
Theobald, Mark R.; Sanz-Cobena, Alberto; Vallejo, Antonio; Sutton, Mark A..
2015. **Suitability and uncertainty of two models for the simulation of ammonia dispersion from a pig farm located in an area with frequent calm conditions.**

Suitability and uncertainty of two models for the simulation of ammonia dispersion from a pig farm located in an area with frequent calm conditions

Mark R. Theobald[a], Alberto Sanz-Cobena[a], Antonio Vallejo[a] and Mark A. Sutton[b]

[a]*Department of Agricultural Chemistry and Analysis, Higher Technical School of Agricultural Engineering, Technical University of Madrid, Spain*
[b]*Centre for Ecology & Hydrology, Edinburgh Research Station, Penicuik, United Kingdom*

Corresponding author:
Mark R. Theobald
Dpto. Química y Análisis Agrícola
E.T.S.I. Agrónomos
Universidad Politécnica de Madrid
Ciudad Universitaria s/n
28040 Madrid
Spain

Tel: +34 91 336 3694
Fax: +34 91 336 5639
Email: mrtheo@ceh.ac.uk

ABSTRACT

We used two atmospheric dispersion models (ADMS and AERMOD) to simulate the short-range dispersion of ammonia emitted by two pig farms to assess their suitability in situations with frequent calm meteorological conditions. Simulations were carried out both using constant and temporally-varying emission rates to evaluate the effect on the model predictions. Monthly and annual mean concentrations predicted by the models at locations within one kilometre of the farms were compared with measured values. AERMOD predicted higher concentrations than ADMS (by a factor of 6-7, on average) and predicted the atmospheric concentrations more accurately for both the monthly and annual simulations. The differences between the concentrations predicted by the two models were mainly the result of different calm wind speed thresholds used by the models. The use of temporally-varying emission rates improved the performance of both models for the monthly and annual simulations with respect to the constant emission simulations. A Monte Carlo uncertainty analysis based on the inputs judged to be the most uncertain for the selected case study estimated a prediction uncertainty of ± a factor of two for both models with most of this due to uncertainty in emission rates.

Keywords: *Ammonia emissions; Atmospheric dispersion modelling; Uncertainty analysis*

1. INTRODUCTION

Ammonia (NH_3) emitted into the atmosphere from agricultural sources can have an impact on nearby sensitive ecosystems either through elevated ambient concentrations or dry/wet deposition to vegetation and soil surfaces (Bobbink et al., 1998). Evidence of impacts of elevated NH_3 concentrations on vegetation has made it possible to define ‘critical levels’ for NH_3 exposure. An annual critical level of 3 µg m^{-3} (with an uncertainty range of 2-4 µg m^{-3}) has been recommended for ecosystems containing higher plants only and a lower critical level of 1 µg m^{-3} for ecosystems containing lichens or bryophytes (Cape et al., 2009). Based on current evidence, impacts to these ecosystems may occur when the annual mean NH_3 concentration is above the critical level. Similarly, impact thresholds of long-term (e.g. 20-30 years) nitrogen deposition rates (critical loads) have also been developed for different ecosystem types (Achermann and Bobbink, 2003).

In Europe, where NH_3 is a regulated pollutant, potential impacts of agricultural emissions to nearby sensitive habitats are normally assessed using atmospheric dispersion models. Model predictions of annual mean atmospheric NH_3 concentrations and nitrogen deposition rates are used to determine whether the critical levels and critical loads, respectively, of nearby sensitive habitats are likely to be exceeded. Since dry deposition rates are calculated by the models from ground-level concentrations using empirically-derived and uncertain deposition parameterisations, the deposition predictions are likely to be more uncertain than the concentration predictions (Environment Agency, 2010). For this reason, it may be preferable to base an environmental impact assessment on critical levels instead of critical loads. A range of different models are used for these assessments, with the choice of model depending on local expertise and model development programmes (Theobald *et al*., 2012). For example, in the United Kingdom, assessments are usually carried out using one of two advanced Gaussian dispersion models (Environment Agency, 2010): the Atmospheric Dispersion Modelling System (ADMS, Carruthers *et al*., 1994) or the AMS/EPA Regulatory Model (AERMOD, Cimorelli *et al*., 2002). These two models have been evaluated for a range of applications, including some agricultural case studies (Hill *et al*., 2001; Theobald *et al*., 2012) and, in general, perform acceptably when all model inputs (emissions rates, meteorological variables etc.) are known with sufficient accuracy.

However, for environmental impact assessments, assumptions and approximations have to be made when model inputs are not known accurately. For example, for assessments of environmental impacts of livestock facilities, emission rates are often assumed to be constant and based on national or international emission factors for each livestock type. Furthermore, meteorological data are normally taken from the nearest ‘representative’ meteorological station, which can be many kilometres from the assessment site. In addition, it may be difficult to obtain complete meteorological records due to sensor downtime or calm periods. Advanced Gaussian dispersion models such as AERMOD and ADMS include routines to simulate periods with low wind speeds. AERMOD, for example, uses a combined solution of a coherent plume (traditional Gaussian shape) and a radially-symmetric plume to simulate dispersion for low wind speeds. The model interpolates between these two plumes, tending to the radially-symmetric plume at very low wind speeds (US EPA, 2003). A similar approach is also used in ADMS, when the non-default calms option is selected (CERC, 2007). However, the default versions of the models cannot simulate ‘calm’ periods when the wind speed in the meteorological data record is zero and so these periods are removed from the model calculations. These are periods when the actual wind speed is less than the anemometer stalling speed but not necessarily zero. This is problematic because high concentrations may occur during these periods as a result of low dispersion rates. This problem is more commonly encountered when routine meteorological data from network stations are used (often the case for impact assessments), which tend to use cup anemometers, compared with meteorological data from research-grade model evaluation studies that use more advanced measurement techniques (e.g. ultrasonic anemometers). AERMOD identifies a calm period when the wind speed is below a user-defined threshold based on anemometer stalling speeds whereas ADMS has a default wind speed threshold of 0.75 m s^{-1} at a height of 10 m when the calms option is not selected.

UK modelling guidance (Defra, 2009) recommends that models can be used for predicting annual mean concentrations when valid non-calm meteorological data are available for more than 75% of the modelling period (provided that there are no gaps of several weeks). However, it may not be possible to meet this criterion in locations with frequent calm periods and so there is a need to evaluate model performance when this criterion cannot be met.

As mentioned above, one of the assumptions frequently made in these assessments is that the emission rates are constant. However, emission rates of agricultural sources are not constant since they depend on many factors as a result of management practices and environmental conditions. The assumption of constant emissions may, therefore, affect the annual mean concentrations predicted by the models, although this has not been tested.

In this paper we simulate the atmospheric dispersion of NH_3 emitted by a Spanish pig farm with two advanced Gaussian dispersion models: ADMS and AERMOD. This case study was chosen because the pig farm is located in a region with frequent calm winds and so is a good candidate to test the suitability of these two models for these meteorological conditions. This is done by statistically comparing monthly and annual mean NH_3 concentrations predicted by the models with those measured at multiple locations up to one km from the farm. Many of the model inputs are uncertain (emission rates, exit velocities, meteorological variables, etc.), as in many real impact assessments and so an uncertainty analysis has been done to assess the influence of these uncertainties on the models' predictions. We also test an emission model that better represents the temporal variability of the pig farm emissions. The objectives of this study were, therefore:

1. To assess the suitability of the two dispersion models ADMS and AERMOD for an agricultural case study with frequent low-wind conditions;
2. To quantify the uncertainty of model predictions due to uncertainties in input data;
3. To assess the effect on the concentration predictions of using an emissions model to simulate the hourly variability of emissions.

2. MATERIALS AND METHODS

2.1. Experimental site

A one year field experiment was carried out in the vicinity of a pig farm (Farm 1 in Figure 1) in the region of Segovia, central Spain. The farm is a pig breeding unit with a fairly constant number of sows (565 animals, on average) with piglets up to 20 kg (1092 animals). The unit consists of three main buildings: the adaptation building for new sows (105 animals) (A in Figure 1), the gestation building (370 animals) (B) and the birthing building (90 sows and 1092 piglets) (C). The buildings have fully-slatted floors with slurry pumped frequently to an outdoor lagoon (D). Slurry is removed periodically for application to nearby arable fields, although no information is available on where and when the slurry is applied. All buildings of the farm are mechanically ventilated with wall inlets. The adaptation building has four wall ventilation outlets, whilst the gestation and birthing buildings have roof outlets (Figure 1). Approximately 1.2 km NW of the unit is another similar unit (Farm 2) with an average of 240 breeding sows in three buildings and an outdoor slurry lagoon. All buildings of this farm have roof ventilation outlets. The area is very flat and the land use around the two farms is arable fields (cereals and sunflowers) with some set-aside and woodland. Detailed management data for the arable fields (e.g. crops grown and fertiliser applications) are not available. There is also a small infrequently used road that passes through the experimental area.

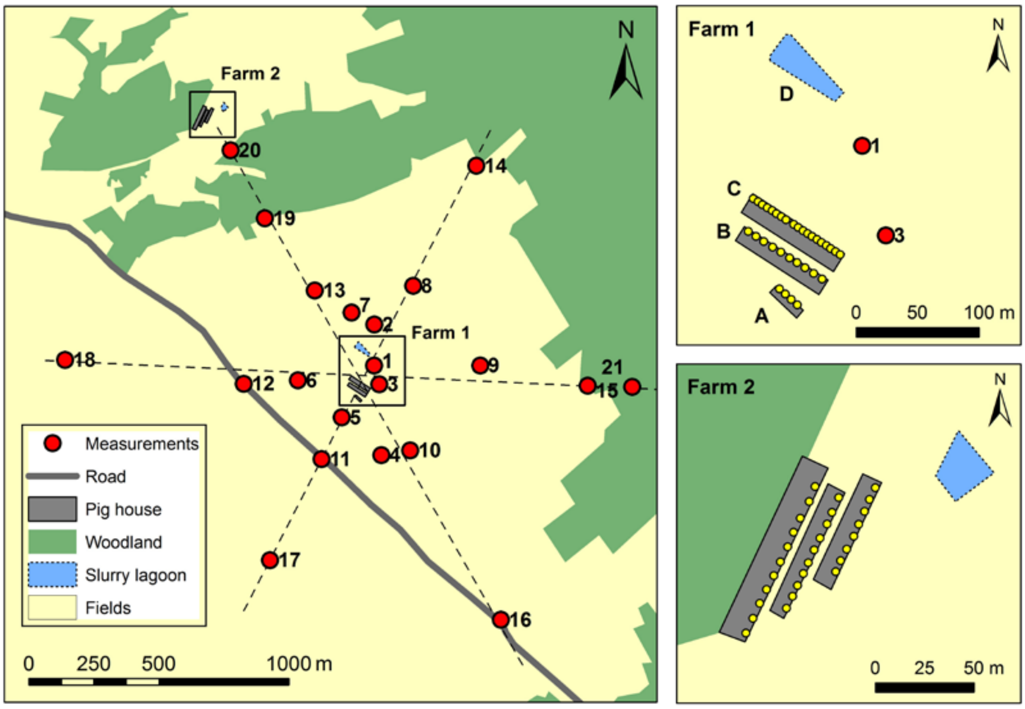

Figure 1: Plan showing land cover, the location of the two farms and the measurement locations within the experimental area. The dotted lines indicate the six radial directions used for the measurements. The insets on the right show the locations of the buildings, slurry lagoons and ventilation points (small circles) of the two farms.

2.2. Ammonia emission estimates

Ammonia emission data are not available for Farm 1 or Farm 2 and, therefore, emission estimates were taken from the emission inventory guidebook produced jointly by the European Monitoring and Evaluation Programme (EMEP) and the European Environment Agency (EEA) (EEA, 2009). The emissions were calculated using the Tier 2 mass flow approach using standard values of nitrogen excretion and emission factors for each stage of the manure management process. The housing emission calculated this way was 6.45 kg NH_3 per sow (including piglets) per year and the slurry lagoon emission was 3.38 kg NH_3 per sow per year.

The use of constant annual emission factors is not ideal because temporal variations will occur due to changes in ambient conditions, especially for the slurry lagoons, which are exposed to large diurnal and seasonal changes in meteorological conditions. In order to take these variations into account, an emissions model was used (Gyldenkærne *et al*., 2005). The model distributes the annual emissions of each source using the ambient air temperature and either the wind speed or ventilation rate, for slurry lagoons and animal houses, respectively, whilst maintaining the annual emission factor. A description of the emission model is included as Supplementary Material.

2.3. Ammonia concentration measurements

An array of 21 passive samplers (Figure 1) was used to measure monthly mean atmospheric NH_3 concentrations over the period mid May 2008-mid May 2009. At each location, triplicate passive (ALPHA) samplers (Tang *et al*. 2001) were exposed beneath a white plastic rain shelter at a height of 1.5 m above ground for 12 consecutive periods of approximately one month (sample periods of 26-35 days). These samplers work on the principal of diffusion of atmospheric NH_3 over a short distance (7 mm) down a short plastic tube to an absorbing medium (acid-coated filter) during the exposure period. Turbulent transport of NH_3 into the sampler is minimized through the use of a PTFE membrane at the sampler entrance. Sampler preparation was according to Tang *et al*. (2001) except phosphoric acid was used as the filter-coating medium instead of citric acid due to its better stability at high air temperatures. Following exposure in the field, the acid-coated filters were each extracted in 3 ml of deionised water and 2 ml of the extract was analysed for ammonium using the indophenol blue method (Searle, 1984) and spectrophotometry. Samples below the detection limit (defined as 3 standard deviations of the laboratory blank samples) and sample outliers were removed from all subsequent analyses. Background concentrations for each sampling period were estimated to be the concentration measured at the site with the lowest concentration during the sampling period. The background concentration was then subtracted from the concentrations measured at the other sampling locations for each period.

2.4. Meteorological data

Measurements of wind speed and wind direction (at heights of 0.51, 1.17 and 2.74 m above ground) and air temperature (0.51 and 2.74 m) were made on a mast located 80 m NE of the birthing building of Farm 1 (Site 1 in Figure 1). This location was probably not out of the region of influence of the farm buildings, since building wakes can extend up to 10-20 building heights downwind (Arya, 2001), whereas the mast was only 14 building heights from the main building, but this was the furthest secure location available. The measurements were made by three weather stations (Davis Weather Wizard III) (one for each measurement height) and hourly data were stored on the internal dataloggers.

2.5. Model simulations

The atmospheric dispersion models AERMOD (version 12345) and ADMS 4.1 were used to predict the monthly and annual mean atmospheric NH_3 concentrations, resulting from the NH_3 emissions of both farms, at the 21 measurement locations. Model simulations were carried out for both the constant emission scenario and for the scenario using the time-varying emission model. More details of the model configurations are provided in the Appendix.

2.6. Uncertainty analysis

In order to evaluate the uncertainty of model predictions due to the uncertainty in the model inputs, a Monte Carlo analysis was carried out for the inputs that we judged to be the most uncertain and/or most influential. The five inputs selected were: emission rates, exit velocity, aerodynamic roughness length (z_0), cloud cover and boundary layer height. These inputs were not measured directly and so the emission rates, exit velocities and z_0 had to be estimated, cloud cover was taken from a weather station more than 30 km away and boundary layer heights were taken from the output of a numerical weather prediction model (see Appendix). We considered that these five inputs were more uncertain than directly-measured model inputs such as source heights and diameters, building dimensions, wind speed, wind direction, air temperature, etc.

Values for the selected inputs were chosen randomly following a midpoint latin hypercube design (Iman and Helton, 1988), in order to ensure the selection of values representative of the entire probability distributions (Table 1) and to reduce the number of required simulations (compared with a standard Monte Carlo design). No correlations were assumed between the selected inputs.

Table 1: Model input variables and their distributions used in the uncertainty analysis

Input	Distribution type	Most probable value	95% Confidence Interval [a]
Emission rate	Lognormal	EMEP/EEA emission factor	± factor 2 [b]
Exit velocity	Normal	Calculated from recommended building flow rates (see Appendix)	± 35%
Roughness length	Lognormal	0.05 m	± factor 3 [c]
Cloud cover	Normal	Observed value	± 40% [d]
Boundary layer height	Lognormal	WRF v3.1.1 prediction	± factor 2

[a] Hourly values of cloud cover and boundary layer height were a combination of variations in the annual mean and hourly fluctuations, both with the same probability distributions (Hanna *et al*., 2007)

[b] EEA (2009)

[c] Hanna *et al*. (2007)

[d] Twice the value used by Hanna (2007) due to the distance between measurement location and modelling domain (34 km)

In order to assess the minimum number of simulations necessary for a robust uncertainty estimate, analyses were carried out for increasing numbers of model simulations until the uncertainty range of the model predictions stabilised. Stabilisation in this case was defined as a change of less than 10% in the annual mean concentrations and their respective standard deviations when the number of simulations was doubled. From this the optimum number of simulations was estimated to be 100.

2.7. Model performance evaluation

Model performance was evaluated from the measured and predicted monthly and annual mean NH_3 concentrations using the acceptability criteria of Chang and Hanna (2004) shown in Table 2. Performance measure values were calculated for all model simulations and compared with the acceptability criteria. Recent work on model performance evaluation by Hanna and Chang (2012) has recognised that, due to stochastic and turbulent processes, even an acceptable model may not meet all acceptability criteria for all experiments. As a result, they propose that an acceptable model is one that meets the criteria for at least half of the performance tests. However, it must be borne in mind that these acceptability criteria were developed for assessing model performance in short-term research-grade evaluations where emission rates, source parameters and required meteorological variables are measured with sufficient accuracy. The criteria may be excessively strict for case studies such as that presented here, where many model inputs have to be estimated, although ADMS and AERMOD have been shown to meet the criteria for similar case studies for averaging times of several months (Theobald *et al*., 2012).

Table 2: Definitions of the performance measures used and their relationships to the observed (C_o) and predicted concentrations (C_p).

Performance measure	Definition	Optimum value	Acceptability Criterion
Fractional bias (FB)	$FB = \frac{2(\overline{C_o} - \overline{C_p})}{(\overline{C_o} + \overline{C_p})}$	0	$\lvert FB \rvert < 0.3$
Geometric Mean Bias (MG)	$MG = \exp\left(\overline{\ln C_o} - \overline{\ln C_p}\right)$	1	$0.7 < MG < 1.3$
Normalised mean square error (NMSE)	$NMSE = \frac{\overline{(C_o - C_p)^2}}{\overline{C_o}\,\overline{C_p}}$	0	$NMSE < 1.5$
Geometric variance (VG)	$VG = \exp\left[\overline{(\ln C_o - \ln C_p)^2}\right]$	1	$VG < 4$
Fraction of model predictions within a factor of two of the observations (FAC2)		1	$FAC2 > 0.5$

2.8. Statistical analyses

To estimate the contribution of each input used in the uncertainty analysis to the model output uncertainty an analysis of relative importance was carried out using the "relaimpo" package of R (Grömping, 2006; R Development Core Team, 2008), which partitions the value of R^2 between the input variables in linear regression models following Lindemann *et al.* (1980).

3. RESULTS

3.1. Meteorological data

Figure 2 shows the mean air temperatures and wind speeds (measured at a height of 2.74 m) for the monthly ALPHA sampler measurement periods and wind frequency rose for the entire experimental period. Mean air temperatures had a large seasonal range of 1-23 °C. Mean wind speeds on the other hand were fairly constant throughout the year with no clear seasonal trends other than increased variability during winter. Zero wind speeds occurred during the monthly measurement periods 9-42% of the period and wind speeds below 0.5 m s^{-1} (including zero) occurred 17-68% of the period. For the entire experimental period, wind speeds were zero or below 0.5 m s^{-1} for 24% and 37% of the time, respectively. Winds during the entire experimental period mainly came from the north or the south to west sector.

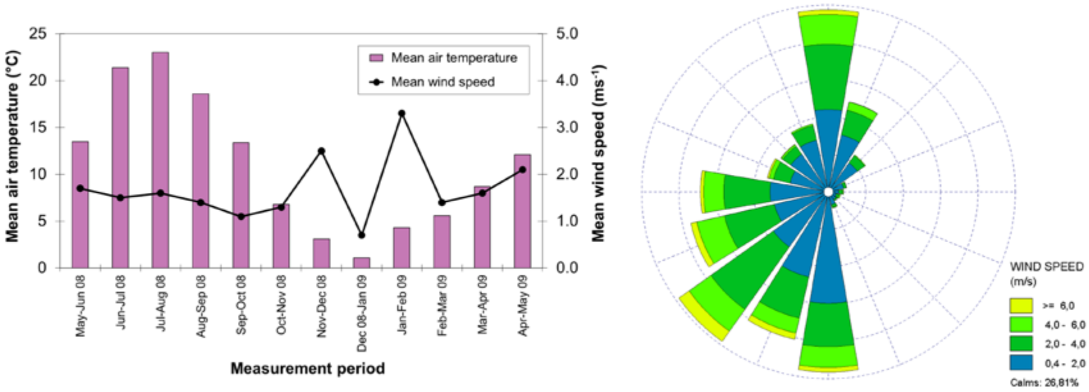

Figure 2: Mean air temperature and wind speed for each ALPHA sampler measurement period (left) and wind frequency rose for the entire experimental period (right) plotted using WRPLOT v7.0 (Lakes Environmental Software).

3.2. Ammonia concentration measurements

Figure 3 shows the annual mean atmospheric NH_3 concentrations for the 21 ALPHA sampler locations. An analysis of the monthly variability shows that at most sites concentrations peak in July-August and March-April. The summer peak is most likely due to a combination of nearby field-application of fertilisers (observed during field visits) and high in-house and/or slurry lagoon temperatures at the two pig farms. In fact the concentration at Site 1 (one of the nearest sites to Farm 1) is well-correlated with the mean air temperature (linear regression: R^2=0.83). The smaller spring peak in concentrations (March-April) is not correlated with air temperature and is probably due to slurry handling operations and the field-application of fertilisers to nearby fields.

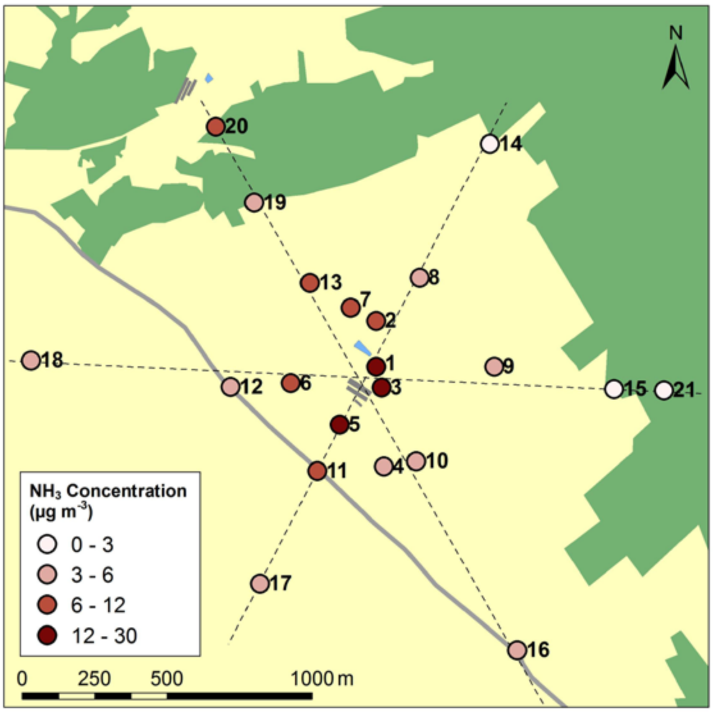

Figure 3: Annual mean atmospheric NH_3 concentrations measured by the ALPHA samplers. The numbers indicate the sample number.

3.3. Emission model estimates

The emission model predicted smaller temporal variability in emissions from the pig houses than from the slurry lagoon due to the smaller air temperature range inside the pig houses (Figure 4). The outdoor slurry lagoon emissions have a large diurnal variability because of the difference between night-time and day-time temperatures and also because of the calm night-time conditions, when the model predicts zero emissions.

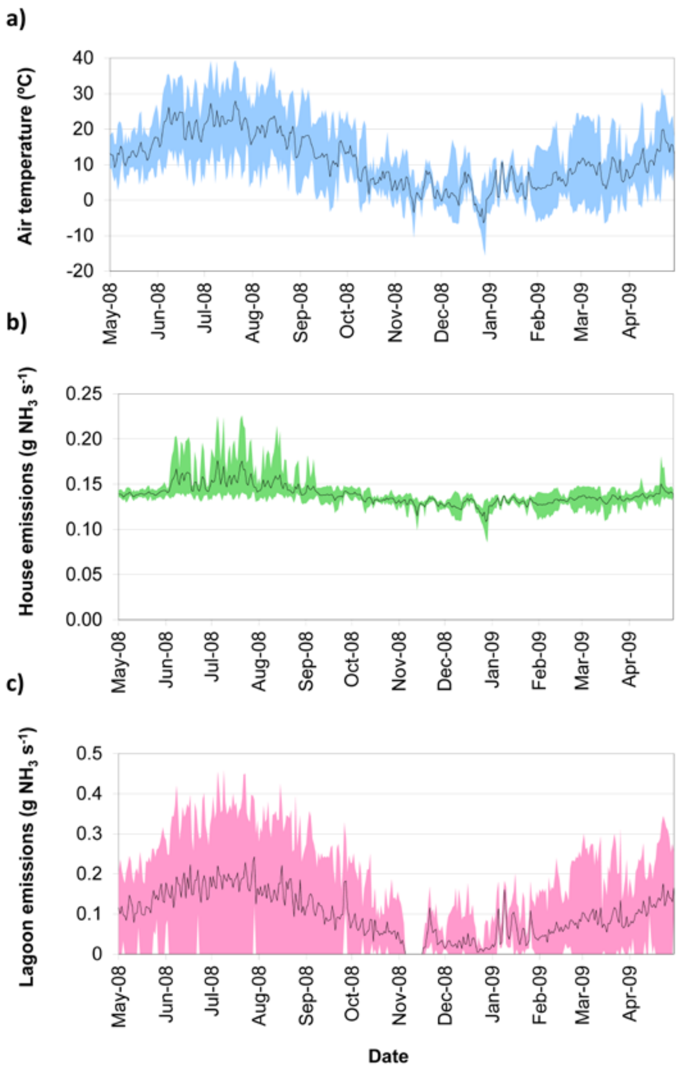

Figure 4: Daily means (solid lines) and daily range (shading) of a) measured air temperature and modelled NH_3 emissions from b) the pig houses and c) the slurry lagoon for Farm 1 calculated using the hourly emission model.

3.4. Modelled concentration predictions

Figure 5a shows the predicted versus measured monthly mean NH_3 concentrations for all sites for the constant emission scenario. The values plotted are the geometric means of the log-normally distributed concentrations of the 100 uncertainty analysis runs. In general, ADMS predicted lower concentrations than AERMOD for all sites by a factor of 1.3 to 41 (mean value: 6.8). For the analysis of annual mean concentrations, only those sites with valid measurement data for more than three quarters of the experimental period were used (sites 1-13, 19 and 20). Similarly to the monthly simulations, AERMOD predicted higher annual mean concentrations than ADMS by a factor of 2.0 to 12.4 (mean value: 5.9) (Figure 5b). The use of time-varying emissions in the simulations increased ADMS concentration predictions by 16% and 12%, on average, and decreased AERMOD concentration predictions by 20% and 28%, on average, for the monthly and annual simulations, respectively (Figure 6). The prediction uncertainty (ninety-five percent confidence intervals of the 100 uncertainty analysis runs) for both the monthly and annual simulations was approximately ± a factor of two for all sites and measurement periods.

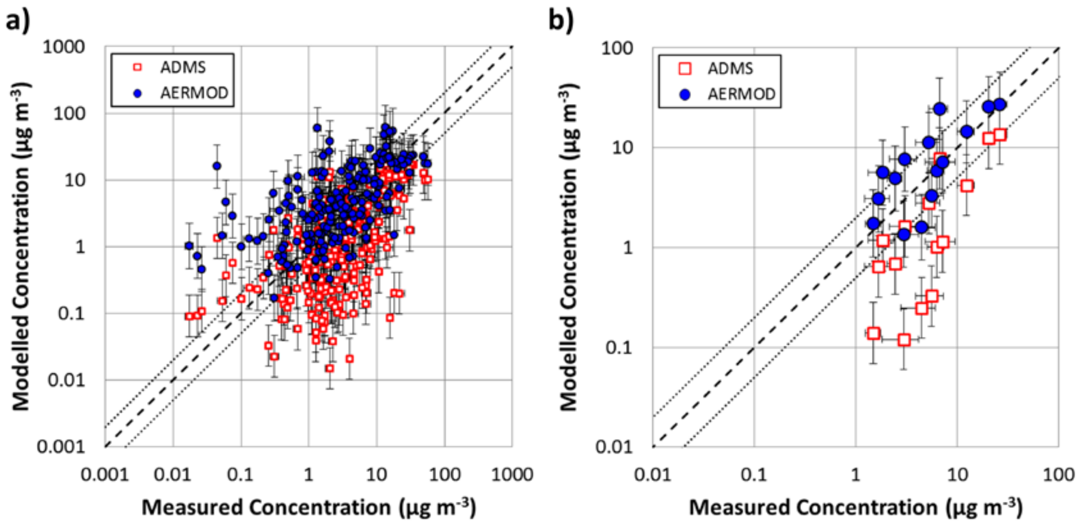

Figure 5: Modelled versus measured a) monthly and b) annual mean NH_3 concentrations for the constant emission simulations. Each point represents the mean measured and modelled value for 100 model runs. Error bars show ±2 standard deviations of predicted concentrations and ±2 standard errors of the measured values. The dashed line shows the 1:1 line and the dotted lines show the limits for predictions within a factor of two of the measured values.

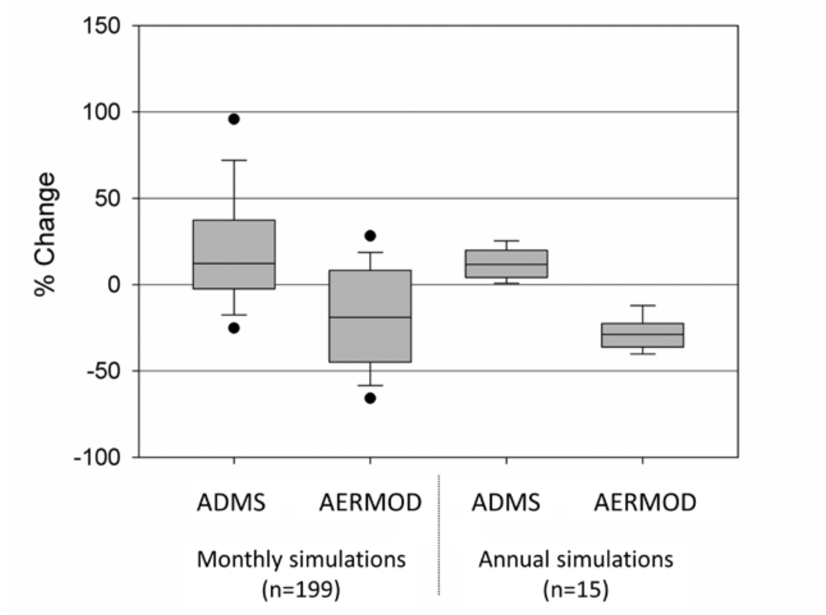

Figure 6: Box plot showing the effect of using time-varying emissions (relative to the constant emission scenario) on the monthly and annual mean predicted concentrations. The boxes show the median value and interquartile range, the whiskers are the 10th and 90th percentiles and the circles are the 5th and 95th percentiles of the distributions.

An analysis of the contribution of each selected input to the overall uncertainty of the annual model predictions shows that three of the five inputs (emission rate, z_0 and exit velocity) contributed more than 90% of the variance of the model output. Less than 10% of the uncertainty at all sites was not accounted for (e.g. due to non-linear interactions). The largest contribution to model uncertainty was the source emission rate, which contributed 77-99% of the prediction uncertainty and when averaged over all sites contributed 96% and 87% of the uncertainty for ADMS and AERMOD, respectively. These contributions were similar for both the constant and the varying emission scenarios.

3.5. Model performance evaluation

For the modelled and measured monthly mean concentrations shown in Figure 5a, 60 performance measure values were calculated for each model (12 measurement periods × 5 performance measures) (data not shown). For the constant emission scenario, ADMS met 15 (25%) of the Chang and Hanna (2004) acceptability criteria whereas AERMOD met 24 (40%). On the other hand, the correlation (R) between modelled and measured mean concentrations was up to 34% higher for ADMS for 10 of the 12 measurement periods, suggesting that this model better represents the spatial variability of the concentrations (data not shown). The use of time-varying emissions improved the performance of both models with ADMS meeting 19 (32%) and AERMOD meeting 34 (57%) of the acceptability criteria.

For the annual simulations AERMOD also met more of the acceptability criteria than ADMS for both the constant and time-varying emission scenarios (Table 3). The poorer performance of ADMS for both scenarios was due to under-prediction of concentrations. Similarly to the annual simulations, the correlation (R) between modelled and measured annual mean concentrations was up to 9% higher for ADMS (data not shown). Again, the use of time-varying emissions improved the performance of both models with respect to the constant emission scenario.

Table 3: Performance measure values for the annual mean concentrations predicted by ADMS and AERMOD for the constant and varying emission scenarios. (Shaded values indicate 'acceptable' model performance according to Chang and Hanna (2004).)

	Constant emissions		Varying emissions	
	ADMS	**AERMOD**	**ADMS**	**AERMOD**
FB	0.77	-0.30	0.68	-0.01
MG	4.0	0.79	3.6	1.1
NMSE	1.2	0.43	0.98	0.29
VG	19	1.6	14	1.6
FAC2	0.40	0.53	0.40	0.67

4. DISCUSSION

4.1. Differences between the models

AERMOD predicted higher concentrations than ADMS both for monthly and annual averaging periods mainly as a result of how the model handles low wind speed conditions. AERMOD uses an estimate of the anemometer threshold to identify calm conditions (0.3 m s^{-1} for this experiment) that is lower than the default threshold in ADMS (0.75 m s^{-1} at a height of 10 m; equivalent to approximately 0.6 m s^{-1} under neutral conditions for the measurement height used here). This means that AERMOD modelled up to 60% more low wind speed periods (< 1 m s^{-1}) than ADMS and since low wind speeds are generally associated with high near-source concentrations, AERMOD predicted higher mean concentrations than ADMS. This can be demonstrated by re-running one of the AERMOD simulations with a similar threshold to that used in ADMS. Changing the threshold in this way reduces annual mean concentrations by 45-80% at all sites and instead of meeting all acceptability criteria, only one (NMSE) is met (data not shown). The performance of AERMOD with the higher threshold, therefore, is comparable to that of ADMS, although the concentrations predicted by AERMOD were still, on average, more than twice those predicted by ADMS. This difference could be the result of the more stable conditions estimated by ADMS and different plume-rise parameterisations (Theobald *et al.*, 2012).

Although with a threshold of 0.3 ms^{-1} AERMOD identifies fewer calm periods than ADMS, there were still a substantial number of hours in the annual simulations (24%) that were not modelled when the wind speed was below this threshold. Since, high near-source concentrations would be expected for these periods, it might be expected that their omission would lead to an underestimate of mean concentrations, which does not seem to be the case for these data. Paine *et al*. (2010) have shown that AERMOD can overestimate concentrations for low wind speeds due to an underestimation of friction velocity for stable low wind conditions, especially for low-level sources. Several solutions have been proposed to correct this overestimation of concentrations (Paine *et al*., 2010; Qian and Venkatram, 2011). It is possible, therefore, that this overestimation during low wind periods is compensated by not modelling calm periods, resulting in concentration predictions that meet the acceptability criteria for model performance.

4.2. Effect of using varying emissions

The use of the emissions model in the simulations effectively increased emissions for warm or windy periods (e.g. day-time, summer) and decreased emissions for cooler, calmer periods (e.g. night-time, winter), when compared with the constant emission scenario. Therefore there was a temporal redistribution of emissions although the total annual emissions were unchanged. This effect was stronger for the slurry lagoons since they are influenced more by the ambient conditions than the livestock houses. This temporal redistribution of emissions increased the ADMS concentration predictions by 14%, on average, because some of the emissions during calm periods (which were not modelled) were shifted to non-calm periods. This shift increased lagoon emissions for the modelled periods by an average of more than 50%, hence increasing the contribution of the lagoon emissions to the overall concentrations. By contrast, the use of the emissions model decreased the AERMOD concentration predictions, on average. Similarly to the ADMS simulations, lagoon emissions during periods with wind speeds above the model threshold increased by nearly 40%, but more importantly lagoon emissions for periods with the lowest modelled wind speeds (< 0.5 ms^{-1}) decreased by about 40%. Since these low wind speed periods make a large contribution to mean concentrations, possibly as a result of overestimating concentrations during these periods, the overall result is a decrease in the predicted mean concentrations.

4.3. Model uncertainty

For both the monthly and annual averaging periods, the 95% confidence intervals of the model predictions were ± a factor of two of the geometric mean concentrations for ADMS and AERMOD, respectively, mainly as a result of uncertainty in emissions. This is maybe not surprising due to the linear relationship between emissions and concentrations in the models and the large uncertainty in the NH_3 emission factors used. Hanna *et al.* (2007) also found that the emission rate was the largest contributor to model uncertainty when AERMOD was used to simulate the dispersion of air pollutants in the Houston Ship Channel area. Bergin *et al.* (1999) and Hanna *et al.* (1998) also identified emission rates as contributing most to model uncertainty for photochemical models, reflecting a widespread uncertainty in emission data.

The five inputs selected for the uncertainty analysis were those judged *a-priori* to be the most uncertain or to have the largest influence on model prediction uncertainty. However, all model inputs are uncertain to some degree and could contribute additional uncertainty although directly measured variables (such as source heights, wind speeds, wind directions etc.) would be expected to contribute less uncertainty than estimated variables (such as emission rates, exit velocities, boundary layer heights etc.). Uncertainty in estimated values of model parameters such as the albedo and the Bowen ratio, will also contribute although AERMOD, for example does not seem to be particularly sensitive to these parameters over small uncertainty ranges (Faulkner, 2008). Concentration predictions will also be influenced by the deposition parameterisation, although Theobald *et al*. (2012) showed that removing the dry deposition entirely changed concentrations by less than 10% for distances up to one km from the source for a similar case study. Neglecting emissions from other sources of NH_3 within the domain, such as fertiliser application, will also add uncertainty to the concentration predictions although this is more important for the monthly than for the annual simulations. However, subtracting the estimated background concentrations and removing outliers should reduce their influence on the measured values. The assumption of the background concentration being the lowest measured value can also add to the uncertainty in the measured concentrations, although it is the best approach available when dealing with long averaging periods. This assumption is most likely to lead to an overestimate of the background concentration since the method assumes that none of the NH_3 measured at the background site came from the emissions of the two farms, which is unlikely for monthly averaging periods. This uncertainty is therefore unlikely to lead to an under-prediction of monthly or annual concentrations.

4.4. Model suitability for impact assessments

Based on the model performance assessment presented here, AERMOD performed better than ADMS for both the monthly and annual simulations, for this case study. However, it is not possible to assess the suitability of the models using this performance assessment, since the acceptability criteria used were designed for model evaluation using research-grade experimental data, not field data with a high degree of uncertainty, such as those used here.

Another way to assess the suitability of the models for impact assessments is to compare the number of sites where the annual critical level of 3 $\mu g\ m^{-3}$ is exceeded (calculated from the measurements) with the model predictions. Measured annual mean concentrations exceeded the critical level at 11 of the 15 measurement sites used in the analyses (taking into account measurement uncertainty). For the scenarios with constant emissions, ADMS predicted exceedance at six of these eleven sites (including prediction uncertainty) and AERMOD predicted exceedance at ten. For the scenarios using the emissions model, ADMS and AERMOD predicted exceedance at six and nine sites, respectively. Again, this indicates that AERMOD performs better than ADMS for this case study.

However, model performance is not only due to model formulation but also depends on the quality of the input data. Model performance in impact assessments could be improved through the use of data with a lower threshold wind speed, e.g. from measurements made using ultra-sonic anemometers, in order to reduce the number of data records with zero wind speed. Routine meteorological measurement networks are starting to use this kind of technology, which has the potential to improve model performance by providing higher quality input data.

5. CONCLUSIONS

- For this case study, AERMOD predicted higher monthly and annual mean atmospheric NH_3 concentrations than ADMS (by an average factor of 6.8 and 5.9, respectively), mainly as a result of the different calm wind speed thresholds used in the models;
- The 95% confidence interval of the model prediction uncertainty due to uncertainty in model inputs was estimated to be ± a factor of two for ADMS and AERMOD, respectively, as a result of a factor of two uncertainty in the emission factors used;
- The use of temporally-varying emission rates improved the performance of both models by increasing the concentration predictions of ADMS and decreasing those of AERMOD;

- Based on established performance measures, AERMOD performed better than ADMS, for this case study;
- These results indicate that AERMOD may be more suited to situations with frequent calm periods, although additional model options for simulating low wind speed periods and improvements in meteorological data quality have the potential to improve the performance of both models.

ACKNOWLEDGEMENTS

We are extremely grateful to the staff of PigCHAMP Pro Europa S.L. for facilitating access to the pig farm and providing farm management data, with special thanks to Gema Montalvo and Juan Carlos Arranz. We are also grateful to Pilar Ortiz García for sample preparation and ammonium analyses, Sim Tang for providing material for the ALPHA samplers and Massimo Vieno for providing the WRF model data. This study was made possible as a result of financial support from the European Commission Project: NitroEurope.

REFERENCES

Achermann, B. and Bobbink, R., 2003. Empirical Critical Loads for Nitrogen, Expert Workshop, Berne, 11–13 November 2002. Environ. Doc. No. 164, 231–257.

Arya, S.P., 2001, Introduction to Micrometeorology, Academic Press, Inc., New York, 415 pp.

Bergin, M.S., Noblet, G.S., Petrini, K., Dhieux, J.R., Milford, J.B., Harley, R.A., 1999. Formal uncertainty analysis of a lagrangian photochemical air pollution model. Environmental science & technology 33, 1116-1126.

Bobbink, R., Hornung, M., Roelofs, J.G.M., 1998. The effects of air-borne nitrogen pollutants on species diversity in natural and semi-natural European vegetation. J. Ecol. 86, 717e738.

Cape, J.N., Eerden, L.J., Sheppard, L.J., Leith, I.D., Sutton, M.A., 2009. Reassessment of critical levels for atmospheric ammonia. Atmospheric Ammonia, 15-40.

Carruthers, D.J., Holroyd, R.J., Hunt, J.C.R., Weng, W.S., Robins, A.G., Apsley, D.D., Thompson, D.J., Smith, F.B., 1994. UK-ADMS: A new approach to modelling dispersion in the earth's atmospheric boundary layer. Journal of Wind Engineering and Industrial Aerodynamics 52, 139-153.

CERC, 2007. ADMS 4 User Guide. Cambridge Environmental Research Consultants Ltd. 331 pp.

Chang, J.C. and Hanna, S.R., 2004. Air quality model performance evaluation. Meteorology and Atmospheric Physics 87, 167-196.

Cimorelli, A.J., Perry, S.G., Venkatram, A., Weil, J.C., Paine, R.J., Wilson, R.B., Lee, R.F., Peters, W.D., Brode, R.W., Pauimer, J.O., 2002. AERMOD: Description of Model Formulation Version 02222.

Defra, 2009. Local Air Quality Management. Technical Guidance LAQM.TG(09). Available from: https://www.gov.uk/government/uploads/system/uploads/attachment_data/file/69334/pb13081-tech-guidance-laqm-tg-09-090218.pdf

EEA (European Environment Agency), 2009. Joint EMEP/EEA Air Pollutant Emission Inventory Guidebook, No. 9/2009. European Environment Agency, Copenhagen.

Environment Agency, 2010. Guidance on modelling the concentration and deposition of ammonia emitted from intensive farming. Available from: http://www.environment-agency.gov.uk/static/documents/Business/Guidance_on__modelling_of_ammonia_from_poultry_pig_farms.pdf

Faulkner, W.B., Shaw, B.W., Grosch, T., 2008. Sensitivity of two dispersion models (AERMOD and ISCST3) to input parameters for a rural ground-level area source. Journal of the Air & Waste Management Association 58, 1288-1296.

Grömping, U., 2006. Relative Importance for Linear Regression in R: The Package relaimpo. Journal of Statistical Software. 17, Issue 1.

Gyldenkærne, S., Skjøth, C.A., Hertel, O., Ellermann, T., 2005. A dynamical ammonia emission parameterization for use in air pollution models. Journal of geophysical research 110, D07108.

Hanna, S.R., Chang, J.C., Fernau, M.E., 1998. Monte carlo estimates of uncertainties in predictions by a photochemical grid model (UAM-IV) due to uncertainties in input variables. Atmospheric Environment 32, 3619-3628.

Hanna, S.R., 2007. Review of Uncertainty and Sensitivity Analyses of Atmospheric Transport and Dispersion Models. In: Borrego, C.; Renner, E., editors. Developments in Environmental Science. Vol. Volume 6. Atlanta: Elsevier Ltd.; 2007. p. 331-351.

Hanna, S.R., Paine, R., Heinold, D., Kintigh, E., Baker, D., 2007. Uncertainties in air toxics calculated by the dispersion models AERMOD and ISCST3 in the houston ship channel area. Journal of Applied Meteorology and Climatology 46, 1372-1382.

Hanna, S.R. and Chang, J., 2012. Setting acceptance criteria for air quality models. Air Pollution Modeling and its Application XXI, 479-484.

Hill, R.A., Parkinson, R.J., Pain, B.F., Phillips, V.R., Lowles, I., 2001. Evaluation of the UK ADMS Buildings Effects Module using data on the near field dispersion of ammonia at an intensive dairy farm. In: Proceedings of the seventh international conference on harmonisation within atmospheric dispersion modelling for regulatory purposes. Joint Research Centre, European Commission, Italy pp. 239–243 (2001)

Iman R and Helton J., 1988. An investigation of uncertainty and sensitivity analysis techniques for computer models. Risk Anal 1988;8:71–90.

Lindeman, R.H., Merenda, P.F. and Gold, R.Z.. 1980. Introduction to Bivariate and Multivariate Analysis, Glenview IL: Scott, Foresman.

Paine, R.; Connors, J.; Szembek, C. AERMOD Low Wind Speed Evaluation Study: Results and Implementation; Paper 2010-A-631-AWMA. Presented at A&WMA's 103rd Annual Conference and Exhibition, Calgary, Alberta, Canada, June 2010.

Qian, W. and Venkatram, A., 2011. Performance of steady-state dispersion models under low wind-speed conditions. Boundary-Layer Meteorology 138, 475-491.

Searle, P.L., 1984. The Berthelot or indophenol reaction and its use in the analytical chemistry of nitrogen-a review. Analyst 109, 549-568.

Seedorf, J., Hartung, J., Schroder, M., Linkert, K., Pedersen, S., Takai, H., Johnsen, J.O., Metz, J., Groot Koerkamp, P., Uenk, G., 1998. A survey of ventilation rates in livestock buildings in northern europe. Journal of Agricultural Engineering Research, 70(1), 39-48.

Tang, Y., Cape, J., Sutton, M., 2001. Development and types of passive samplers for monitoring atmospheric NO2 and NH3 concentrations. The Scientific World 1, 513-529.

Theobald, M.R., Løfstrøm, P., Walker, J., Andersen, H.V., Pedersen, P., Vallejo, A., Sutton, M.A., 2012. An intercomparison of models used to simulate the short-range atmospheric dispersion of agricultural ammonia emissions. Environmental Modelling & Software 37, 90-102.

US EPA, 2003. AERMOD: Latest Features and Evaluation Results. Report EPA-454/R-03-003. US Environmental Protection Agency. Available from: http://www.epa.gov/scram001/7thconf/aermod/aermod_mep.pdf

Vieno, M., Dore, A., Stevenson, D.S., Doherty, R., Heal, M.R., Reis, S., Hallsworth, S., Tarrason, L., Wind, P., Fowler, D., 2010. Modelling surface ozone during the 2003 heat-wave in the UK. Atmospheric Chemistry and Physics 10, 7963-7978.

Wieringa, J., 2001. New revision of davenport roughness classification‖. Proc., 3EACWE, Eindhoven, The Netherlands

APPENDIX: Dispersion model configurations

A1. Emission source data

The location of the ventilation outlets of the pig houses were taken from aerial photographs and their emissions were modelled as elevated point sources with a diameter of 0.5 m. The source height was assumed to be 0.5 m above the building height to simulate the effect of the roof chimney. No data were available for the vertical exit velocities of the emissions and so values were calculated from recommended livestock building flow rates (Seedorf *et al.*, 1998). The mean value (and standard deviation) of the recommendations for sows with piglets is 196 (34) m^{-3} h^{-1} sow^{-1}. For the simulations with time-varying emissions, the modelled exit velocity was scaled by the exit velocity parameter (V) in the emissions model. The modelled sources were assumed to emit at a constant temperature of 22 °C (the mean internal house temperature for Farm 1) for the constant emission simulations and the in-house temperature predicted by the emissions model for the simulations with time-varying emissions. The slurry lagoons were modelled as area sources at ambient temperature with no vertical exit velocity and no building effects, since this feature is not available for area sources in the models. Emissions from the application of organic or mineral fertilisers were not modelled due to insufficient data on the location and timing of these emissions.

A2. Buildings

Buildings were modelled as cuboids with a height equal to the average of the building wall and apex heights (range: 3.9-4.8 m; measured for Farm 1 and estimated for Farm 2). The buildings modules of ADMS and AERMOD were used to take into the account the effect of all buildings of Farms 1 and 2 on the atmospheric dispersion of the building emissions.

A3. Meteorological data

Minimum meteorological data requirements for the AERMOD pre-processor (AERMET) are wind speed, wind direction, air temperature, cloud cover and an estimate of the boundary layer height (e.g. from a radiosonde sounding). No on-site data were available for could cover and boundary layer height and so data were taken from the nearest meteorological station with available data (Segovia, 34 km from the site) and from simulations of the numerical weather prediction model WRF (version 3.1.1), respectively. NCEP/NCAR GFS data were used to initialise the WRF model, as described in Vieno *et al.* (2010).

Minimum meteorological data requirements for ADMS 4.1 are wind speed, wind direction and a way to estimate atmospheric stability (one of either the reciprocal of the Monin-Obukhov length (L), surface sensible heat flux or cloud cover). Segovia cloud cover data were used for the calculation of atmospheric stability. The WRF boundary layer height estimation was also included to be consistent with the AERMOD simulations. The ADMS ‘calms’ option was also tested but the simulations produced an internal error and did not finish correctly.

A4. Aerodynamic roughness length and dry deposition parameterisations

Both models also need an estimate of the mean aerodynamic roughness length (z_0) for the simulation domain. This was estimated from the wind speed profile based on the assumption of a logarithmic profile for near neutral conditions (when there is a negligible vertical temperature gradient). Under these conditions the intercept of the wind speed plotted against the natural logarithm of the measurement height gives z_0 (ignoring any displacement height). From the log-normal distribution of z_0 values the geometric mean value was calculated to be 0.05 m. This value is between the "level country with low vegetation" and "cultivated area with low crops" classifications of Wieringa *et al*. (2001), which is appropriate for the study area.

The dry deposition parameterisations of Theobald et al. (2012) for agricultural land cover were used.

Supplementary Material: Description of the emission model

The ammonia (NH_3) emission model of Gyldenkærne *et al.* (2005) was used to estimate the hourly emissions from the livestock houses and slurry lagoons of the two pig farms. The model distributes the annual emissions of each source using the ambient air temperature and either the wind speed or ventilation rate, for slurry lagoons and animal houses, respectively, whilst maintaining the annual emission factor.

Slurry lagoon emissions

The hourly emissions (kg NH_3 h^{-1}) from the slurry lagoon were calculated as:

$$E(t) = E_{annual} \frac{T(t)^{0.89} \times V(t)^{0.26}}{\sum_{s=1}^{n} T(s)^{0.89} \times V(s)^{0.26}} , \qquad \text{(Equation 1)}$$

where E_{annual} is the total annual emission (kg NH_3 yr^{-1}), T is the air temperature (°C), V is the wind speed (m s^{-1}) and t and s are the calculation time step (hour) and the annual simulation time step (hour). The wind speed and temperature data were taken from the measurements made at a height of 2.74 m. This emission parameterisation has the limitation that emissions are zero when the wind speed is zero, which is not realistic and should be improved in subsequent versions of the model.

Pig house emissions

The hourly emissions (kg NH_3 h^{-1}) from the pig houses were calculated using the same equation as for the slurry lagoon (Equation 1), but with estimates of in-house temperature and ventilation rates instead of the external air temperature and wind speed, respectively. In-house temperatures were estimated for three temperature regimes:

$$T = T_{rec} + \Delta T_{low} \times (T_{out} - T_{\min}), \qquad T_{out} \in [-\infty; T_{\min}]$$

$$T = T_{rec}, \qquad T_{out} \in [T_{\min}; T_{\max}]$$

$$T = T_{rec} + \Delta T_{high} \times (T_{out} - T_{\max}), \qquad T_{out} \in [T_{\min}; \infty]$$

where T_{rec} is the recommended in-house temperature, T_{min} and T_{max} are the temperature boundaries where the ventilation rate is at its minimum and maximum, respectively, ΔT_{low} is the temperature dependence for temperatures below T_{min}, ΔT_{high} is the temperature dependence above T_{max}, and T_{out} is the external air temperature. The parameter values recommended by Gyldenkærne *et al.* (2005) were used for T_{min}, T_{max}, ΔT_{low} and ΔT_{high} (0°C, 12.5°C, 0.5 and 1.0, respectively) and the recommended temperature suggested by the pig house manager was used for T_{rec} (22°C). However, since this model was developed for farms in Denmark, which are subjected to lower external temperatures than the case study used here, this model parameterisation estimated maximum in-house temperatures of 47 °C, which is much higher than the maximum temperature recorded in the building (35 °C). The maximum predicted in-house temperatures were fitted to the recorded value by increasing the parameter T_{max} (the value of the outside temperature at which the ventilation system is fully on) from 12.5 to 26.4 °C. This modification represents a ventilation system that functions over a larger temperature range, which is what is needed in a warmer climate.

The ventilation rate (m s^{-1}) within the pig house (V in Equation 1) was estimated for the same three temperature regimes as the in-house temperature:

$$V = V_{\min}, \qquad T_{out} \in [-\infty; T_{\min}]$$

$$V = V_{\min} + T_{out}\left(\frac{V_{\max} - V_{\min}}{T_{\max} - T_{\min}}\right), \qquad T_{out} \in [T_{\min}; T_{\max}]$$

$$V = V_{\max}, \qquad T_{out} \in [T_{\min}; \infty]$$

where V_{min} and V_{max} are the minimum and maximum ventilation rates (0.2 and 0.38 m s^{-1}, respectively, as suggested by Gyldenkærne *et al.* (2005)).

More details of the derivation and parameterisation of the model and its application to other source types (e.g. natural ventilated livestock buildings) can be found in the original paper (Gyldenkærne *et al.*, 2005).