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Ammonia emissions from a dairy housing and wastewater treatment plant quantified with an inverse dispersion method accounting for deposition loss

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Abstract

Ammonia (NH₃) emissions negatively impact air, soil, and water quality, hence human health and biodiversity. Significant emissions, including the largest sources, originate from single or multiple structures, such as livestock facilities and wastewater treatment plants (WWTPs). The inverse dispersion method (IDM) is effective in measuring total emissions from such sources, although depositional loss between the source and point of measurement is often not accounted for. We applied IDM with a deposition correction to determine total emissions from a representative dairy housing and WWTP during several months in autumn and winter in Switzerland. Total emissions were 1.19 ± 0.48 and 2.27 ± 1.53 kg NH₃ d⁻¹ for the dairy housing and WWTP, respectively, which compared well with literature values, despite the paucity of WWTPs data. A concurrent comparison with an inhouse tracer ratio method at the dairy housing indicated an offset of the IDM emissions by <20%. Diurnal emission patterns were evident at both sites mostly driven by changes in air temperature with potential lag effects such as following sludge agitation. Modelled deposition corrections to adjust the concentration loss detected at the measurement point with the associated footprint were 22-28% of the total emissions and the cumulative fraction of deposition to emission modelled with distance from the source was between 7-12% for the measurement distances (60-150 m). Although estimates of depositional loss were plausible, the approach is still connected with substantial uncertainty, which calls for future validation measurements. Longer measurement periods encompassing more management activities and environmental conditions are required to assess predictor variable importance on emission dynamics. Combined, IDM with deposition correction will allow the determination of emission factors at reduced efforts and costs, thereby supporting the development and assessment of emission reducing methods and expand the data availability for emission inventories.

Introduction

Ammonia (NH_3) emissions have detrimental effects on soils and aqueous ecosystems via acidification, eutrophication, and subsequent loss of biodiversity but also on air quality due to the formation and growth of particulate matter which impacts human health (Fowler et al., 2015; Galloway et al., 2021; Sutton et al., 2011). Agriculture and more specifically, the livestock sector is the largest emitter of NH_3 with 80-85% globally and approx. 90% in Switzerland (Kupper et al., 2015; van Damme et al., 2021). In countries with high livestock densities, primary sources include animal housing and manure application, but other point sources such as wastewater treatment plants (WWTPs) are also densely distributed in Switzerland.

Often NH_3 emissions from point sources are quantified by combining concentration measurements in the housing with estimated ventilation rates (Calvet et al., 2013). Many studies have measured NH_3 emissions from dairy housings, while calculated emission factors are used to normalise emissions by animal, taking different climatic conditions into account (Flesch et al., 2009; Hempel et al., 2016; Schrade et al., 2012). However, difficulties remain when comparing these emissions due to significant effects of different housing types, management and measurement methods (Poteko et al., 2019), as well as the timing and duration of measurements (Kafle et al., 2018). For example, loose housings with natural ventilation are the prevalent dairy housing system in western Europe (Sommer et al., 2013), yet annual mean emissions for these systems despite standardisation by livestock unit ($\text{LU} = 500 \text{ kg live weight}$) vary by orders of magnitude from $3.4 \text{ g NH}_3 \text{ LU}^{-1} \text{ d}^{-1}$ to $98.4 \text{ g NH}_3 \text{ LU}^{-1} \text{ d}^{-1}$ (Poteko et al., 2019; Wu et al., 2012) depending on the climate and farm conditions during the measurements, as well as the measurement method itself. Not all methods are equally suitable for different housing configurations adding further variability to the already wide emission range. Although the tracer ratio method is considered state-of-the-art, it has limited applicability for farm-scale measurements due to its extensive experimental setup (Mendes et al., 2015). It is generally used for single structures as it must be carefully tailored to scale with the source strength in the case of multiple or inhomogeneous sources. Furthermore, the frequently used tracer, sulphur hexafluoride (SF_6), is the most potent and extremely long-lived greenhouse gas making it unsuitable for longer measurement durations necessary to capture emission variability.

WWTPs are another widespread source in Switzerland (64% of WWTPs are small to medium-sized) with high uncertainties in the national emissions inventory. They represent a similar source configuration and measurement difficulties for NH_3 emissions as dairy housings (Kupper et al., 2013). Generally, emissions are poorly quantified worldwide due to their spatial arrangement and temporal variations. The major source of NH_3 is expected from the sludge line, although there is a paucity of data on whole plant and individual source emissions. The sludge line comprises of thickening of primary and excess sludge, anaerobic treatment in a digester, and subsequent storage of the anaerobically stabilised liquid sludge. Optionally, the sludge can be dewatered and stored before disposal. For storage of liquid sludge in open

tanks, emission peaks are expected due to regular agitation for further transport and processing. Samuelsson et al. (2018) estimated total emissions of $4.3 \text{ g NH}_3 \text{ PE}^{-1} \text{ yr}^{-1}$ for a plant with 805000 person equivalents (PE) in Sweden, of which 66% likely originated from the sludge line. Sutton et al. (1995a) estimated up to $27 \text{ g NH}_3 \text{ PE}^{-1} \text{ yr}^{-1}$ for a medium-sized facility (165000 PE) relying on several assumptions. Upscaled emissions from laboratory studies on wastewater or from livestock manures can be a further source for emissions estimates (Dai et al., 2015).

Measuring gas emissions from open structures is challenging due to the source configuration and scale, as emissions are heterogeneous and dynamic in space and time (Bühler et al., 2022). Schrade et al. (2012) measured NH_3 emissions from six dairy loose housings with outdoor exercise areas in Switzerland using a tracer ratio method throughout the year. Although this method is considered state-of-the-art, it has limited applicability for farm-scale measurements due to its complex experimental setup and high operating costs for extended measurement durations (Mendes et al., 2015). This method is suited for single structures as it must be carefully tailored to scale with the source strength in the case of multiple or inhomogeneous sources, such as from WWTPs, which complicated of may impede its application. Another building-scale measurement type is the inverse dispersion method (IDM) which combines concentration measurements up- and downwind of the source with results from modelling the inverse dispersion of the plume to relate the measured concentration difference to the source emission rate (Flesch et al., 2004). This method has been successfully applied to determine emissions from whole farms, including animal housings (Flesch et al., 2005; Flesch et al., 2007; Flesch et al., 2009; Harper et al., 2010), feedlots (Flesch et al., 2007; McGinn et al., 2007; McGinn et al., 2016), and manure management (Baldé et al., 2018; Flesch et al., 2013; Grant et al., 2013; Kamp et al., 2021; Lemes et al., 2022). Using this method in a controlled methane release experiment in a barn Gao et al. (2010) achieved a recovery rate, i.e. the fraction of the modelled release rate to the emitted trace gas, of 0.93-1.03 and more recently Lemes et al. (2023) determined recovery rates between 0.66-0.91.

The dispersion model relies on micrometeorological assumptions including constant wind speeds, large fetch, and homogeneous terrain. Since buildings disturb the wind field, the measurement location needs to be outside of this disturbance. Downwind locations should be at a distance of >10 times the building height (Gao et al., 2010; Harper et al., 2011). Other requirements include homogeneous sources and an emission plume that is clearly distinguished from ambient concentrations. The latter is particularly challenging in Switzerland, where emission sources are small (farms with 22 dairy cows on average) but farms are densely dispersed (Federal Statistical Office, 2020) resulting in weaker emission plumes compared to high background levels. Despite this and the sometimes non-ideal terrain conditions, we previously demonstrated the effective application of IDM to quantify methane emissions under these constraints by optimising the filtering criteria (Bühler et al., 2021; 2022). The optimised method yielded a good comparability with the inhouse tracer ratio method (iTRM) as a reference (inhouse referring to the release of the tracer inside the housing) (Mohn et al., 2018), with differences in methane emissions of 1 -

8%, which were well within the uncertainty ranges of both methods (i.e., <10% for tracer ratio and <24% for IDM (Bühler et al., 2021; Mohn et al., 2018)).

A backward Lagrangian stochastic (bLS) model was applied to model the inverse dispersion of the plume assuming that the target gas is inert, which is satisfied for methane but not for NH₃. Ammonia is very reactive and soluble with a high affinity to adsorb to surfaces and thus experiences greater losses mostly by deposition (Loubet et al., 2009; Schrader and Brümmer, 2014b). As a general rule, 20-25% of total emitted NH₃ and ammonium (NH₄⁺) is expected to deposit within 1 km of the source (Asman et al., 1998; Asman, 1998; Asman and van Jaarsveld, 1992; Loubet et al., 2006; Loubet et al., 2009; Sutton et al., 1998b). Measurements along deposition transects from pig farms showed deposition losses of 6% up to 500 m (Bajwa et al., 2008), but Loubet et al. (2017) found that the cumulative deposition within only 200 m downwind on a fertilised grassland site ranged from 4% to 34% and therefore may not always be negligible. This deposition loss with distance from the source reduces measured concentrations from which emissions are calculated. It is worth noting that this concentration reduction depends on the corresponding areal extent, i.e. footprint, of each measurement and thus differs from the ratio of total deposition to emissions of a source. Although line-integrating instruments for IDM are located 100-150 m from the source (i.e., 10x the building height), the deposition loss is often not accounted for in emission measurements, which leads to a systematic underestimation when determining total emissions from source areas (Häni et al., 2018).

Horizontal deposition profiles vary strongly with source height and depend on wind speed, atmospheric stability, surface resistances and roughness, as well as surface compensation points (Flechard et al., 2013). Since deposition data from direct flux measurements are limited (Famulari et al., 2004; Ferrara et al., 2012; Sintermann et al., 2011; Swart et al., 2022; Vendel et al., 2023; Zöll et al., 2016), deposition models remain widespread, which modify the concentrations with a calculated deposition velocity. The velocity can either be a static value or derived from a series of resistances including aerodynamic, boundary layer and canopy resistances, the latter consisting of stomatal and non-stomatal resistances (Sutton et al., 1995b). Häni et al. (2018) conducted a source release experiment using IDM which had recovery rates between 0.69 and 0.91 without correcting for deposition. They then demonstrated that a simple deposition algorithm could achieve a recovery rate of 1 using realistic values for the deposition velocity. The algorithm adjusts the emissions calculated without deposition to include deposition loss based on a user defined deposition velocity. Therefore, the optimum deposition velocity and thus canopy resistance needed to achieve a recovery rate of 1 was determined from multiple runs to match a known emission source during the NH₃ release experiments. Without a known source, canopy resistances are based on parameterisations assuming a single 'big leaf' canopy surface and are associated with large uncertainties (Massad et al., 2010). In addition to impacts of environmental conditions, uncertainties depend on changing vegetation and surface types and can remain high even with more detailed models which require highly-resolved input data (Flechard et al., 2013; Walker et al., 2020). For practical emissions monitoring, estimating canopy resistance is time and computationally intensive

providing only marginally improved results and uncertainties. A simpler approach using maximum and minimum canopy resistance and hence deposition to constrain emissions determined by IDM may prove useful without incurring further uncertainties.

The aim of this study was to 1) determine emission estimates for building-scale sources in different emission sectors (i.e., dairy housing and WWTP) as reported for the Swiss NH₃ emissions inventory using longer IDM measurements over weeks to months, 2) apply a simple approach to account for dry deposition losses to determine total emissions of building-scale NH₃, 3) compare NH₃ emissions by IDM with iTRM for the dairy housing, and 4) model the depositional loss in relation to total emissions for the distances relevant to the IDM instrument setups. Here we present measurements from a dairy loose housing and the first facility-scale measurements of NH₃ emissions from a representative WWTP in Switzerland.

Methods

Site descriptions

Loose dairy housing

Measurements were conducted at an experimental loose housing for dairy cows in Aadorf, Switzerland (47.489175° N, 8.919663° E, 544 m altitude) (Mohn et al., 2018). The building is naturally ventilated with the long axis positioned perpendicular to the prevailing wind directions (NE and SW) for optimal ventilation. While the building itself is situated on a flat plain extending >1 km to the SW, the surrounding topography consists of a descending slope (9% over 50 m height difference) 220 m to the NE, a small forest 200 m west and several buildings and trees (<15 m height) to the north, as well as other livestock housings beyond a radius of 250 m but within 600 m, plus surrounding pastures with grazing cattle and sheep (Figure 1).

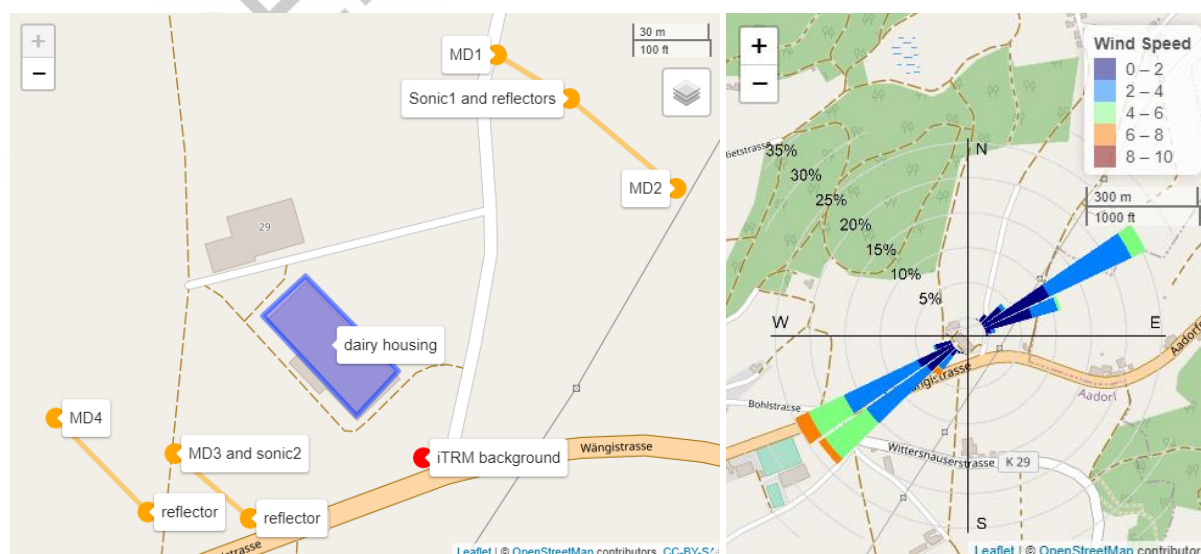


Figure 1. Map of the loose dairy housing (emitting area in blue) and instrument setup up and downwind of the housing (left) consisting of miniDOAS sensors and reflectors (MD1 to 4, orange dots and lines for measurement paths), sonic anemometers (sonic1 and 2), as well as the inlet for background concentration measurements needed for the iTRM (red dot). Wind speeds and frequencies during the entire measurement period (right) indicated two main directions. Map data © OpenStreetMap contributors. Please see the online version for the full colours.

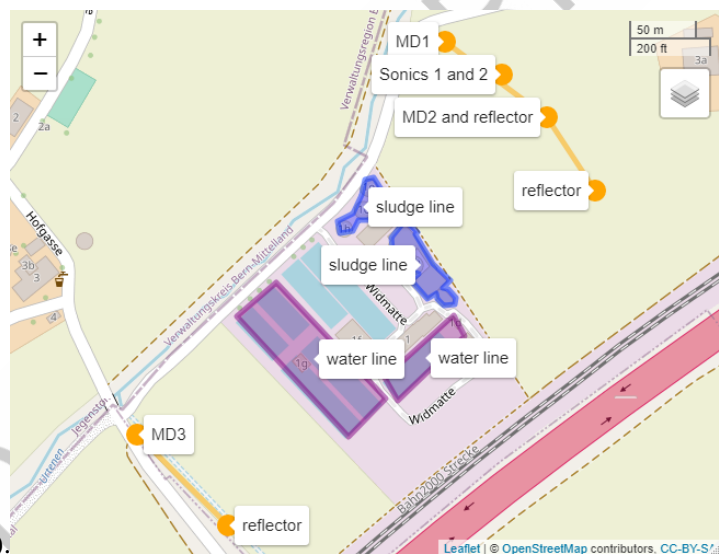
The total area of the dairy housing was 1205 m² and consisted of two compartments for 20 dairy cows each with straw mattress cubicles and a solid floor covered by a rubber mat (KURA P, Gummiwerk KRAIBURG GmbH, Tittmoning, Germany). The milking and waiting area were situated between the compartments along with other technical installations and an office. Each housing compartment was equipped with a cross channel which ran perpendicular to the building's long axis and led to an adjacent underground slurry store to the SW of the housing and was separated from the housing compartments by rubber flaps. The slurry store consisted of two compartments (one for each housing compartment) of 252 m³ in total with a solid cover and four openings. The housing compartments were not thermally insulated. Ventilation was adjusted with flexible curtains along façades, which during the measurement periods were varied from completely open, through a combination of completely closed on the NE side and partially or completely closed on the SW side, to completely drawn on all sides. The curtains were fully closed for the last four days of the first measurement period and during the entire second measurement period. More information on the study site can be found in (Bühler et al., 2021; Poteko et al., 2018).

Management routines included milking twice daily (05:30 and 16:30 local time), as well as dung removal with stationary scrapers 12 times per day. During the measurements 40 primiparous and multiparous lactating Brown Swiss and Swiss Fleckvieh cows were housed in the building. The diets of the dairy cows consisted of either hay, maize pellets, and a mixture of maize and bean pellets, or grass silage, maize silage, and hay. Both diets were supplemented with concentrates individually allocated by an automatic feeder according to milk yield and lactation stage. The average body weight was 701 kg in autumn and 685 kg in winter with a mean daily milk yield of 23.2 kg and 25.7 kg, respectively. The cows had no access to the pasture or outdoor exercise areas during the experiment.

Wastewater treatment plant

Measurements were conducted at a medium-sized WWTP (47.055620° N, 7.539515° E, 507 m altitude) which used a conventional activated sludge treatment with complete nitrification and denitrification. The plant processed waste of 43534 PE, which corresponded to 33126 connected inhabitants plus industrial waste. The site encompassed an area of 2.18 ha and consisted of multiple emitting structures totaling 5617 m² including sand traps, primary and secondary clarifiers, activated sludge tanks, thickener and digester towers (total volume of 2200 m³), a gas storage tank and open sludge storage tanks. It was selected based on the suitability of the location for IDM measurements i.e., minimal topographic obstructions (Figure 2). No large undulations or additional emission sources were present within 1 km in

the dominant wind direction (SW). The plant applied conventional activated sludge treatment with complete nitrification and denitrification. The thickened sludge had a dry matter content of 4% before entering the anaerobic digester where it resided for 20 days. It was then further dewatered again to 8% dry matter content through the addition of flocculants by means of a rotary screen. Afterwards the sludge was stored in open tanks with a total volume of 1960 m³ (632 m³ in use at the time) and a surface of 331 m², which were regularly agitated typically in the morning before part of the sludge was transported to another facility for further treatment and incineration. Mean operational data during the measurement period were comparable to annual means (see Table SI2.1 for average operational data during the measurement periods). The mean incoming N load from ammonium (NH₄-N) during the measurement period was 350 kg NH₄-N d⁻¹ in total or 7.8 g NH₄-N PE⁻¹ d⁻¹ which is within the expected range for



Swiss WWTPs (Kupper and Chassot, 1999).

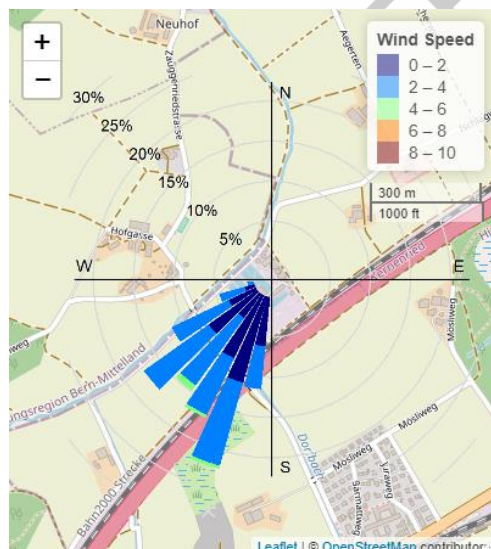


Figure 2. Map of the wastewater treatment plant with emitting structures marked on the left (blue shading for the sludge line and purple for the water line). Yellow dots and lines represent the miniDOAS sensors and measurement paths (MD1 to 3) and sonic anemometers. Wind speeds and frequencies during the

measurement period are shown on the right. Map data © OpenStreetMap contributors. Please see the online version for the full colours.

Measurement campaigns

Line-integrated concentration measurements

Measurements at the dairy housing were split into two periods of 36 days (autumn) and 23 days (winter) from September to December 2018, but only one measurement period at the WWTP lasting 21 days from late September to mid-October 2019. Ammonia concentrations were measured using miniDOAS instruments placed up- and downwind of the emission sources (Figures 1 and 2). The instruments are open-path optical devices which measure line-integrated gas concentrations between a light source and detector by UV absorption (200-230 nm wavelengths) (see Sintermann et al., 2016 for more details). The path length, here 50 m, is determined by the placement of the reflectors and the miniDOAS, which houses both the light source and detector in an environmentally controlled container. Downwind instruments were placed at a distance of approx. 10 times the maximum building height (i.e., the dairy housing was 8.5 m and the WWTP up to 15 m high) to avoid wind flow disturbance by the structures (Harper et al., 2011). At the dairy housing one set of instruments was placed 120 m NE of the building, while the second set was around 60 m SW due to topographic restrictions, while at the WWTP the up and downwind placements were 100 – 150 m.

In-house tracer ratio method

The measurement setup and processing is described in detail by Mohn et al. (2018) and results of the tracer release experiment for methane at this dairy housing were presented in (Bühler et al., 2021). Here the results of the NH_3 emission measurements are shown. In brief, each housing compartment was dosed with a different tracer gas (i.e., sulphur hexafluoride (SF_6) and trifluoromethyl sulphur pentafluoride (SF_5CF_3)) to identify emissions from each location. The tracer and target (NH_3) gas concentrations were concurrently quantified from a representative air sample collected in the housing using a GC-ECD and a Picarro analyser (G2301, Picarro Inc., USA). Emissions were calculated based on the ratio of the background-corrected gas concentrations of each target to tracer gas multiplied with the dosed mass flow of the tracer gases. This method assumes that the dispersion of the tracer gas behaves the same way as the target gas and mimics its emissions (Demmers et al., 2001). The iTRM comparison measurements consisted of 2 intervals lasting 4 or 8 days during each IDM measurement period. Background concentrations were sampled 30 m SE of the housing (Figure 1). The iTRM data represented average emissions for 10 min intervals for each compartment, which were summed and then averaged to 30-min means before being compared with the corresponding 30-min IDM measurement intervals.

Meteorological measurements

Three-dimensional sonic anemometers (Gill Windmaster, Gill Instrument Ltd., Lyngington, Hampshire, UK) were installed up- and downwind of the source structures at around 1.4 m height to measure the wind flow and turbulence for the bLS model and logged at 10 Hz. Since the wind direction

at the dairy housing alternated between SW and NE, only data from the respective downwind sonic was used for bLS model. Additional meteorological data (air temperature, pressure, precipitation) were obtained at both sites using an OTT WS700 weather station (OTT Hydromet GmbH, Germany) and at the dairy housing a nearby weather station (Tänikon, MeteoSwiss) provided additional meteorological data. Coordinates of the instruments and source locations are needed for the bLS model and were collected with a handheld GPS (Trimble Pro 6T, Trimble Navigation Limited, Westminster, USA). At both sites methane was also measured and the results were published in Bühler et al. (2021; 2022).

Inverse dispersion modelling

Inverse dispersion modelling is a micrometeorological method to determine gaseous emissions in a downwind plume from sources of a known dimension and spatially constrained area. The concentration difference ΔC (mg m^{-3}) between background levels in upwind C_{BG} and downwind C_{DW} measurements of a source is combined with the bLS model based on the measured turbulence characteristics to calculate the dispersion factor D_{bLS} (s m^{-3}) needed to estimate an emission source strength Q in mass over time (eq. 1).

$$Q = \frac{C_{DW} - C_{BG}}{D_{bLS}} \quad (\text{eq. 1})$$

The bLS model based on (Flesch et al., 2004), which is a surface layer model covering distances <1 km, was used to calculate the dispersion factor (eq. 2) from the total backward trajectories of the line-integrated concentration measurements. The measurement path was approximated by a series of points spaced 1 m apart and propagated backward to the source area and hence is proportional to the ratio of the simulated concentration to the emission rate E , i.e., source strength per area, $(C/E)_{bLS}$ as follows,

$$D_{bLS} = \frac{(C/E)_{bLS}}{A_{source}} = \frac{1}{N_{Traj}} \sum TD_{inside} \left(\frac{2}{w_0} \right) * \frac{1}{A_{source}} \quad (\text{eq. 2})$$

For each point and measurement interval, 250000 backtrajectories were calculated and analysed for touchdowns (TD_{inside}) within the source area (A_{source}), which provide the touchdown location coordinates and vertical velocity (w_0).

Trace gas emissions determined by this IDM method have previously been compared with an inhouse tracer ratio method at this dairy housing site and have been discussed in Bühler et al. (2021). Means from both methods were within the uncertainty range of the tracer method ($<10\%$).

Deposition modelling

The bLS model modified by (Häni et al., 2018) (R package *bLSmodelR* available at <https://www.agrammon.ch/documents-to-download/blsmodelr/>) was used to calculate dry deposition by multiplying the measured concentration increase with a proportionality constant called the deposition velocity (Wesely and Hicks, 2000). The deposition velocity was approximated by a resistances approach (Sutton et al., 1995b), whereby the velocity is the inverse of the sum of several resistances to deposition that must be overcome. These include the aerodynamic, boundary layer and canopy resistances across the

surface-atmosphere interface, which vary with micrometeorological, canopy and surface properties of the ecosystem. The first two resistances are calculated, while the latter can be either laboriously derived from models or depends on parameterisations which can vary significantly at the canopy level where the exchange processes take place (Flechard et al., 2011). Instead, a more practical approach was applied, in which the average between the possible upper and lower boundaries was used to estimate deposition. For the maximum possible deposition velocity ($v_{d,max}$), the canopy resistance was set to 0, i.e., assuming that the surface acts as a perfect sink, while the effects of boundary layer and canopy resistances on deposition were not considered for the minimum case. This method allows a simple, yet reasonable approximation of the likely deposition and corrected emissions therefore represent an average of the possible lower and upper flux magnitudes. The deposition flux F_d was modelled following Flechard et al; Häni et al. (2018) by

$$F_d = -v_d * C_{TD} \quad (\text{eq. 3})$$

Where v_d is the deposition velocity and C_{TD} was the modelled concentration at the touchdown of a trajectory at the adsorbing surface. The aerodynamic resistance (R_a) usually included in the v_d calculation was omitted, since v_d was investigated for the layer between the modelled effective ground level i.e., z_0 plus the displacement height d and the adsorbing surface z_0' and was therefore implicitly accounted for in the bLS dispersion model. Since the canopy resistance R_c was set to 0 in the maximum deposition flux model, the deposition velocity v_d was calculated as

$$v_d = \frac{1}{R_b + R_c} \rightarrow v_{d,max} = \frac{1}{R_b} \quad (\text{eq. 4})$$

The boundary layer resistance R_b was calculated from equation 5 in Flechard et al. (2010) using the input variables: friction velocity u^* , canopy temperature, roughness length z_0 , and ambient air pressure. Although wet deposition was not taken into account, there were only few periods of rainfall or fog with valid measurements, hence the underestimation of calculated emissions during such periods is likely small. Also, the deposition of particulate NH_4^+ was not investigated. The R package *bLSmodelR* was used for all of the modelling (Häni, 2021).

In addition, based on the average conditions during the measurement campaign at the dairy housing, the fraction of the emission deposited within different distances (10, 20, 50, 100, 200 m) downwind of the source was modelled. From a simple mass balance perspective, the total horizontal flux through a vertical plane P_x at distance x which is perpendicular to the average wind direction (and sufficiently wide to capture the entire plume) will match the net flux, i.e., the sum of emissions Q_i and the integrated deposition flux $F_{d,upw} = \iint_{x < x_{\text{plane}}} F_d dx dy$, upwind of the plane:

$$\iint_{P_x} \bar{u} \bar{c} dy dz = \sum_i Q_i + F_{d,upw} \quad (\text{eq. 5})$$

Where $\bar{u} \bar{c}$ is the temporal average of the product between the horizontal wind speed component along the main wind direction u and the concentration c .

If we solely focus on a single source (and its contribution to the horizontal flux) and divide equation 5 by its source strength Q , we obtain the following relationship:

$$\iint_{P_x} \frac{\overline{uc}}{Q} dy dz = 1 + \frac{F_{d,upw}}{Q} \quad (\text{eq. 6})$$

The ratio $F_{d,upw}/Q$ represents the ratio between the total deposition (related to the source emission) between source and plane P_x and the source emission strength. The ratio of the horizontal flux and the source strength is obtained from bLS model results as:

$$\frac{\overline{uc}}{Q} = \left(\frac{\overline{uc}}{E}\right)_{bLS} A_{source}^{-1} \quad (\text{eq. 7})$$

With A_{source} being the area of the emitting source and E the emission rate per area. Due to biases stemming from both the discretization and the stochastic nature of the bLS model, the integral of the horizontal flux from model results without accounting for deposition will never be exactly equal to 1. Thus, equation 5 has been reformulated and discretised to provide:

$$\frac{F_{d,upw}}{Q} = A_{source}^{-1} \sum \sum \left(\left(\frac{\overline{uc}}{E}\right)_{bLS,dep} - \left(\frac{\overline{uc}}{E}\right)_{bLS} \right) \Delta y \Delta z \quad (\text{eq. 8})$$

Where the subscripts bLS and bLS, dep stand for model results without deposition and with deposition, respectively. The plane's extensions were taken as 200 m in the crosswind direction and 80 m from model ground in the vertical direction. The discretization in the vertical was done in 20 steps on a logarithmic scale, while the discretization in the crosswind direction was equally spaced with 1 m distances.

Uncertainty analysis

The uncertainty analysis followed Bühler et al. (2021), whereby the uncertainty ε_Q of the mean IDM emission rate \bar{Q}_i over a time period Δt was estimated from the standard deviation SD of consecutive measurements of increasing lengths from 1 h to 45 h using the longer first measurement period at the dairy housing (eq. 9) and represents the upper uncertainty boundary of the mean emission rates across the entire campaign.

$$\varepsilon_Q(\Delta t) = 2 * SD\{\bar{Q}_i(\Delta t)\} \quad (\text{eq. 9})$$

Since the SD included true variations of Q , such as diel variability, the uncertainty, $\varepsilon_Q(\Delta t)$, corresponds to a 95% confidence interval of an assumed constant Q over time and is thus larger than the true uncertainty of the varying Q .

Data processing and assessment

For the IDM measurements, the 3D wind vectors (u , v , w) underwent two-axis coordinate rotations, corrections for a known bug affecting the w wind component of the Gill Windmaster instruments (Gill Instruments, 2016), and were averaged to 30 min intervals. To correct for possible underestimation of NH_3 concentrations from loss by dry deposition the maximum dry deposition was modelled (Section 2.4), which represented an upper total emission boundary compared to the uncorrected emissions, which represented the lower boundary.

The bLS model cannot deal with periods of low wind speed, very high stability or instability, as well as extreme turbulence which are typically filtered out (Flesch et al., 2014; Gao et al., 2009; Harper et al., 2010). However, to avoid substantial data loss due to the meteorological conditions, a custom filtering procedure was applied based on observed variations of u and v , as well as literature values as described in Bühler et al. (2021). The filters at the dairy housing and the WWTP included friction velocity $u^* > 0.1 \text{ m s}^{-1}$ or $> 0.05 \text{ m s}^{-1}$ and for canopy height (z_H) and roughness length z_0 , $z_H / 100 < z_0 < z_H / 3$ or $z_0 < 0.1$, respectively, while the ratio of the SD of the along-wind (σ_u) or crosswind speeds (σ_v) to u^* , i.e., σ_u / u^* and σ_v / u^* were < 4.5 at the dairy housing and < 6 at the WWTP, and at both sites $|L| > 2 \text{ m}$ for the Obukhov length and the Kolmogorov constant of the Lagrangian structure function was $3 < C_0 < 10$. Finally, only wind directions perpendicular to the instrument paths and therefore including the plume were retained (e.g., SW at the WWTP or SW and NE at the dairy housing). This resulted in 282 and 379 valid half-hourly measurements corresponding to a data loss of 80% and 62% during autumn and winter, respectively, at the dairy housing and 241 intervals at the WWTP corresponding to an average of 69% data loss. Since data loss occurred mostly at night, daytime data retention was as high as 40 to 60% (Figure SII.2). There was a general bias towards more daytime data and greater loss at dawn and dusk due to the rapid changes in atmospheric turbulence conditions and the higher frequency of fog which obscures the sensors during these periods. The iTRM included 290 valid datapoints with corresponding 30-min IDM means for the comparison split across the autumn (190 intervals) and winter (100 intervals) measurements.

The WWTP consisted of several structures that acted as sources with different emission strengths within the general source area, therefore a relative source weighting factor w_i based on known literature values from Samuelsson et al. (2018) for a WWTP and Kupper et al. (2020) for pig slurry tanks representing the sludge storage emission was applied to each source (see Bühler et al., 2022). Each area A_i for each source emission Q_i with its corresponding modelled dispersion factor D_i was combined to a single source emission Q_{tot} based on the measured total concentration difference ΔC_{tot}

$$Q_{tot} = \frac{\Delta C_{tot}}{\sum_{i=1}^N \left(\frac{w_i D_i}{A_i} \right)} \sum_{i=1}^N w_i \quad (\text{eq. 10}).$$

Welch t-tests and Pearson correlations were used to compare means and establish synchronous correlations with predictor variables, while a cross correlation function was used to investigate lagged correlations. A reduced major axis regression was used to compare the independent IDM and iTRM results. Other R packages used for data processing and plotting included *ibts* (Häni, 2022), *RgoogleMaps* (Loecher and Ropkins, 2015), *scales* (Wickham and Seidel, 2020), *ggplot2* (Wickham, 2016), *dplyr* (Wickham et al., 2022), *leaflet* (Cheng et al., 2022) and *openair* (Carslaw and Ropkins, 2012).

Results

Dairy housing

The meteorological conditions were generally representative for the respective seasons with mean \pm standard deviation (*SD*) temperatures of 11.2 ± 4.6 °C and 3.6 ± 4.1 °C and mean monthly precipitation of 46 mm and 97 mm, respectively, for the autumn and winter periods at the dairy housing. Air temperatures were in the upper ranges during the autumn period and there were multiple events with high wind speeds particularly during winter at the dairy housing (Figure SII.1 and SII.3).

NH₃ emissions and diel profiles

The mean (\pm *SD*) NH₃ emissions during the autumn and winter measurement periods are shown in Table 1 for both emissions without accounting for dry deposition, which indicated the lower emission range, and with corrections for maximum dry deposition representing the upper range. The averages of these ranges present a mean estimate of the total emissions resulting in 1.57 ± 0.59 kg NH₃ d⁻¹ and 0.81 ± 0.38 kg NH₃ d⁻¹ during autumn and winter, respectively. The concentration increase required to correct for dry deposition raised the uncorrected emissions by approx. 22% and the overall uncertainty of the IDM was 27%. This resulted in mean (\pm *SD*) corrections of the emissions to account for deposition of 0.28 ± 0.10 and 0.15 ± 0.07 kg NH₃ d⁻¹ for the autumn and winter periods, respectively, with mean modelled deposition velocities of 1.1 ± 0.52 cm s⁻¹ and 1.4 ± 0.55 cm s⁻¹. The measurement timeseries (Figure 3) presents the emissions accounting for mean deposition loss. Mean (\pm *SD*) NH₃ concentrations as measured by the miniDOAS for the entire measurement duration were 10.65 ± 6.86 µg NH₃ m⁻³ and 19.19 ± 14.23 µg NH₃ m⁻³ for up- and downwind instruments, respectively (Figure SII.4).

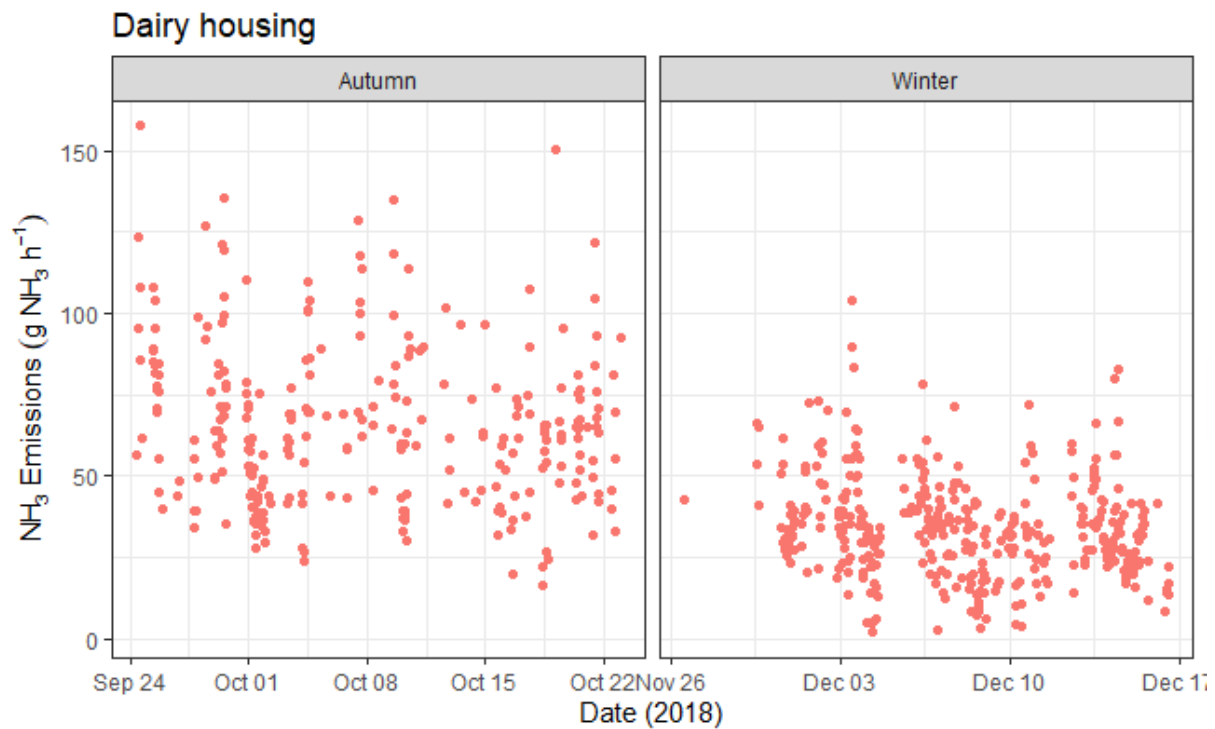


Figure 3. Timeseries of NH_3 emissions including averaged deposition loss for the first (left) and second (right) measurement periods at the dairy housing.

Table 1. Mean and standard deviation (SD) of non-gap filled emissions for the entire dairy housing and normalised by livestock units.

Measurement period	Type of deposition correction	Mean \pm SD NH_3 emissions	
		($\text{kg NH}_3 \text{ d}^{-1}$)	($\text{g NH}_3 \text{ LU}^{-1} \text{ d}^{-1}$)*
Autumn	None	1.28 ± 0.5	22.8 ± 8.9
	Max	1.85 ± 0.69	33.0 ± 12.3
	Mean	1.57 ± 0.59	28.0 ± 10.5
Winter	None	0.66 ± 0.31	12.0 ± 5.6
	Max	0.97 ± 0.44	17.6 ± 8.0
	Mean	0.81 ± 0.38	14.7 ± 6.9
All	None	0.97 ± 0.40	17.6 ± 7.2
	Max	1.41 ± 0.56	25.6 ± 10.2
	Mean	1.19 ± 0.48	21.6 ± 8.7

* Livestock units ($\text{LU} = 500 \text{ kg live weight}$)

Profiles indicated a median diurnal pattern of NH_3 emissions with mean deposition corrections from the dairy housing with peak emissions during the afternoon which were more pronounced in autumn compared to winter (Figure 4). Emissions correlated best with air temperature ($r = 0.51$, $p < 0.001$), which slightly increased with a lag of 1-2 h for both periods and wind directions (up to $r = 0.55$, see Table S11.1). Due to the relatively larger loss of data during the night, only the daytime emission profiles were considered robust.

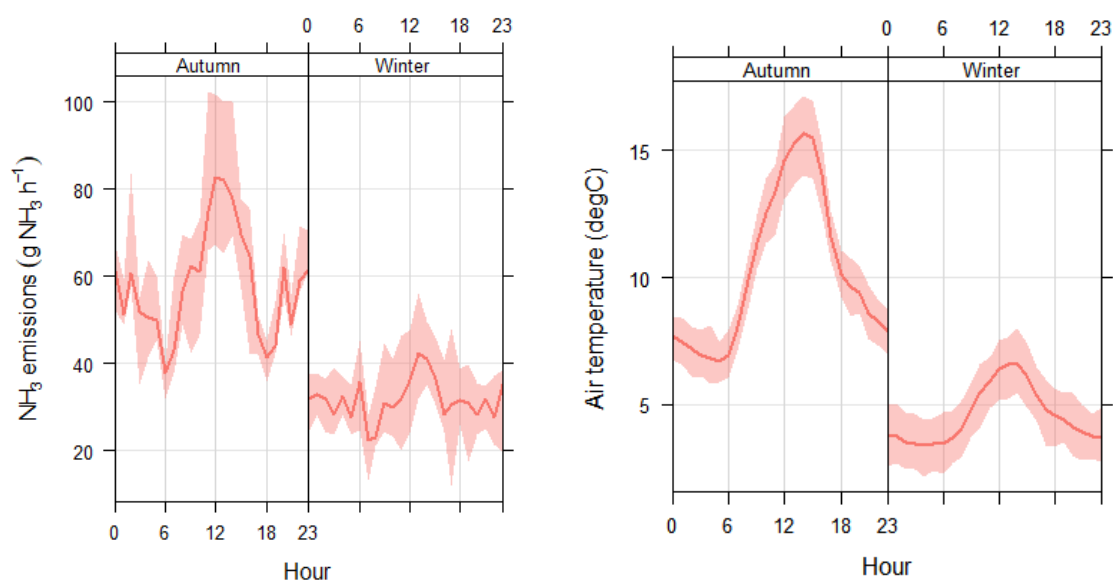


Figure 4. Diurnal patterns of median NH₃ emissions with average deposition correction (left) and the mean air temperature profile (right) at the dairy housing for autumn and winter periods, respectively.

Wind sector differences

Winds were predominantly from the NE and the SW during the autumn and winter measurement periods, respectively (Figure SII.5). Winds speeds were only higher from the SW during the winter with $3.9 \pm 1.4 \text{ m s}^{-1}$ compared to $1.9 \pm 0.6 \text{ m s}^{-1}$ from the NE while speeds were approx. $2.1 \pm 0.8 \text{ m s}^{-1}$ from both directions in autumn (Figure 5). Although wind speeds were different only in winter, emissions in autumn were 20% higher with NE winds over SW winds (Figure SII.6a). During this period, emissions additionally correlated with u^* with NE but not the SW winds (Figure SII.6b).

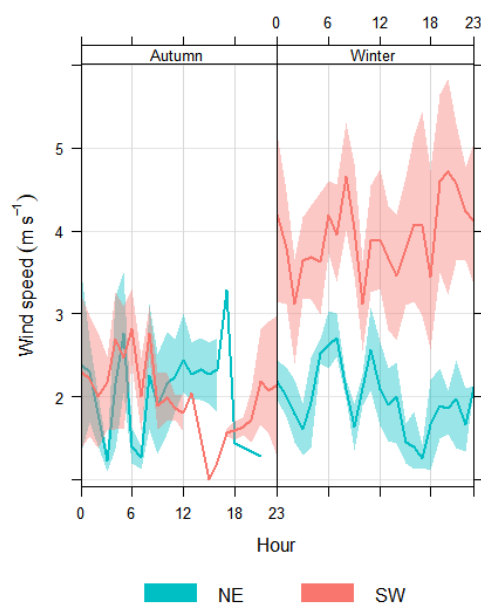


Figure 5. Mean diel profiles of wind speed separated by wind direction (northeast, NE, and southwest, SW) during autumn and winter periods.

Comparisons with iTRM

Daily averaged NH_3 emissions quantified by iTRM from both compartments were summed and compare to the entire dairy housing emissions quantified by IDM. Mean \pm SD emissions by iTRM were $1.90 \pm 0.58 \text{ kg NH}_3 \text{ d}^{-1}$ and $1.00 \pm 0.31 \text{ kg NH}_3 \text{ d}^{-1}$ for autumn and winter measurements, respectively (SI Figure SI1.7). Mean IDM using only concurrent time intervals with iTRM were $1.53 \pm 0.59 \text{ kg NH}_3 \text{ d}^{-1}$ and $0.83 \pm 0.34 \text{ kg NH}_3 \text{ d}^{-1}$, hence the IDM means differed from those reported in Table 1. Therefore, mean corrected emissions by IDM for these respective periods varied up to 20% from the iTRM results (Figure 6) which represented a statistically significant difference (t -statistic = 5.73, $p < 0.001$).

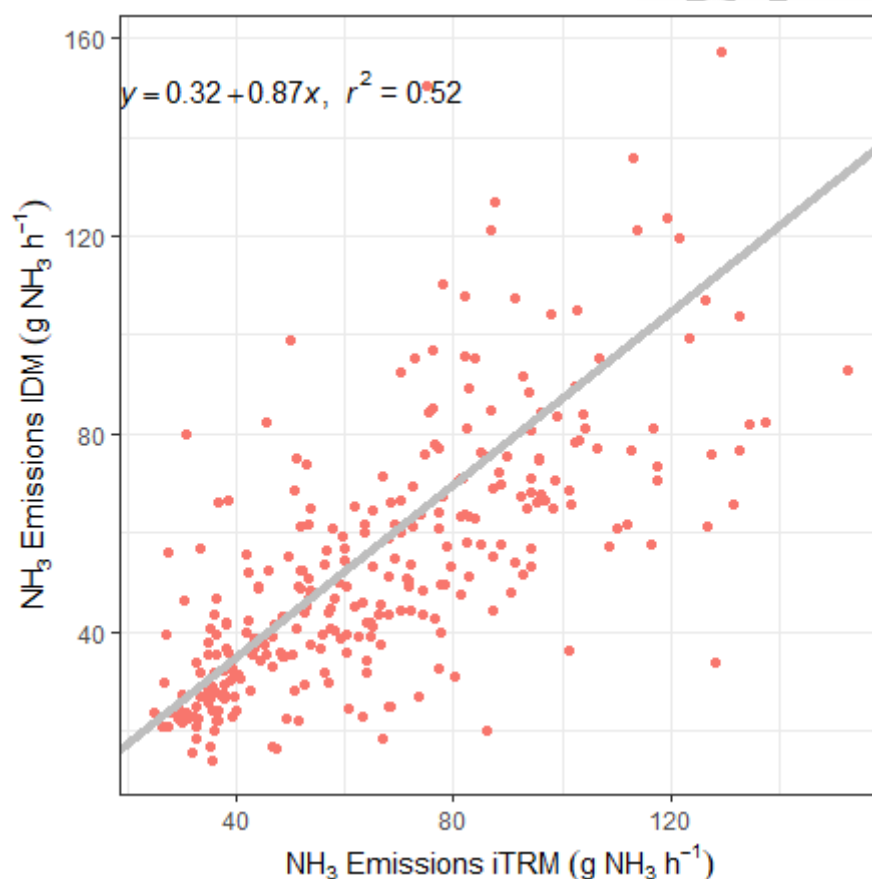


Figure 6. Scatterplot of corresponding 30-min iTRM and IDM measurements of total NH_3 emissions ($\text{g NH}_3 \text{ h}^{-1}$) at the dairy housing with the linear regression and coefficient of determination.

Modelled deposition loss with distance from the source

The fraction of mean deposition to emission with distance from the source based on the measurement setup at the dairy housing was highest for stable atmospheric conditions and lowest for unstable conditions (Figure 7). For each respective condition the highest relative deposition within the first 50 – 150 m from the source, as relevant at our sites, ranged from 9 to 12% and 7 to 9%, respectively.

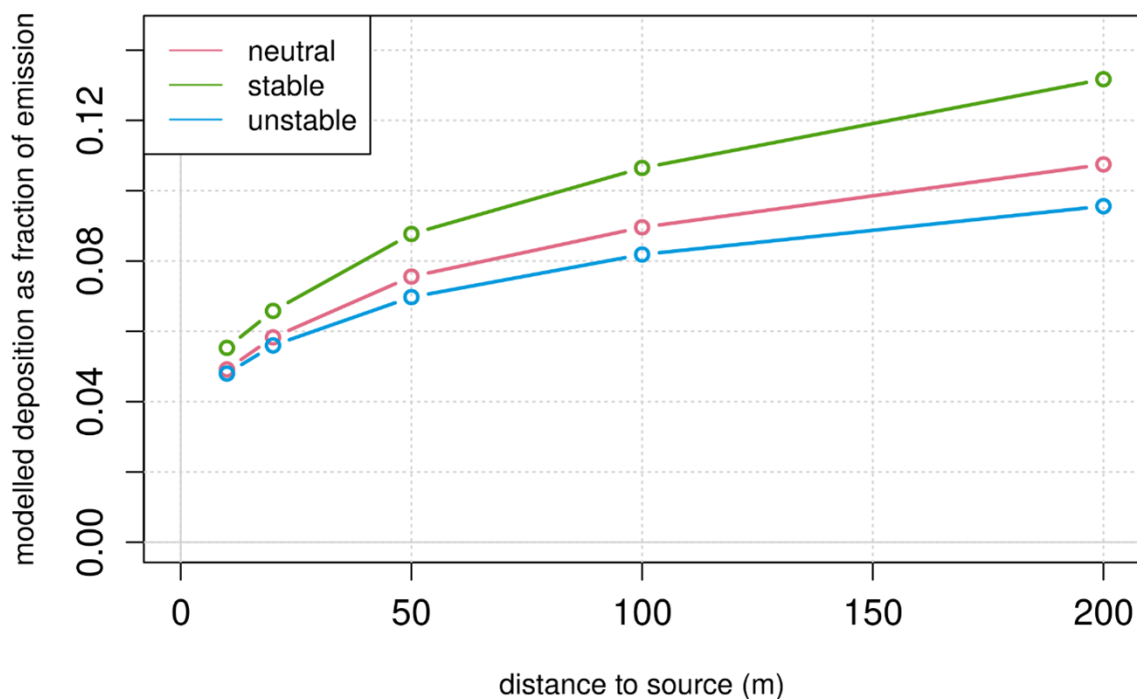


Figure 7. Relative NH₃ deposition rate as a cumulative fraction of total emissions deposited with distance from the source for different atmospheric stability conditions (defined as Obukhov length, L , $-500 < L < 50$ for unstable, $|L| > 500$ for neutral, and $10 < L < 500$ for stable).

WWTP

The meteorological conditions during the measurements at the WWTP in 2019 were representative for the season with a mean \pm SD temperature of 9.38 ± 2.58 °C and monthly precipitation of 126 mm, albeit with periods where temperatures were in the upper ranges or with high wind speeds (Figure SI2.1).

Mean NH₃ emissions

Average (mean \pm SD) NH₃ emissions from the WWTP assuming no deposition loss (lower range) were 1.77 ± 0.84 kg NH₃ d⁻¹, while emissions corrected for maximum deposition (upper range) were 2.79 ± 1.28 kg NH₃ d⁻¹. Averaged emissions representing a mean corrected estimate of deposition were 2.27 ± 1.53 kg NH₃ d⁻¹ (Figure 8 top), which resulted in a deposition correction of 28% with an overall uncertainty of IDM at 24%. The per person equivalent of the mean emissions corresponded to 18.4 g NH₃ PE⁻¹ yr⁻¹, which represented 0.63% of the nitrogen (NH₄-N) inflow. The mean deposition correction for the emissions was 0.51 ± 0.23 kg NH₃ d⁻¹ and the mean deposition velocity was 0.65 ± 0.40 cm s⁻¹. Mean (\pm SD) concentrations as measured by the miniDOAS instruments up- and downwind of the WWTP were 4.71 ± 0.82 µg NH₃ m⁻³ and 7.19 ± 1.55 µg NH₃ m⁻³, respectively (Figure SI2.3).

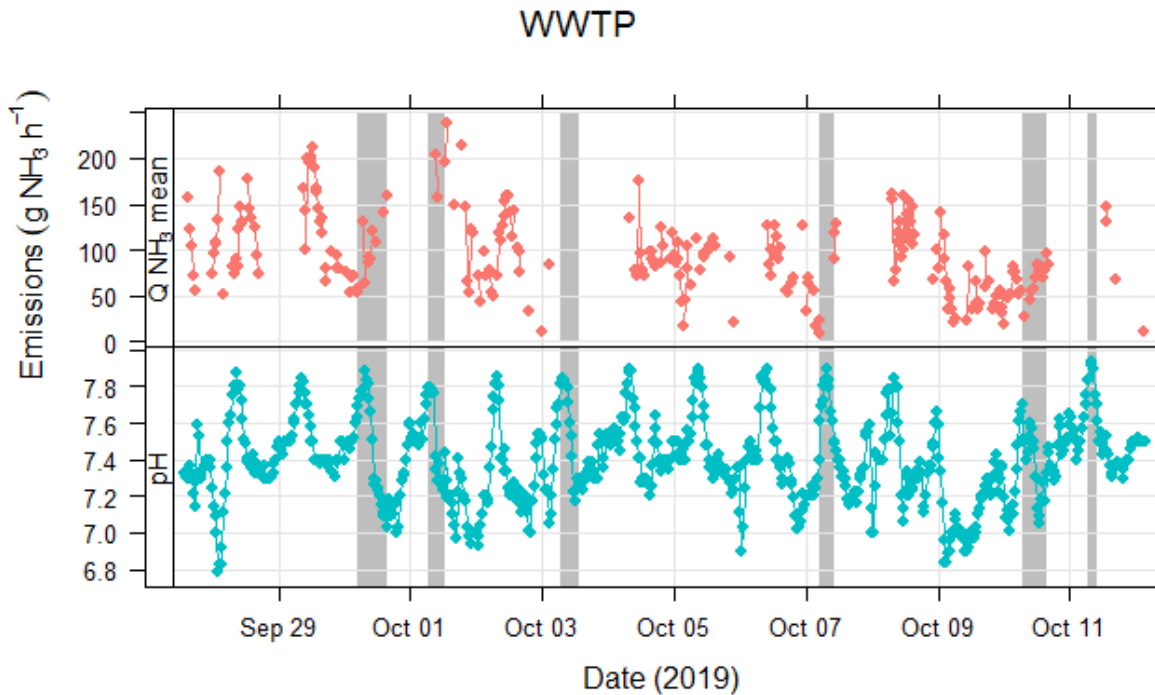


Figure 8. Timeseries of mean deposition-corrected NH_3 emissions (top) and pH (bottom) at the inflow of the WWTP with agitation events (grey shaded bars) of the slurry storage tanks during the measurement period. Note that pH, was measured at the inflow to the water line and not at the sludge tanks.

Emission responses to management activities

Although a pH above 7 increases the partitioning of NH_3 to NH_4^+ and the temporal pattern of emissions was similar to that of pH (mean 7.4 ± 0.2) measured at the inflow (Figure 8 bottom), there was only a weak concurrent correlation (Pearson's $r = 0.15$, $p < 0.05$). Using a cross correlation function to determine asynchronous leads and lags, statistically significant lagged relationships were found up to 5.5 h after changes in pH with peak correlations at 3 h ($r = 0.52$, F-statistic = 29.3, $p < 0.001$). All sewage sludge tank agitators were operated on Monday mornings (06:00 – 12:00 local time on 30th September and 7th October), while one tank was additionally agitated more frequently during the week. However, no direct changes in emissions could be observed in response to agitation, as the two prominent agitation events coincided with measurement gaps.

As with the dairy housing, diurnal patterns in emissions were also evident and emissions peaked again around midday lagging behind management activities affecting pH or agitation times by 3-6 h, both of which peaked early morning (Figure 9). Emissions correlated moderately with air temperature (Pearson's $r = 0.48$, $p < 0.001$) which improved with a 3-hour offset ($r = 0.75$, $p < 0.001$), while incoming solar radiation also showed a high instantaneous correlation ($r = 0.67$, $p < 0.001$). A moderate correlation was also observed between emissions and wind speed ($r = 0.30$, $p < 0.001$).

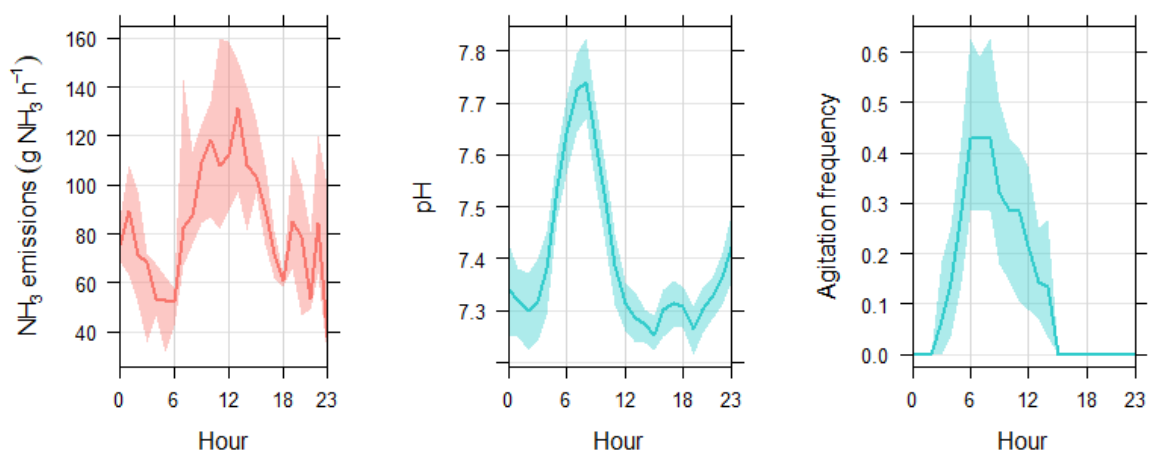


Figure 9. Diel profiles of median NH₃ emissions with deposition corrections (left), mean pH measured at the inflow (middle), and counts of agitation events mixing the sludge tanks (right).

ACCEPTED MANUSCRIPT

Discussion

Dairy housing emissions

There were up to 20% differences in IDM-based emissions between main wind directions during the autumn measurements with higher emissions for NE compared to SW winds, although these were still within the estimated uncertainty range, i.e., <27%. Increased emissions with NE winds, might have resulted from slightly higher wind speeds further indicated by the increased but still weak correlation of emissions with u^* for this direction only. However, in the winter period, wind speeds were even higher from the SW but did not result in differences in emissions. A more likely explanation was the diel shift in wind direction which was observed only in autumn reflecting daily emission patterns, whereby NE winds were more prevalent during the day when emissions were higher and SW winds during the evening. However, we could not exclude the possibility that emissions may have been affected by different curtain settings, which were either half or fully open during autumn but fully closed in winter. Therefore, it is possible that the curtain settings affected the emission measurements from their position which increased aeration but also their presence affecting source geometry due to changes in turbulence conditions.

The diurnal pattern with higher daytime emissions, which was more prominent during the autumn measurement at the dairy housing, was probably mostly driven by air temperature (Bougouin et al., 2016; VanderZaag et al., 2015). Other studies found similar patterns for dairy housing emissions with daytime peaks being highest in summer, then transitional seasons and lowest in winter (Flesch et al., 2009; Saha et al., 2014). While wind speed showed no distinct diel patterns, it was higher during the winter period when emissions were lower further indicating the dominant effect of temperature compared to wind speed. This follows similar findings by Schrade et al. (2012), in which total emissions across six farms showed some correlation with wind speed but the effects of air temperature and milk urea content were more dominant. This has also been observed for different housing types and manure management systems (Bougouin et al., 2016; Liao et al., 2021; Saha et al., 2014).

The comparison of NH_3 emission estimates between IDM and iTRM showed good agreement with an overall deviation of <20%, i.e., within the uncertainty of IDM. The possible underestimation by IDM with an averaged deposition correction may indicate that the correction was too low. In fact, the estimates with corrections for maximum deposition more closely matched emissions determined by iTRM (<3% difference). Although instrument precision and dispersion modelling may contribute to this difference, our results indicate that deposition is likely the main contributor. This conclusion agrees with results from Bühler et al. (2021) who observed smaller differences between IDM and iTRM of <10% for methane emissions, which act as a good reference since deposition is not relevant (Lassman et al., 2020). Even though the slurry tank contributions were minor since it was covered, it is worth noting that iTRM did not include these emissions.

In terms of magnitude, our measurements were within typical ranges from the literature for loose housings with solid floors and no exercise yard during transition seasons (i.e., spring and autumn) and winter, which ranged from 3.5-92.9 g NH₃ LU⁻¹ d⁻¹ and 5.2-87.9 g NH₃ LU⁻¹ d⁻¹, respectively (Poteko et al., 2019). Compared to ranges published by Schrade et al. (2012) in Switzerland, the IDM measurements agreed well for the respective seasons, i.e., 28.0 ± 10.5 in autumn and 14.7 ± 6.9 g NH₃ LU⁻¹ d⁻¹ in winter compared with 16-44g NH₃ LU⁻¹ d⁻¹ and 6-23 g NH₃ LU⁻¹ d⁻¹, respectively. Other seasonal means for naturally ventilated barns with solid floors from a meta-analysis by Bougouin et al. (2016), which covered multiple measurement methods, were comparable with autumn and winter averages around 31.6 ± 4.3 and 43.5 ± 29.9 g NH₃ cow⁻¹ d⁻¹, respectively, compared to our 39.2 ± 14.7 g NH₃ cow⁻¹ d⁻¹ and 20.2 ± 9.5 g NH₃ cow⁻¹ d⁻¹ (note the different units, as LU was not available). Their winter mean emissions were higher due to two studies in the UK and USA. Including all seasons, mean annual emissions were 47.7 g NH₃ cow⁻¹ d⁻¹, therefore the autumn and winter means were a factor of 0.66 and 0.91 of the annual mean, respectively, while the seasonal weighting factors derived from means in Schrade et al. (2012) were 0.96 and 0.49, respectively. The factors of seasonal to annual mean emissions were closer to those from Schrade et al. (2012), in which winter emissions were a factor of 0.6-0.8 lower than the autumn ones, while the values in this study were 0.53 of those in autumn. Saha et al. (2014) measured seasonal averages for winter and autumn ranging between 7.9-35.3 and 5.5-54.7 g NH₃ LU⁻¹ d⁻¹ for naturally ventilated housings using the carbon dioxide balance method. Their seasonal average weightings were 0.54 and 0.75 for winter and autumn, respectively, compared to the annual mean and the mean ratio of winter to autumn, i.e., 0.71, were similar to those listed above. Although comparing very well to other studies, it is likely that our daily means had a slight bias towards higher values due to the lower representation of transitional (dawn and dusk) and nighttime periods when emissions are low because atmospheric conditions during these times deviate from the assumed ideal conditions for IDM leading to data rejection. In addition to the daily data representativeness, it is important to consider the seasonal impacts when comparing emission measurements and applying annual emission factors.

Specifically compared to other line-integrated measurements from Flesch et al. (2009) who used a bLS model and measured whole farm, as well as separate source structures by relocating the sensors, at free-stall barns also equipped with semi-drawn curtains, our measurements were in similar seasonal ranges and ratios, as their barn emissions amounted to 21.2 ± 10.7 g NH₃ animal⁻¹ d⁻¹ in autumn and 11.7 ± 10.2 g NH₃ animal⁻¹ d⁻¹ in winter. They also found similar but still weak diurnal patterns for the barns modulated only somewhat by air temperature in autumn and winter with u^* being of minor importance. Moreover, they found that the management practices across the different farms were the most influential on emissions (i.e., naturally ventilated free-stall barns, sand bedding, regularly scraped barn floors) and that the same IDM technique allowed good comparisons between locations.

WWTP emissions

Total emissions at the WWTP were higher than at the dairy housing and although fewer data were available, there was a much more prominent diurnal profile. Higher emissions are expected during the day due to higher N loads in the wastewater from human activity, but also from the higher temperatures driving emissions through increased molecular diffusion of NH_3 which enhances emissions from both the sludge and water line. However, peak air temperatures were often reached in the late afternoon while emissions peaked around noon leading to the higher offset correlation. Incoming solar radiation peaked at noon and showed a high synchronous correlation with emissions. Since many of the emission sources at the WWTP had open surfaces that were exposed to the sun (especially the sludge storage tanks, but also much of the water line), localised surface heating likely facilitated this higher correlation compared to ambient air temperature. With emissions being affected by weather conditions, as with the dairy housing, it is important to consider the impact of seasonality and representativeness of the measurement period for future comparisons. The emission profile also matched changes in pH and tank agitation following a time offset of 3-5.5 hours. Although NH_3 emissions are controlled by pH which determines the speciation of volatile NH_3 and non-volatile NH_4^+ with even microsite variations leading to observable emission changes (Hafner et al., 2013; Kim and Or, 2019), the pH was measured at the inflow of the water line, which is expected to contribute only 34% to total emissions (Samuelsson et al., 2018), as opposed to the storage tanks by which point the pH will have changed. More likely, agitation of the open sludge tanks caused larger contributions to the total emission variations, as they are expected to produce the most emissions (Samuelsson et al., 2018), assuming they act as an open slurry tank (Kupper et al., 2020). However, agitation did not occur daily and regrettably, no direct effects of the agitation could be observed on emissions due to gaps in the measurements directly following the larger agitation events. Following the immediate initial emission response after disturbance in a swine slurry storage tank, Blanes-Vidal et al. (2012) observed a 1.5-3.5 h delay with a subsequent NH_3 emission increase without additional disturbance. This was explained by the development of a pH gradient during the transient-state conditions after disturbance and lasted up to 48 h. Therefore, the morning agitation of the sludge most likely promoted movement to and subsequent release of NH_3 at the surface and other buffers changing the pH profile and hence contributed to emission increases with delayed and lasting effects (Kupper et al., 2021). Longer measurement periods would be required to collect sufficient data and events to definitively determine the impact on emissions, as well as more expansive monitoring of facility parameters.

The NH_3 emissions of the WWTP ($94.9 \pm 44.1 \text{ g NH}_3 \text{ h}^{-1}$) were smaller than measurements by Samuelsson et al. (2018) who reported $400 \pm 100 \text{ g NH}_3 \text{ h}^{-1}$ for a much larger WWTP (805000 PE) in Gothenburg and using individual samplings at each processing stage during the day. When normalised by PE our measurements were over 4 times higher, i.e., $18.4 \text{ g NH}_3 \text{ PE}^{-1} \text{ yr}^{-1}$ compared to $4.3 \text{ g NH}_3 \text{ PE}^{-1} \text{ yr}^{-1}$ at the WWTP in Gothenburg. However, that plant included an activated sludge system comprised of nitrifying trickling filters and moving bed biofilm reactors for post-denitrification, which can be effective

in lowering total NH_3 levels (Colón et al., 2009; Xue et al., 2010). The sludge after digestion was also dewatered with a centrifuge without intermediate liquid storage (personal communication S. Tumlin, Gryaab AB, Gothenburg). The solids were then stored for three weeks in open-air stockpiles before being transported off-site. Data from Aguirre-Villegas et al. (2019) on anaerobically digested dairy manure with and without solid-liquid separation using a screw press or centrifuge suggested that NH_3 emissions from the solid fraction can be lower by at least one order of magnitude compared to unseparated manure. If these findings are extrapolated to our WWTP to compare anaerobically digested liquid sewage sludge in open stores (i.e., without solid-liquid separation) with the storage of dewatered sludge in the Gothenburg plant, the discrepancy in PE normalised emissions seems plausible. Furthermore, our results come closer to the estimate by Sutton et al. (1995a) of $27 \text{ g NH}_3 \text{ yr}^{-1}$ for a WWTP in the UK.

Due to the lack of emission data from WWTPs, another comparison may be drawn with emissions from slurry stores. The matrix of sewage sludge is comparable to that of pig slurry, although it contains less NH_4^+ and the formation of a natural surface crust is less pronounced, which is expected to enhance emissions (Kupper et al., 2020). However, sewage sludge stores are agitated more frequently than slurry tanks (Kupper et al., 2015), likely enhancing NH_3 emissions. Emissions from slurry stores with different types of slurry and tank configurations are well documented (Kupper et al., 2020), allowing an estimation of the sewage sludge tank emissions based on pig slurry emission factors for comparable stores (i.e., $0.17 \text{ g NH}_3 \text{ m}^{-2} \text{ h}^{-1}$). Upscaled to the same source area as the WWTP, these emissions would amount to $80 \text{ g NH}_3 \text{ h}^{-1}$ which compare well with the measured emissions of $95 \pm 64 \text{ g NH}_3 \text{ h}^{-1}$, further confirming that the majority of measured emissions likely originated from the sludge storage.

Dry deposition corrections

Maximum dry deposition was modelled at both sites and the range averages between the maximum and no deposition resulted in concentration corrections of 22-28% with higher percentages at the WWTP. At the dairy housing, the correction differed with wind direction by 19% for NE winds but 27% and 23% for SW winds in autumn and winter, respectively. Although emissions were lower from the SW, atmospheric conditions were mostly stable which extended the footprint and allowed for a greater proportion of the emissions to be deposited. Since the relative correction with SW winds was greater in autumn than winter, it may be feasible that the curtain position, which was more frequently open during autumn, could have affected emission estimates by altering the turbulence conditions and hence possibly also the deposition corrections. Alternatively, the lower correction of 19% during both periods with NE winds may have resulted from the shorter measurement distance for NE emissions reducing the cumulative deposition loss. Overall, the absolute corrections were lower during winter even with higher wind speeds since temperatures and hence emissions were also low.

While both the heights of the emission source and measurements can affect the proportion deposited with distance, the bLS model assumes an emission at surface level, while the measurement heights and

distances from the emission source varied between 1.2-1.7 m and 60-150 m, respectively, depending on site and direction. To estimate the relative deposition loss with distance, we modelled the fraction of deposition to emission for each stability condition for the dairy housing site. For the measurement distance of 150 m, the model indicated that <12% of the emissions were deposited during stable and <9% during unstable conditions, while minimal deposition at 50 m ranged from 7-9%. This proportion is in contrast to the emission correction value discussed previously, which adjusted the percent concentration loss detected at the measurement point and is different to the percent deposited to emitted gas, since the concentration-associated footprint changes the contribution weightings of both source and deposition areas.

Mean deposition velocities (i.e., the inverse of the sum of all resistances) where $R_c = 0$ at both sites were comparable to or lower than estimates from other studies (Aksoyoglu and Prévôt, 2019; Phillips et al., 2004; Schrader and Brümmner, 2014a) for agricultural surfaces, i.e., 0.65-1.55 cm s^{-1} and therefore boundary layer resistances were 0.64-1.54 s cm^{-1} . The model produced highest velocities at the dairy housing with SW winds during winter when emissions were lower, but wind speeds were higher, also evident from higher correlations with u^* . In contrast, Phillips et al. (2004) found higher daytime v_{dep} in summer than winter of $3.94 \pm 2.79 \text{ cm s}^{-1}$ and $2.41 \pm 1.92 \text{ cm s}^{-1}$, respectively, when emissions were higher even though wind speeds were lower than in winter. The surfaces between the source and measurement paths at both sites consisted of pastures and fallow fields which led to low roughness lengths z_0 of $0.01-0.03 \pm 0.01 \text{ m}$. The higher value of $0.03 \pm 0.01 \text{ m}$ was only during NE winds at the dairy housing, which traversed a taller pasture.

Although most studies rely on simple resistance-based deposition models (Nemitz et al., 2000) and only few use bi-directional models with a calculated compensation point (Massad et al., 2010; Sutton et al., 1998a), the emission component downwind of a plume will be negligible for calculating the deposition loss relative to the total emission of a point source (McGinn et al., 2007). However, direct measurements of net NH_3 fluxes within the downwind plume would allow a better comparison but to date only very few data of net flux measurements are available (Swart et al., 2023 and references therein). Overall, the relative deposition values at the given measurement distances, surface roughness, stability conditions, and source heights were close to the ranges from studies including results of bi-directional models (Asman et al., 1998; Loubet et al., 2006; Shen et al., 2016; Spindler et al., 2001; Walker et al., 2008; Yi et al., 2021). For comparable conditions, Asman et al. (1998) estimated a dry deposition loss within 100 m of 10% with greater amounts expected over rough surfaces with more turbulence due to higher deposition velocities. Using a N deposition model Schou et al. (2006) calculated cumulative losses from a dairy farm resulting in only 5% deposition within 100 m, 10% in 300 m, and 12% by 500 m, while Fowler et al. (1998) estimated 3-10% deposition within 300 m from a poultry farm based on a transect of concentration measurements from which the canopy resistance for the deposition model was determined by a regression fit. Estimates of dry deposition using resistance and dispersion models based

on emission measurements from a hog farm in North Carolina (barns with forced ventilation and a slurry lagoon) were found to be even lower, i.e., 2.9-7% within 500 m and added up to only 16.6% with a distance of 2500 m (Bajwa et al., 2008). Although pig slurries can produce slightly higher NH₃ emissions from lagoons (Kupper et al., 2020), their deposition velocities were an order of magnitude lower than observed here, which the authors explained by the tendency of the deposition model used to underpredict under the given field conditions. Another study at a pig farm in South China estimated monthly means of 4.1-14% NH₃ deposition at 500 m applying passive samplers and a multi-resistance model (Yi et al., 2021). A multiapproach study downwind of a beef feedlot in southern Alberta also found relative deposition amounts of approx. 14% at 500 m using resistance modelling and flux gradient measurements (McGinn et al., 2016), while Staebler et al. (2009) found dry deposition to account for 6-12% loss within 1 km of the same feedlot using airborne concentration measurements and a deposition model. Similarly, Shen et al. (2016) determined 8% deposition at 1 km from a feedlot in Victoria, Australia. Generally, the greater the source emissions the longer the distance of deposition (Fowler et al., 1998; Shen et al., 2016). It is worth noting that these values represent individual deposition gradients and differ from top-down estimates of total deposition budgets which include the entire surroundings of a source which can help to reduce uncertainties at regional scales (Griffis et al., 2019). Our modelled relative deposition loss compared well with these studies but highlighted the variability due to different surface properties and estimation methods of emissions, as well as models needed to derive deposition fluxes. Hence, there is a need to further constrain deposition estimates through improved models or validation by direct measurements of deposition fluxes within emission plumes.

Overall, IDM allows continuous measurements of multiple gases such as methane and NH₃ at sub-daily resolutions with lower labour costs and without interfering with local operations, making it easily deployable for longer periods to capture a wider set of environmental and management conditions. Using line-integrated measurements also offers greater representation of emissions considering wind and dispersion impacts than point measurements. Corrections for the downwind deposition loss ideally require bi-directional modelling, which is complex with high computational demands often not accessible to practical users. Furthermore, due to the considerable reliance on parameterizations, it frequently does not yield a practical reduction in uncertainty. We demonstrated that the deposition loss can be reasonably well estimated using a simple approach to constrain total emissions that still provides meaningful information, while maintaining sufficient data quality, especially considering the need for more data from buildings or sites with multiple emission sources under a variety of conditions. These emission measurements are needed to verify inventory estimates, but more urgently, to assess potential emission reducing technologies and management methods to curb emissions (Insausti et al., 2020). Particularly when testing new strategies, it is vital that emissions are not transferred elsewhere onsite or substituted with other pollutants, e.g., greenhouse gases, therefore whole farm or whole site emissions must be quantified. With accessible IDM modelling tools and simple deposition corrections, a wider adoption of

this method could improve the data availability and implementation of emission monitoring and control strategies to achieve much needed advances to decrease NH₃ emissions and improve N use efficiency.

Conclusion

This study demonstrated the application of IDM accounting for deposition losses to determine total NH₃ emissions from large-scale structures with multiple sources, such as animal housings and WWTPs, the latter of which were the first such measurements in Switzerland. The mean deposition-corrected NH₃ emissions agreed with comparable literature data where available. Clear diurnal patterns were observed, which were likely driven by air temperature and to a smaller degree wind speed, as well as management practices with indications of lagged responses. Longer datasets spanning several months across all seasons would be required to determine the contributions of the predictor variable variations and capture more representative management activities. This could be achieved with IDM's continuous measurement operations but can be biased towards daytime periods due to requirements of specific atmospheric and site terrain conditions.

Currently, the main limitation of IDM is the relatively high uncertainty introduced by the deposition corrections which can also limit the ability to link emissions to specific activities if the effects on emissions fall within the uncertainty. We applied a simple deposition model to correct emissions assuming maximum possible deposition which we averaged with emissions assuming no deposition. These mean deposition-corrected IDM estimates were lower compared to a reference tracer ratio method albeit still within the overall uncertainty but possibly underestimating deposition. Even though the modelled deposition to emission fractions with distance compared well to previous studies, reducing the uncertainty of determining dry deposition or conducting validation experiments of deposition models are needed to further improve the usefulness of IDM. With improved deposition corrections and longer measurement durations, IDM provides an excellent tool to evaluate complex facility-scale NH₃ emissions needed to assess potential management activities and technologies to increase the implementation of emission abatement strategies.

Data availability statement

Due to the data volume only the 30-min averaged data that support the findings of this study are openly available in Zenodo (DOI: 10.5281/zenodo.8047581 for the dairy housing data and DOI: 10.5281/zenodo.8032908 for the WWTP data). The raw data are available from the corresponding author, AV, upon reasonable request.

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Implication statement

Ammonia emissions must be measured to determine emissions factors and reporting national inventories. Measurements from structures like farms and industrial plants are complex due to the many different emitting surfaces and the building configuration leading to a poor data availability. Micrometeorological methods provide high resolution emission data from the entire structure, but suffer from uncertainties, as the instruments must be placed at a distance from the structure resulting in a greater loss of the emitted ammonia via dry deposition before it reaches the measurement. This study constrains such emission measurements from a dairy housing and wastewater treatment plant by applying a simple correction to account for the deposition loss and compares the results to other methods.

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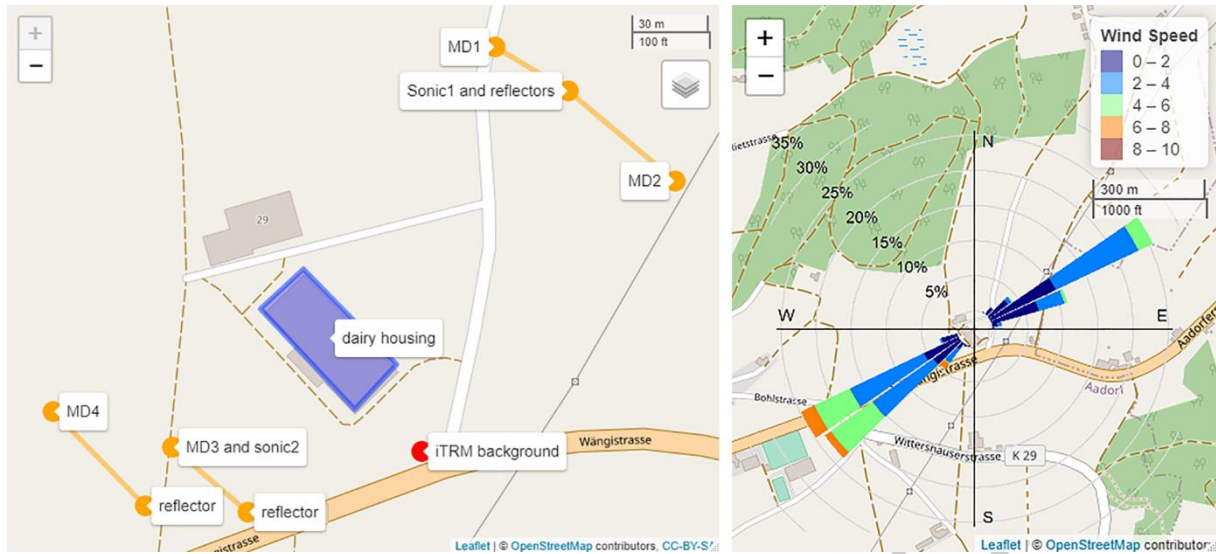
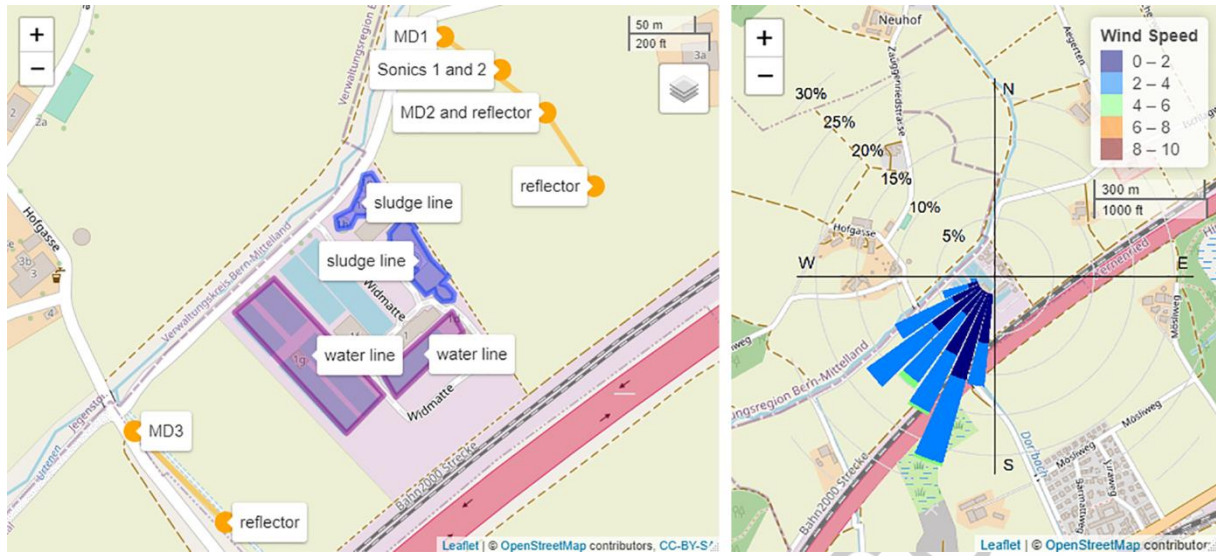


Fig 1

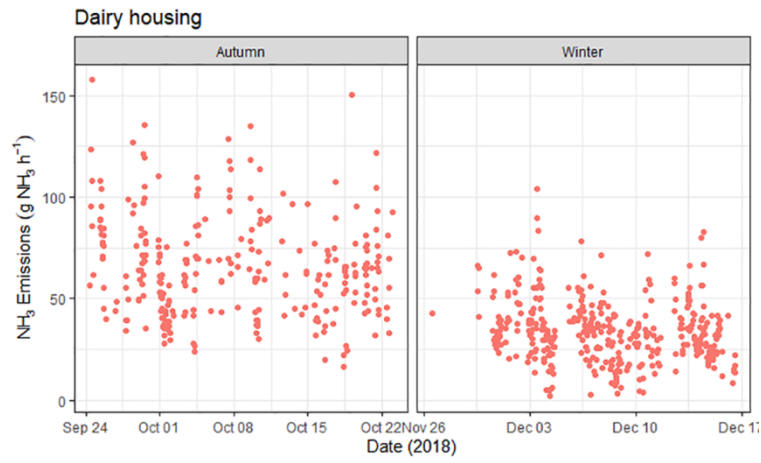
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Fig 2



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Fig 3



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Fig 4

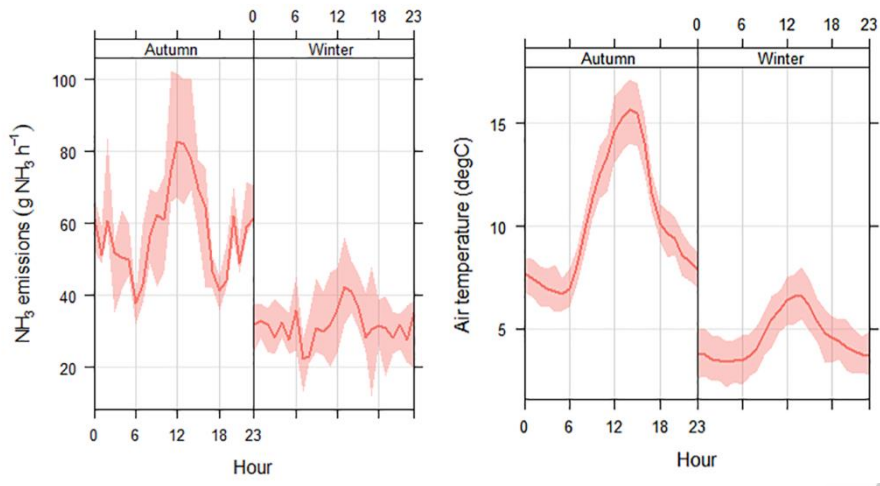
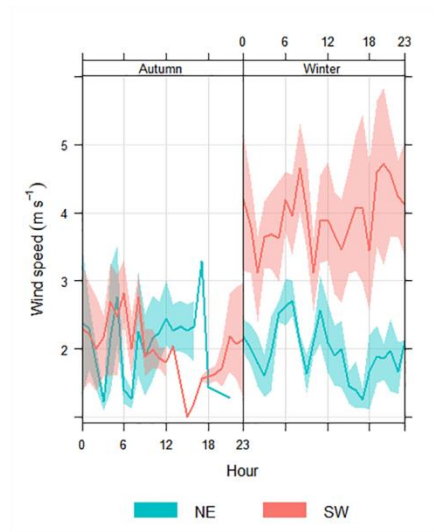
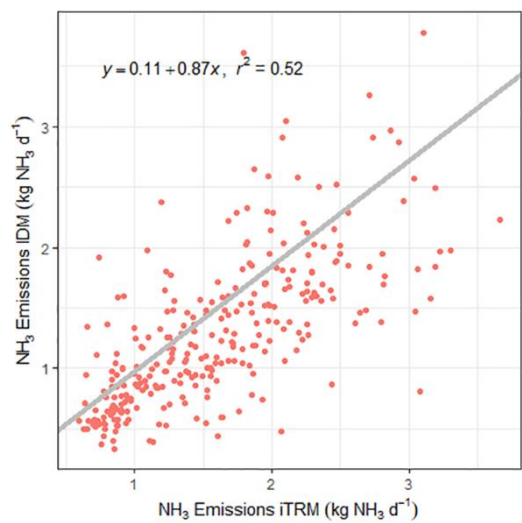


Fig 5



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Fig 6



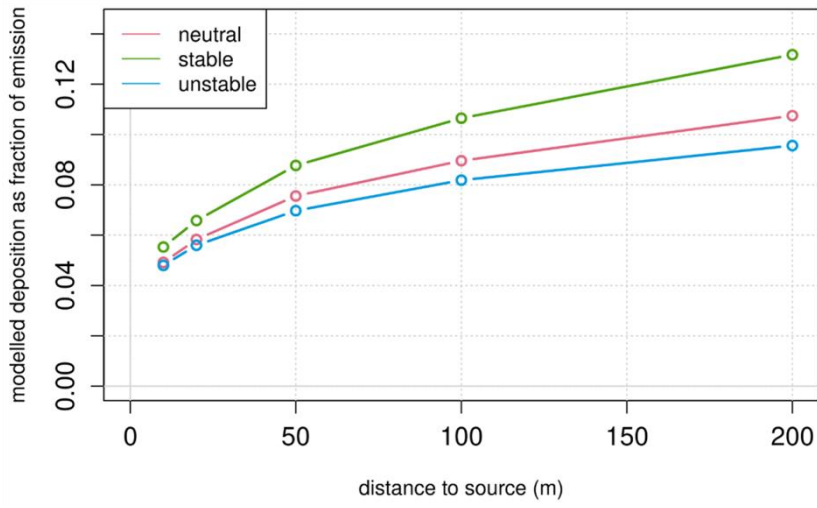
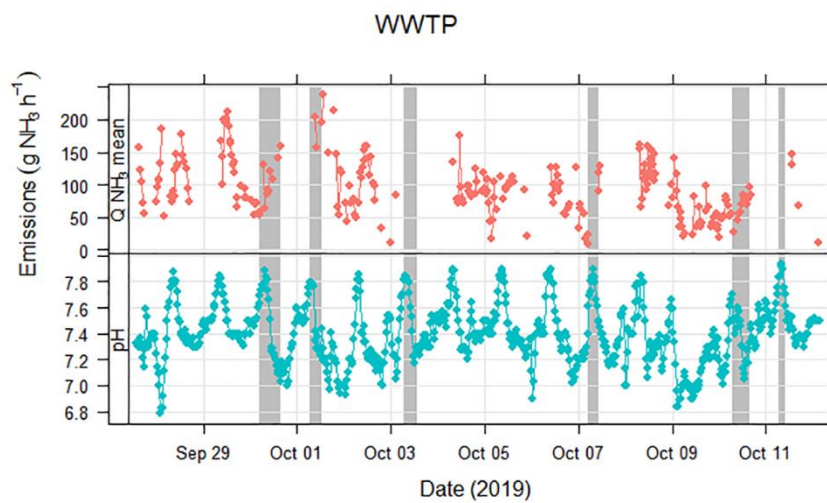


Fig 7

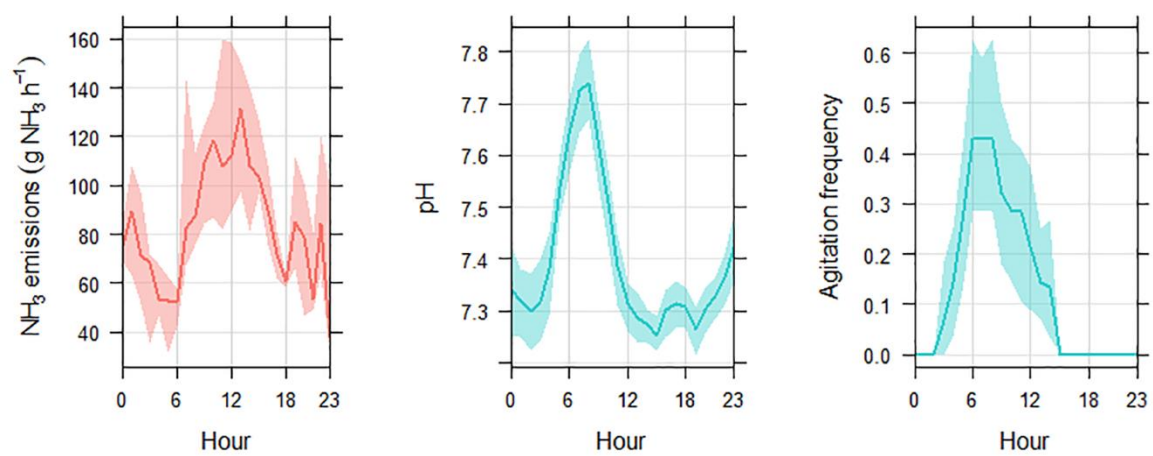
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Fig 8



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Fig 9



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Supplement: Ammonia emissions from a dairy housing and wastewater treatment plant quantified with an inverse dispersion method accounting for deposition loss

Alex C. Valach, Christoph Häni, Marcel Bühler, Joachim Mohn, Sabine Schrade, and Thomas Kupper

1. SI1. Dairy housing

SI1.1 Supplementary data

Additional figures on the weather conditions, site operations, data processing, and instrument outputs are presented below for both sites.

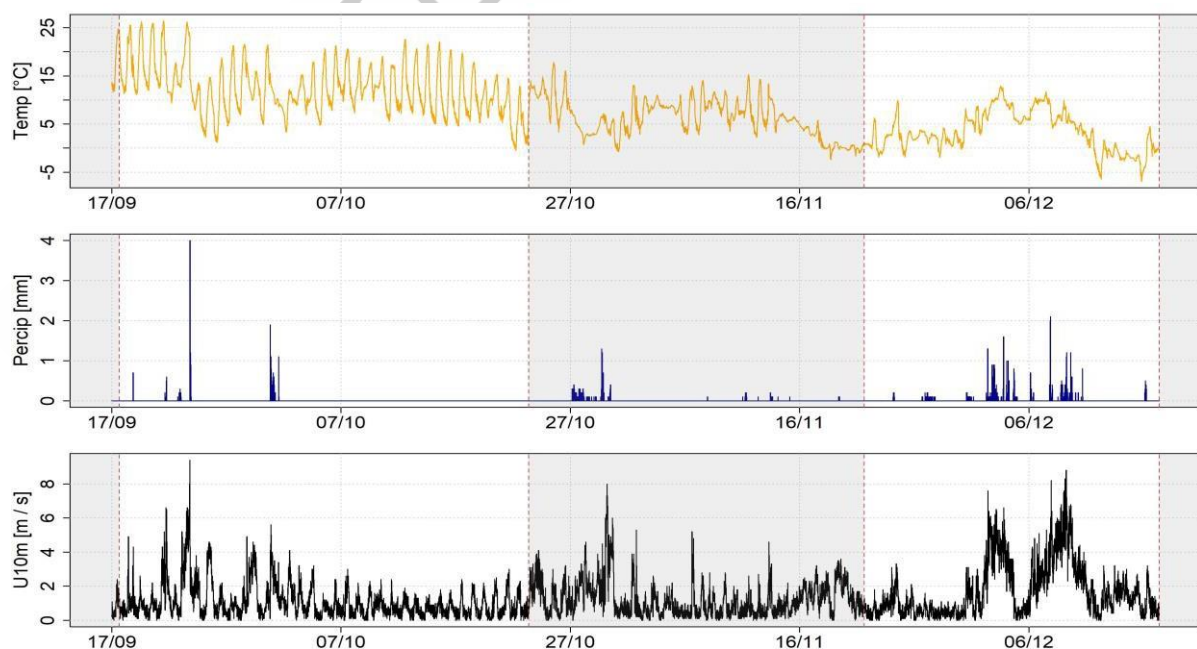


Figure SI1.1. Meteorological conditions (temperature, precipitation, and wind speed) from the nearby weather station (Taenikon in Aadorf, MeteoSwiss) during the first (autumn) and second

(winter) measurement periods (non-shaded sectors) at the dairy housing in 2018. Reproduced from the supplemental information in Bühler et al. (2022).

Due to the filtering requirements (including valid wind directions, atmospheric turbulence and stability conditions, and bLS outputs) data retention was relatively low with a bias towards higher retention during the day than at night averaging in 69% data loss across all measurements (Figure S11.2).

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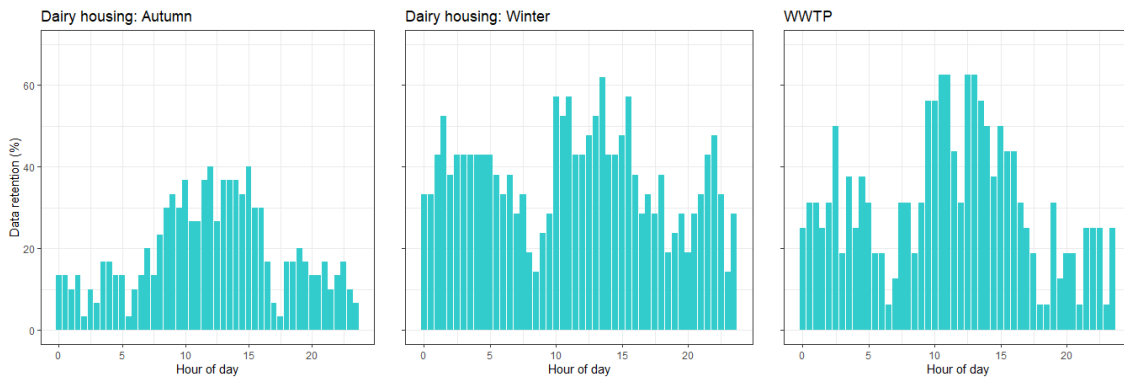


Figure S11.2. Diel averages of data retention (as % of data available during instrument uptime) for both measurement periods at the dairy housing (left and middle panels) and the WWTP (right panel).

Since emissions could only be determined for wind directions perpendicular to the line-integrated measurements Figure S11.3 indicates the dominant wind directions with time. In order to calculate the concentration increase due to the emission source, the instruments were assigned the up- or downwind position according to the dominant wind direction for each 30-min period.

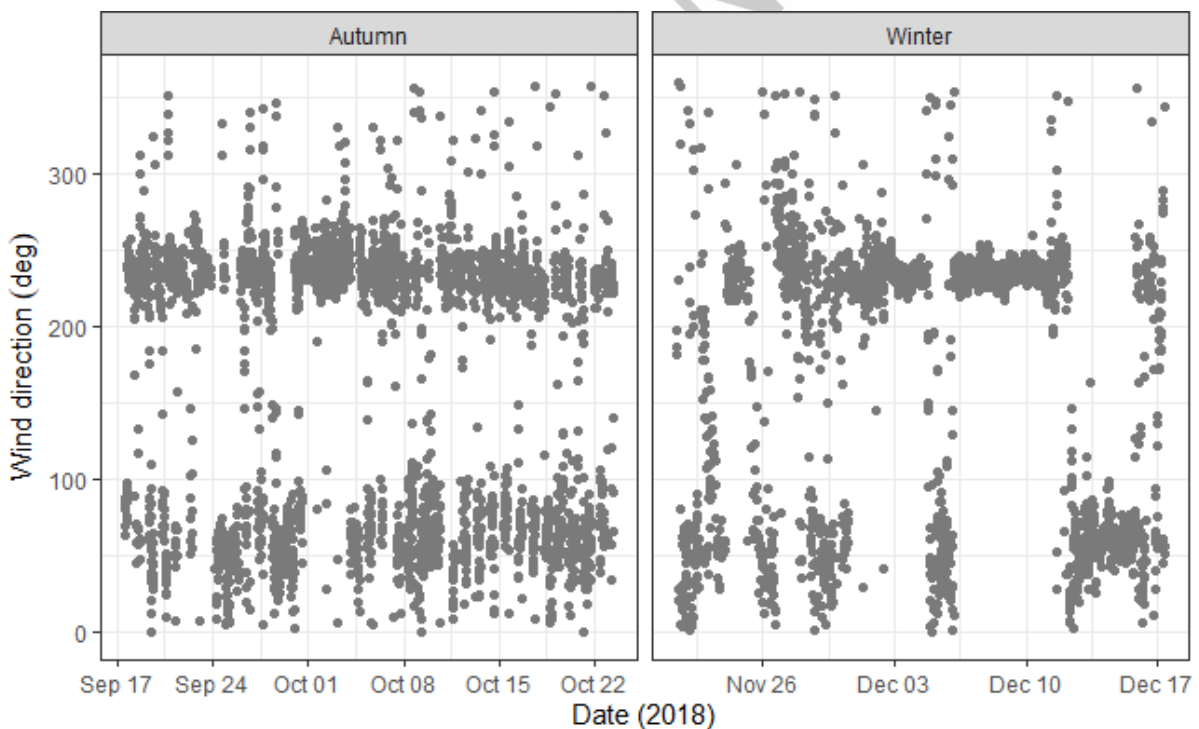


Figure S11.3. Wind directions for each 30-min averaging period at the dairy housing during both measurement campaigns.

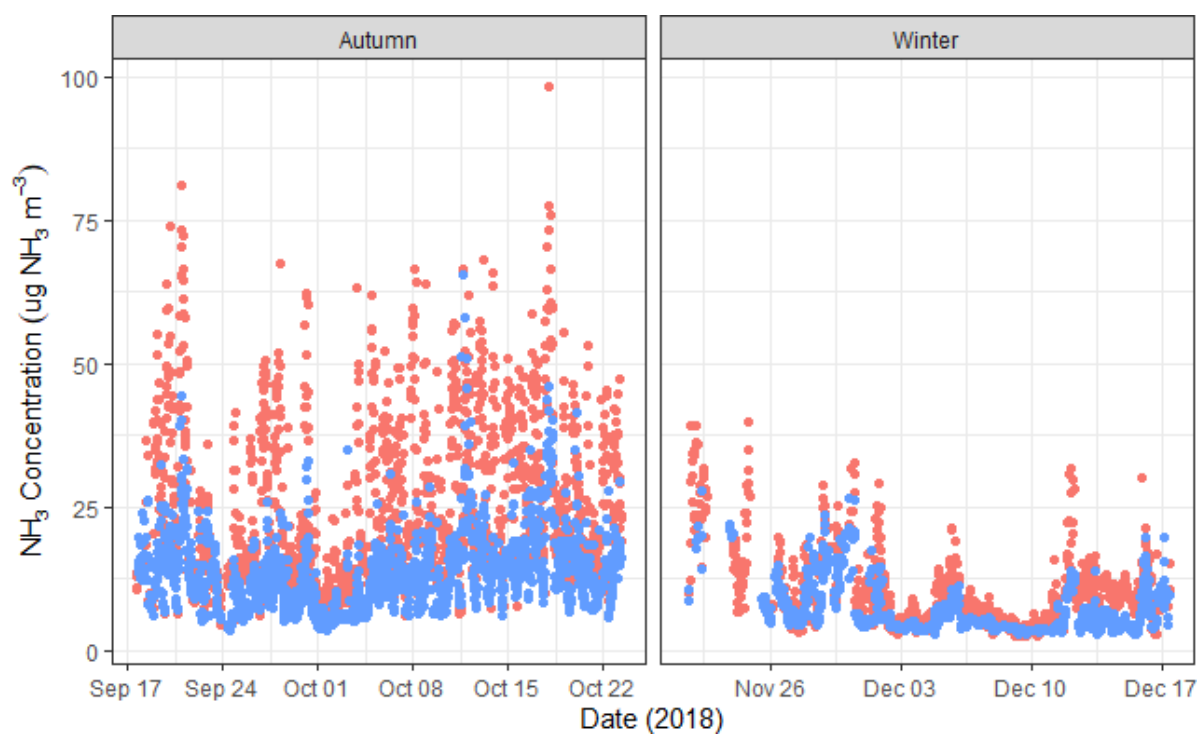


Figure S11.4. Raw (unfiltered by micrometeorological conditions) 30-min NH_3 concentration data of the upwind background (blue) and downwind plume (red) levels.

2. S11.2 Correlation statistics

Correlations of NH_3 emissions with environmental conditions by measurement period and wind direction are summarized in Table S11.1.

Table S11.1 Correlation statistics of NH_3 emissions with environmental variables at the dairy housing.

Time offset (h)	Measurement period	Wind direction	Temperature		Wind speed	
			coefficient r	p - value	coefficient r	p - value
0	Autumn	SW	0.07	0.62	0.005	0.96
		NE	0.14	0.18	0.33	<0.001***
		all	0.27	<0.001***	0.27	<0.001***
	Winter	SW	0.16	0.06	-0.01	0.83
		NE	0.42	<0.01**	0.13	0.18
		all	0.06	0.37	-0.03	0.62
	all	all	0.51	<0.001***	-0.18	<0.001***

1-2	<i>Autumn</i>	SW	0.28	<0.01**	0.08	0.4
		NE	0.24	<0.01**	0.37	<0.001***
		all	0.33	<0.001***	0.30	<0.001***
	<i>Winter</i>	SW	0.31	<0.001***	-0.06	0.35
		NE	0.26	<0.01**	-0.03	0.70
		all	0.21	<0.001***	-0.06	0.25
	<i>all</i>	all	0.55	<0.001***	-0.20	<0.001***

Statistically significant correlations are marked with * for $p < 0.05$, ** for $p < 0.01$, and *** for $p < 0.001$.

3. SI1.3 Wind direction dependencies of emissions

The winds were more dominant from the NE during autumn and from the SW during winter (Figure SI1.5).



Figure SI1.5. Wind roses superimposed on the site map showing the wind speed frequency by wind direction for the autumn (left) and winter (right) measurement periods at the dairy housing. Map data © OpenStreetMap contributors.

Daily profiles of NH_3 emissions and correlations with friction velocity u^* are shown in Figure SI1.6 with approx. 20% higher emissions with NE winds during autumn, which also showed slightly higher correlations.

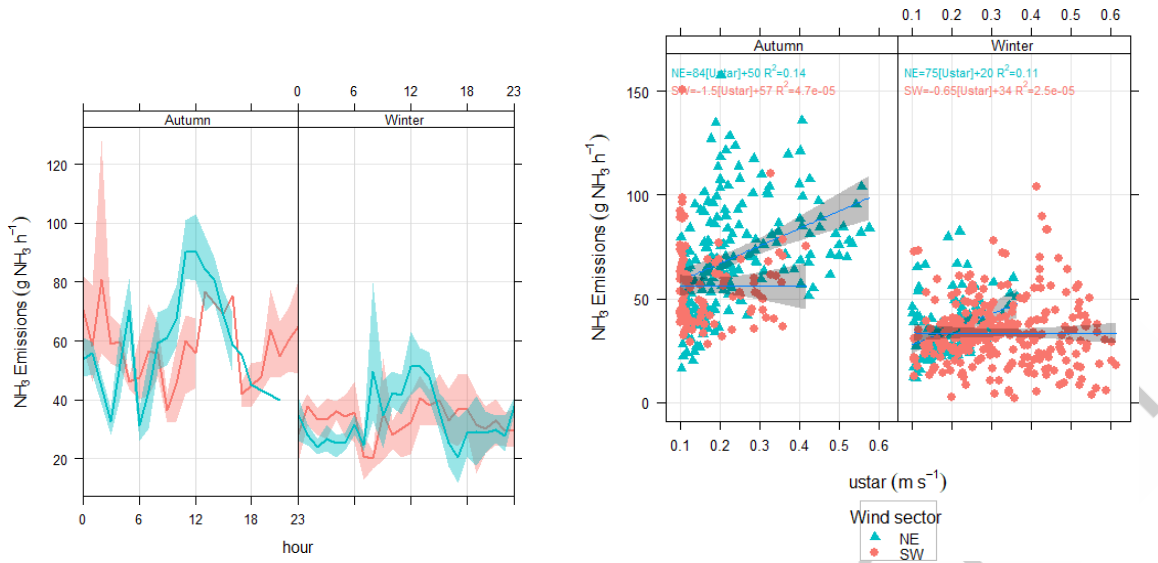


Figure S11.6 Mean diel NH_3 emissions (a, left) separated by sector (red SW, blue NE) and correlated with friction velocity u^* (b, right) with linear regressions and coefficients for each wind direction and measurement period at the dairy housing.

4. S11.4 iTRM measurements

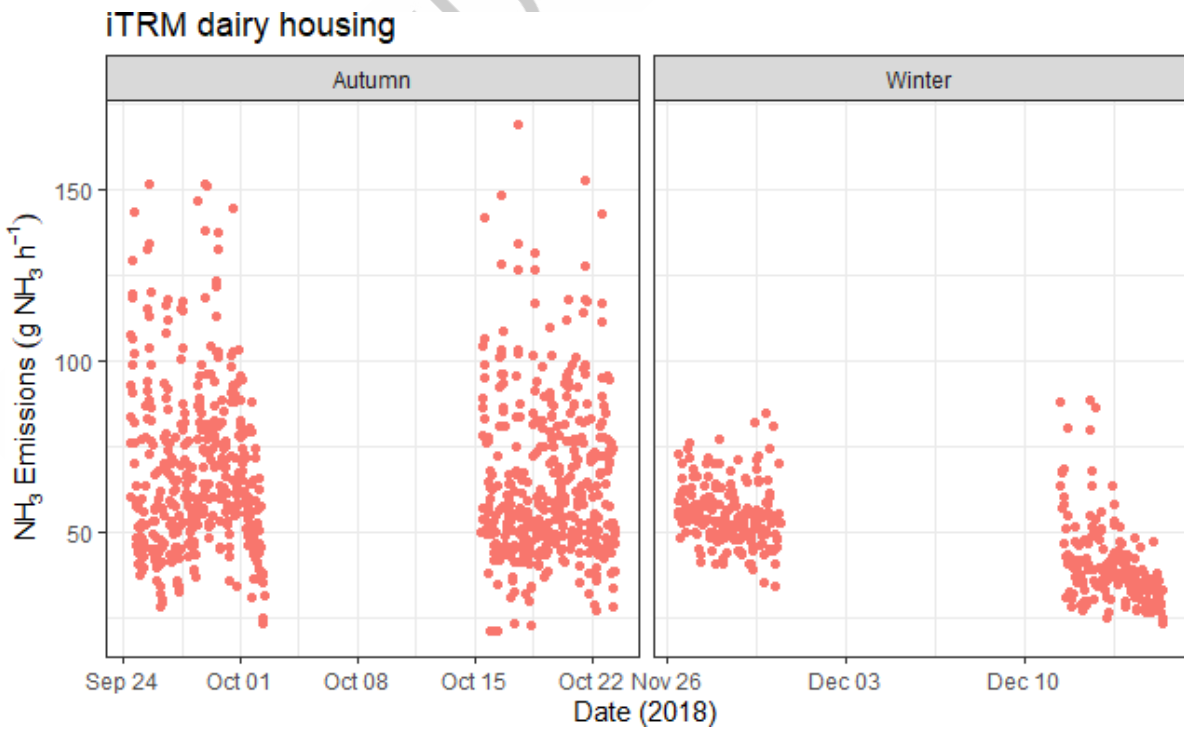


Figure SI1.7. 30-min averaged NH_3 emissions ($\text{g NH}_3 \text{ h}^{-1}$) from the inhouse tracer ratio method (iTRM) at the dairy housing during the autumn and winter measurement periods.

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5. SI2. Wastewater treatment plant

SI2.1 Supplementary data

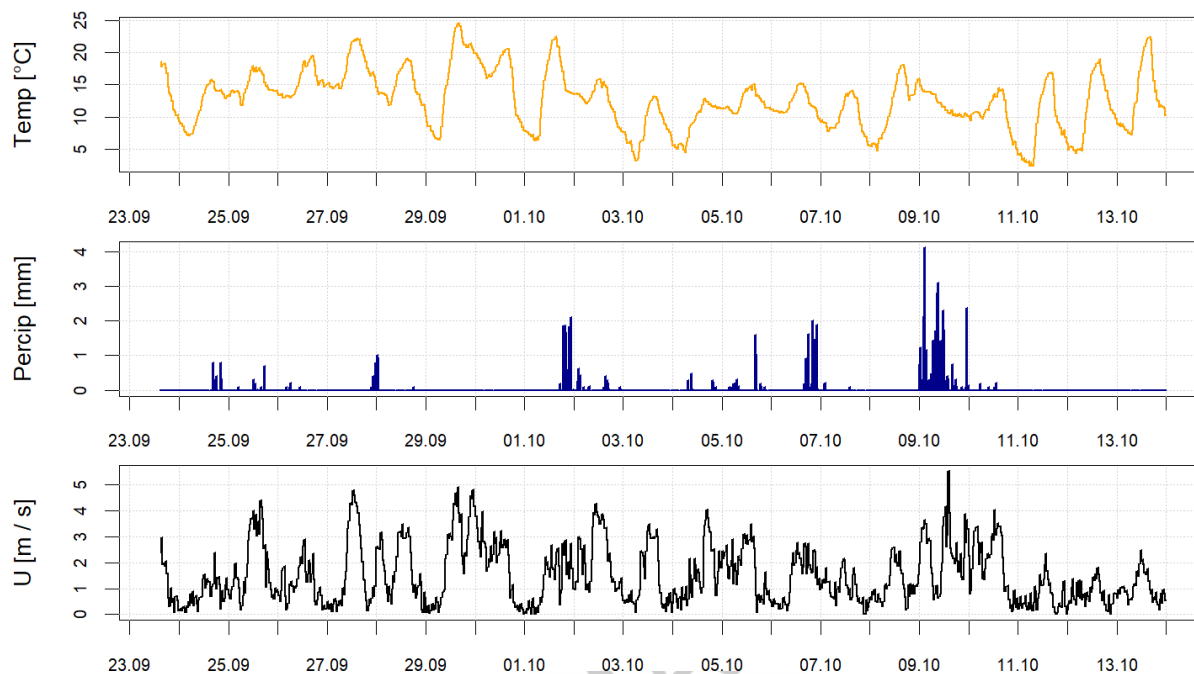


Figure SI2.1. Meteorological conditions (temperature, precipitation, and wind speed) recorded on site during the measurements at the WWTP. Reproduced from the supplemental information in Bühler et al. (2022).

Operational parameters and conditions at the WWTP during the measurement period are shown in Table SI2.1 and bLS weightings for each emitting structure at the WWTP are given in Table SI2.2. The WWTP had a total digester volume of 2200 m³, and 1960 m³ for the sludge storage tanks (surface of 331 m²), of which 632 m³ were in use during the measurement period. Measured concentrations by the up- and downwind miniDOAS instruments, as well as the wind directions at the WWTP are shown in Figures SI2.2 and 2.3.

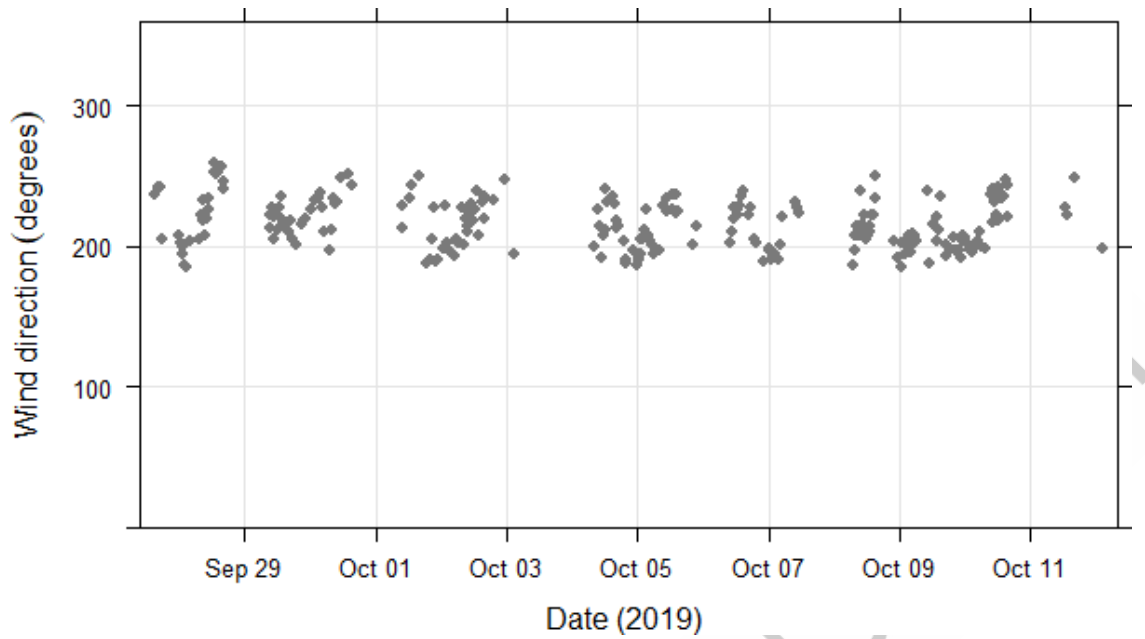


Figure SI2.2 Wind directions for the instrument placements up- and downwind of the WWTP after filtering for valid measurement conditions.

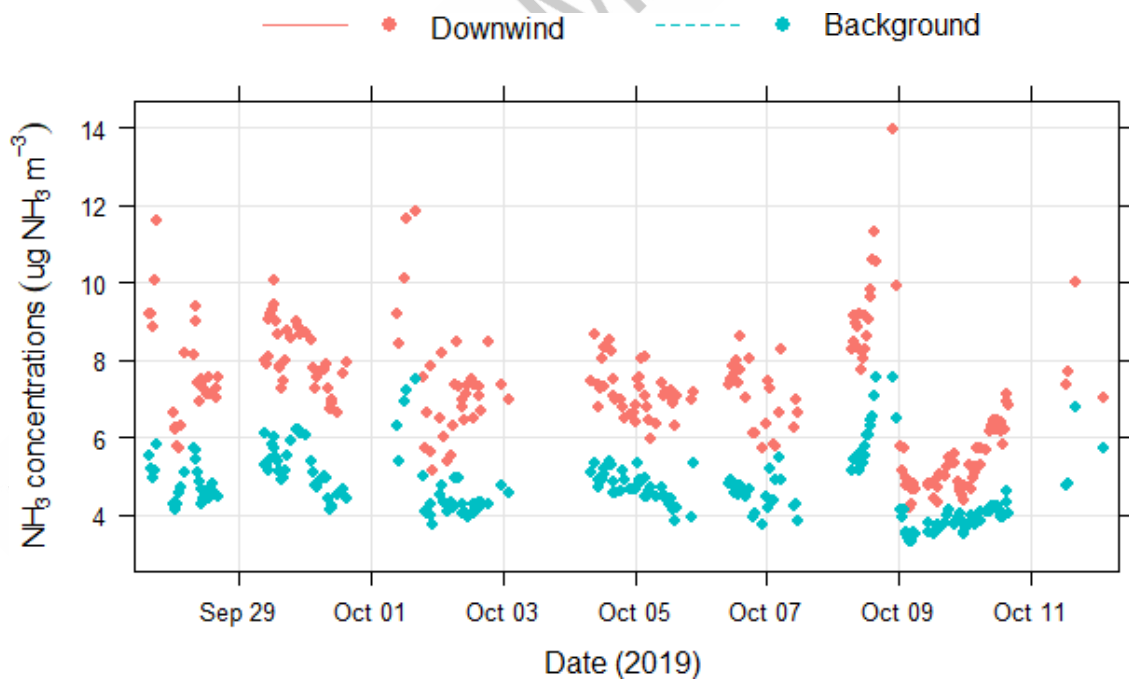


Figure SI2.3 Filtered ammonia concentrations for valid conditions measured by the miniDOAS instruments placed up- (blue) and downwind (red) of the emission source at the WWTP.

6. SI2.2 Operational data and statistics

Table SI2.1 Mean operational conditions at the wastewater treatment plant during the measurement period.

<i>Parameter</i>	<i>Units</i>	<i>Mean</i>	<i>Median</i>	<i>SD</i>	<i>Min</i>	<i>Max</i>
Inflow pH		7.39	7.39	0.23	6.79	7.93
Temperature	°C	17.7	18.0	1.3	15.3	20.
Dry matter (fresh sludge)	%	5.46	5.34	0.76	3.52	6.72
Inflow rate	L s ⁻¹	141	118	54	91	314
Inflow volume	m ³ d ⁻¹	11168	9380	6497	7829	30660
NH ₄ -N (inflow)	mg l ⁻¹	33.1	36.2	8.67	15.7	42.3
NH ₄ -N (outflow)	mg l ⁻¹	0.09	0.05	0.14	0.02	0.52
Total NH ₄ -N flow	kg d ⁻¹	336	341	233	230	1034
COD* concentration	g l ⁻¹	299	310	42	156	424
COD* Flow	kg d ⁻¹	2862	2933	1090	1866	6133

*COD: Chemical Oxygen Demand

Table SI2.2 Weightings applied to the emission of the different source structures at the wastewater treatment plant.

<i>Structure</i>	<i>Weighting*</i>	<i>Reference</i>
Storage tanks	0.81	Kupper et al., 2020
Sand trap	0.1	Samuelsson et al., 2018
Digester	0.4	Samuelsson et al., 2018
Primary clarifier	0.04	Samuelsson et al., 2018
Secondary clarifier	0.004	Samuelsson et al., 2018
Aeration tanks	0.006	Samuelsson et al., 2018

*Weightings are calculated as fractions from emission factors for each source area, which were based on values from the literature.

Table SI2.3 summarises correlations of NH₃ emissions with different driver variables using a synchronous correlation, as well as lagged correlations, whereby a positive lag indicates that the predictor variable lags behind the NH₃ emissions, while a negative lag shows that it precedes changes in emissions. The lagged correlations are only presented if they were higher than the synchronous correlation and only the lag with the maximum offset correlation is shown.

Table SI2.3 Correlations of predictor variables for NH₃ emissions at the WWTP.

<i>Parameter</i>	<i>Time offset (h)</i>	<i>Correlation coefficient</i>	<i>p-value</i>
Air temperature	0	0.48	<0.001***
	+3	0.75	<0.001***
pH	0	0.15	0.022*
	-4	0.53	<0.001***
Sludge tank agitation	0	-0.11	0.088
	-4	-0.19	0.002**

	+4	0.33	<0.001***
Incoming solar radiation	0	0.68	<0.001***

Statistically significant correlations are marked with * for $p < 0.05$, ** for $p < 0.01$, and *** for $p < 0.001$.

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