<sup>-1</sup> (FOEN 2014) and 2.27 g AAP<sup>-1</sup> d<sup>-1</sup> (EUROPEAN ENVIRONMENT AGENCY 2013). At 0.40 g animal<sup>-1</sup> d<sup>-1</sup> (year-round indoor housing) and 0.32 g animal<sup>-1</sup> d<sup>-1</sup> (with grazing), emission factors derived from PM10 measurements in four perforated cubicle loose housing systems (MOSQUERA et al. 2011) are lower than the emission factor based on our own measurements and hence also significantly below the values used in the inventories. The average PM10 emission from more up-to-date measurements in the Netherlands is significantly lower ( $P < 0.05$ ) than the emission factor reported in MOSQUERA et al. (2011) (MOSQUERA et al. 2012). Hence the emission factors for dairy cattle so far used in the inventories seem to be too high.

Our PM10 emission values are the currently best data for Swiss dairy cow emissions. They are representative for the most important housing system with cubicles, solid floors and an outdoor exercise

area. For other systems, no specific data is available, and they were thus, included using the same emission factors. As no differentiation could be made between housing systems when extrapolating emissions, the trend in PM10 emissions runs parallel to the cattle population. Were PM10 emissions to be differentiated according to housing system and production technique, a different trend might become evident due to the increase in loose housing at the expense of tied housing over the course of the year, by analogy with extrapolations for ammonia emissions (SCHRADE and KECK 2012).

## Conclusions

For the first time, PM10 concentrations and emissions on dairy-cattle farms with cubicles, natural ventilation and an outdoor exercise area were systematically quantified in different seasons on a large database with measurements on six farms.

PM10 concentrations in the animal area (housing; the outdoor exercise area or the cubicle access aisle/outdoor exercise area) were in many cases slightly over or within the background concentration range. Accordingly, the calculated emissions are fraught with a high level of relative uncertainty. Within the farms, differences in emission levels are recognisable between the seasons and between measuring periods. With the present volume of data and based on the very slight differentiation from the background concentration, there was no statistically significant relationship to the influencing variables investigated. A detailed qualitative study of dust samples for source apportionment analogous to that which HENSELER-PASSMANN (2010) carried out in dairy housings or HUANG et al. (2013) conducted in cattle feedlots could serve to help determine relevant variables influencing PM10 emissions. Online measurement methods with a high temporal resolution would be necessary to model daily schedules or short-term activities. While grazing and alpine pasturing are frequently practised in Switzerland, PM10 emission data from alp and pasture or from housing used only occasionally have not as yet been quantified. Future PM10 measurements should, thus, investigate changes in housing emission levels during half-day and full-day grazing and permanent access to pasture. Further type and processing of bedding as well as effects of different feed ratios (hay or silage) has to be quantified.

The emission factor derived from our own measurements as well as other emission factors from more recent measurements in dairy cubicle loose housing, indicate that the emission factors used to date in inventories overestimate PM10 emissions in dairy farming.

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