NERC Open Research Archive
sharing our research

Centre for Ecology & Hydrology
NATURAL ENVIRONMENT RESEARCH COUNCIL.

Article (refereed) - postprint

Contact CEH NORA team at
noraceh@ceh.ac.uk

Upwind impacts of ammonia from an intensive poultry unit

Jones L[1*], Nizam M.S.[3], Reynolds B[1], Bareham S[2], Oxley ERB[4#]

[1] Centre for Ecology and Hydrology Bangor, Environment Centre Wales, Deiniol Road, Bangor, LL57 2UW, UK

[2] Countryside Council for Wales, Maes-y-Ffynnon, Penrhosgarnedd, Bangor, LL57 2DW, North Wales, United Kingdom.

[3] School of Environmental and Natural Resource Sciences, Faculty of Science and Technology, The National University of Malaysia, 43600 UKM Bangi, Selangor, Malaysia

[4] School of Biological Sciences, University of Bangor, Deiniol Road, Bangor LL57 2UW, UK

[#] Deceased

*Corresponding author: LJ@ceh.ac.uk, Telephone: +44 (0) 1248 374500, Fax   +44 (0) 1248 362133

Abstract:

This study investigated potential ammonia impacts on a sand dune nature reserve 600 m upwind of an intensive poultry unit. Ammonia concentrations and total nitrogen deposition were measured over a calendar year. A series of ammonia and nitrogen exposure experiments using dune grassland species were conducted in controlled manipulations and in the field. Ammonia emissions from the intensive poultry unit were detected up to 2.8 km upwind, contributing to exceedance of critical levels of ammonia 800 m upwind and exceedance of critical loads of nitrogen 2.8 km upwind. Emissions contributed 30% of the total N load in parts of the upwind conservation site. In the nitrogen exposure experiments, plants showed elevated tissue nitrogen contents, and responded to ammonia concentrations and nitrogen deposition loads observed in the conservation site by increasing biomass. Estimated long-term impacts suggest an increase in the soil carbon pool of 9% over a 50-year timescale.

Keywords:

Nitrogen deposition; dune grassland; air pollution; agriculture; chicken farm

Capsule:

Ammonia from a poultry unit has upwind impacts, exceeding critical levels 800 m and critical loads 2.8 km upwind, and increasing biomass and tissue N of dune grassland species.

Introduction

Ammonia (NH_3) is a major source of atmospheric nitrogen (N) pollution which has negative consequences for ecosystems adapted to low levels of nitrogen. In Europe, while oxidised nitrogen emissions and deposition have declined since around the 1990s (Erisman et al., 2001; Verstreng et al., 2008), emissions of ammonia have remained virtually the same, with the result that ammonia is increasingly the dominant source of atmospheric nitrogen pollution (Fowler et al., 2004; RoTAP, 2012).

Both dry gaseous ammonia and wet deposited ammonium are damaging to natural systems. Experimental field-release of ammonia to a bog system has shown that ammonia is toxic to both vascular plants such as *Calluna vulgaris* and particularly lower plants such as *Cladonia* lichens (Sheppard et al., 2008, 2009). Lichen photosynthetic capacity is reduced at high ammonia concentrations (Paoli et al., 2010). Critical levels represent the thresholds defined for gaseous pollutant concentrations above which harmful effects are likely to occur, and critical levels for ammonia have been reduced to 3 µg m^{-3} for vascular plants and to the lower level of 1 µg m^{-3} to protect lichens and bryophytes (Cape et al., 2009). Ammonia is also damaging as a component of the total nitrogen deposition load, where the input of nitrogen causes both acidification and eutrophication effects. Ammonia effects on vegetation are documented in a number of reviews (e.g. Bobbink et al., 2010; Krupa, 2003) while nitrogen deposition as a whole causes declines in higher plant diversity (Sala et al., 2000; Stevens et al., 2004), increased susceptibility to secondary stresses (e.g. Power et al.,1998; Strengbom et al., 2002), and altered soil processes (Phoenix et al., 2012).

Sand dunes are a highly biodiverse habitat (Grootjans et al., 2004; Howe et al., 2010) providing many ecosystem services to society (Everard et al., 2010; Ford et al., 2012; Jones et al., 2012). They are also sensitive to N deposition, showing loss of plant species diversity, altered soil processes and accumulated N in plant and soil pools under enhanced deposition (Jones et al., 2004, 2008; Plassmann et al., 2009). As in other habitats, many of these changes are driven by increased biomass of nitrophilous species, particularly graminoids (Remke et al., 2010; van den Berg et al., 2005). The current critical load for 'Fixed dunes with herbaceous vegetation' ("grey dunes") is 8 – 15 kg N ha^{-1}yr^{-1} (Bobbink and Hettelingh,

2011), and bryophytes and lichens comprise a major component of the vegetation cover (Plassmann et al., 2009).

Ammonia gas is primarily a local pollutant, it is highly reactive and deposits readily to vegetation and other surfaces close to its source (Heij and Schneider, 1991) although, in its wet or aerosol NH_4^+ form, it can be transported much greater distances. Intensive animal husbandry units such as poultry farms are major point sources of ammonia. Downwind impacts can be substantial, completely altering vegetation communities close to the farm, causing declines in moss abundance and replacement of sensitive lower plant species by more nitrophilous species at distances of up to 200 m from the farm (Pitcairn et al., 2002, 2009). It is estimated that roughly 4 % of the total ammonia emissions from a farm are deposited within 250 m and are largely responsible for the observed downwind effects (NEGTAP, 2001). The remaining 96 % is assumed to contribute to short and medium range transport of N. Previous impact studies on ammonia point sources have concentrated on downwind effects with the assumption that concentrations upwind of the prevailing wind direction represent a clean background. Upwind effects are assumed to be insignificant and remain unquantified.

The influence of less prevalent wind directions on the pollutant deposition upwind of point sources is a major knowledge gap. Therefore, this study aimed to quantify the upwind influence of a poultry unit on a Natura 2000 designated sand dune system in an area with low background nitrogen deposition, and explore the implications for exceedance of the relevant critical levels and loads. This was conducted by measuring ammonia concentrations and resulting N deposition at varying distances upwind of the poultry unit, and by using experimental exposure in a range of controlled environment and in situ studies to test the sensitivity of dune vegetation to gaseous ammonia and to N deposition. The following hypotheses were tested: 1) Can ammonia emissions from a poultry unit be detected upwind of the prevailing wind direction, and to what distance? 2) If so, are critical levels of ammonia and critical loads of N exceeded upwind? 3) Is ammonia from the local point source likely to be affecting plant species composition upwind?

Methods

Site description and history

Newborough Warren is one of the largest UK sand dune systems, located on the island of Anglesey, North Wales, UK (53:08N 4:21W). It is a Natura 2000 designated site, up to 2 km wide and approximately 1300 hectares in area. The site contains the full succession of habitats from strandline and mobile dunes to dune grassland and scrub, and includes wet and dry slacks. The dominant habitat at the site is Fixed dunes with herbaceous vegetation (Natura code 2130), the older parts of which show succession towards a de-calcified dune grassland containing acidophile species including *Cladonia* species and other lichens, which comprise special interest features of the site (Plassmann et al., 2010). Annual rainfall is 850 mm (Curreli et al., 2013) and the prevailing wind direction is from the south west.

The intensive poultry unit is located approximately 600 m north east of the Natura site. It comprises twelve barns with roof-top ventilation, with capacity for 160,000 – 180,000 birds. There are approximately six rearing cycles per year. Growing cycles run for 56 days, with a clearout period of 1 week in between growing cycles. Ammonia emissions are strongly cyclic, reflecting the birds' growth pattern and management cycle. Continuous ammonia sampler data from other monitoring studies at the poultry unit (NETCEN, 2006; Sheppard, 2003) showed that emission peaks increase through the 56 day growing cycle, and are highest in the last two-thirds of the growing cycle and during the clearing period between cycles when the sheds are emptied and manure removed. The poultry unit was established in 1972 with eight buildings, with a further four buildings constructed in 1978.

This study reports on a twelve-month monitoring campaign of dry and wet N deposition upwind and around the poultry unit, combined with three experiments to test the sensitivity of sand dune species to N. Gaseous ammonia concentrations were measured over a twelve-month period, sampled at eight locations (Figure 1) using a combination of passive diffusion samplers (badge samplers) well suited to monitoring low ammonia concentrations (Tang et al., 2001) at locations upwind of the prevailing wind direction (i.e. to the south west of the poultry unit), and standard ammonia diffusion tubes (Gradko International Ltd, Winchester, UK) at the poultry unit and some additional locations experiencing high ammonia concentrations. Upwind sampling locations for gaseous ammonia were at sites labelled A to F (Figure 1), ranging in distance from 0 to 2800 m to the south west, i.e. upwind from the poultry unit. Locations A to C were at the poultry unit itself and locations to 300 m upwind but outside the Natura 2000 site. Locations D to F were further upwind from 800 to 2800 m,

within the Natura site boundary. The other monitoring sites were located near potential local ammonia sources: a sewage treatment works 520 m to the north-west and improved grazing land at a small livestock farm 1500 m to the south-east. Triplicate badge samplers and triplicate diffusion tubes were exposed in batches lasting one calendar month over an 12-month period, December to the following November. Field blanks and laboratory blanks were used to control for contamination. Missing data for sampling site B (months December and January) and site E (months December to April) were recreated using ratios of the mean ammonia concentrations of each site with means at the immediately adjacent monitoring locations for all periods without missing data. The gap filling was necessary in order to calculate annual means. Data from a separate study funded by the UK Environment Agency (NETCEN, 2006) are reported for comparison. In that study, gaseous ammonia concentrations were measured using triplicate diffusion tubes for a 7-month period, January to July at 17 locations at the farm, upwind and downwind, at distances of 850 m upwind to 450 m downwind. All diffusion tube locations are shown in Figure 1.

Estimating background ammonia concentrations
It was assumed that months with low ammonia concentrations at the poultry unit (monthly mean $< 20 \, \mu g \, m^{-3}$) (January, February, March and June) coincided broadly with the early phase of the growth cycle and could be used to estimate background ammonia concentrations at the Natura site. Wind directions during these sampling periods incorporated a range of directions, although winds were predominantly south-westerly. The calculated average ammonia concentration for site F, furthest from the poultry unit, for months with low ammonia concentrations was $0.60 \, \mu g \, m^{-3}$. This was assumed to represent the true local background in the absence of inputs from the poultry unit. This low background was also observed at sampling locations close to the farm when emissions from the unit were low.

Other components of N deposition
Other components were measured in order to calculate total N deposition. Gaseous nitrogen dioxide concentrations were sampled at one location only, within the farm, using diffusion tubes supplied and analysed by Gradko International Ltd, Winchester, UK, exposed monthly for a seven-month period from January to July. Nitrogen dioxide concentrations show a fairly standard seasonal pattern (Jones et al., 2005), and these data were used to calculate a mean

annual nitrogen dioxide concentration for the 12 month period. This estimate was in close agreement with modelled estimates from national maps (RoTAP, 2012).

Wet deposited nitrogen was sampled in bulk precipitation at a location 650 m east-north-east of the farm at roughly 1 month intervals over a 12-month period. Total inorganic NH_4 and NO_3 concentrations in rainfall samples were measured using Ion Chromatography on a Dionex DX-120 instrument (Dionex, California, USA). Detection limits for NH_4-N and NO_3-N were 0.02 ppm. Organic N was not measured, leading to a likely underestimate of the total N deposition in this study. Rainfall volume and wind direction data were obtained from a coastal meteorological station at RAF Valley 8 km north west of the site. Comparison of wind speed and direction data with on-site measurements taken over a shorter duration showed no significant differences between the two measurement locations.

Estimating total nitrogen deposition load

Ammonia deposition was estimated as the product of the measured ammonia concentration with a fixed deposition velocity of 22 mm s^{-1} established experimentally for sand dune vegetation in a separate study. That study interpreted continuous monitoring of ammonia concentrations in air before and after passing over transplanted turves of sand dune vegetation, along with other micro-meteorological measurements (Jones, 2006). Nitrogen dioxide concentrations were converted to fluxes using a deposition velocity of 1.13 mm s^{-1} (Jones et al., 2004). Concentrations of inorganic N in bulk precipitation were converted to fluxes using rainfall volumes from RAF Valley.

Experiments with sand dune vegetation mesocosms

Mesocosms were constructed with seven sand dune grassland species comprising a mix of fast- and slow-growing species (*Dactylis glomerata, Plantago lanceolata, Festuca rubra, Centaurea nigra, Achillea millefolia, Leontodon hispidus* and *Galium verum*), planted in sand in 4 L pots, 20 cm diameter, containing two individuals of each species in random positions. The mesocosms were exposed to different levels of N in three experiments: as wet-deposited nitrogen; as ammonia, and as phytometers in a gradient study. Each experiment lasted 28 weeks, with an interim vegetation cut at 5 cm above soil surface after 18 weeks. At the end of the experiments the vegetation was cut to ground level and biomass sorted to species, then dried at 105°C to constant weight. In the ammonia fumigation and the phytometer study,

tissue N content of each species was determined by grinding biomass to a fine powder before combustion on a Leco 2000 CHN analyser (LECO corporation, Michigan, USA). There was insufficient biomass to measure tissue N for *G. verum*.

In the first experiment, mesocosms were exposed to four nitrogen deposition levels of 2, 10, 20 and 55 kg N $ha^{-1}yr^{-1}$ in a misting tunnel facility with a background N deposition of 4 kg N $ha^{-1}yr^{-1}$ giving total doses of 6, 14, 24 and 59 kg N $ha^{-1}yr^{-1}$. The facility consisted of eight, fan-vented polytunnels, fitted with fine spray nozzles for automated delivery of nitrogen treatments as wet deposition (Jones, 2005). Nitrogen was applied as NH_4NO_3 in a fine spray, three times weekly. There were two replicate tunnels of each N treatment, with three sand dune mesocosms in each tunnel.

In the second experiment, mesocosms were exposed in an ammonia fumigation experiment using replicated open-top chambers (N=2 for each ammonia treatment), with three mesocosms in each. Ammonia was supplied using open dishes of ammonia solution concentrations of 0 (control), 1, 5 and 10%. Gaseous ammonia concentrations in the open top chambers were measured using passive diffusion tubes at canopy level, exposed in triplicate over a monthly duration. These gave average (±s.d.) measured concentrations over the duration of the 28 week experiment of 0.4 (±0.2) (control), 4.3 (±0.1), 22.1 (±3.2) and 35.0 (±0.5) $\mu g\ m^{-3}$, giving deposition loads of 2, 24, 126 and 200 kg N $ha^{-1}yr^{-1}$ respectively, using a deposition velocity to sand dune vegetation of 22 mm s^{-1} (Jones, 2006).

In the third experiment, mesocosms were exposed as phytometers placed in the field to assess effects of ammonia concentration in a gradient study of declining ammonia concentrations upwind of the prevailing wind direction from the poultry unit. Four replicate phytometers were exposed at each of the upwind ammonia sampling sites A to E. Phytometers at site B suffered an unknown growing problem during the early exposure period and were excluded from subsequent analysis.

Potential impact of additional N on soil development
Two approaches can be used to estimate impacts of additional N on soil chemistry and soil C pools. The first makes the assumption that C stocks remain more-or-less static and all additional N drives down the C:N ratio. The second assumes that C:N ratios remain constant,

therefore all additional N translated into plant growth essentially fixes the same proportion of C, and both C and N stocks in the soil increase (Emmett et al., 2010; Rowe et al., 2011). The latter assumption is the more common. Using this approach, we can calculate the theoretical input of carbon into sand dune soils in the Natura site as a result of extra N deposition resulting from the poultry unit. We assumed an average of 92.3 % retention of additional N into the soil pool, derived from a leachate study, where the remaining 8% is leached 0-1 kg NO_3-N ha^{-1}yr^{-1}, and 2-3 kg dissolved organic N (DON) ha^{-1}yr^{-1} (Jones, 2010), and a C:N mass ratio of 16.9 (data from Jones et al., 2004) to calculate the additional N and C added to the soil pool in the sand dune system over a 50-year period, assuming no change in ammonia emissions from the poultry unit over that period.

Statistical analysis

Statistical relationships between monthly measured ammonia concentrations at all passive sampler and diffusion tube monitoring sites and monthly concentrations measured at the farm were compared using Pearson correlations. Total biomass, individual species biomass, and species tissue N concentration data from replicate mesocosms were pooled for each N treatment before analysis. Statistical differences between treatments in N manipulation experiments, or distances from the farm were assessed by analysis of variance using Generalised Linear Models. All statistical analyses were carried out using Minitab v15.

Results

Ammonia profile

Results from the badge-sampler measurements of ammonia concentrations upwind showed that the highest concentrations occur nearby the poultry unit, as expected (Figure 2). Annual mean concentrations declined rapidly upwind from 60.1 µg m^{-3} at the poultry unit to 6.3 µg m^{-3} at 300 m upwind, dropping to 1.2 µg m^{-3} at site D (800 m upwind, inside the Natura site boundary), and 0.9 µg m^{-3} at the furthest sampling site F (2800 m upwind, 1200 m inside the Natura site). Ammonia concentrations showed a similar pattern of decline both upwind and downwind. Wind direction data from RAF Valley showed that wind directions were predominantly from the south west quarter during the 12-month monitoring period, but wind came from the north east quarter for some of the time (mean daily windspeed exceeded 9 m s^{-1} for a total of 16 days, with 2 days in December where mean daily windspeed exceeded 14 m

s^{-1}). When winds did come from the north east quarter, this occurred during periods of both low and high emissions from the poultry unit.

Monthly concentrations showed a sequence of peaks and troughs (Figure 3), with the highest monthly mean at the poultry unit (Site A) reaching 178 µg m^{-3} in September. Examination of the temporal profile showed that the monthly peaks at all sites upwind corresponded with peaks at the poultry unit, even at the maximum monitored distance of 2800 m upwind at Site F (Figure 3). Ammonia concentrations at all sites upwind were highly correlated with those at the poultry unit itself (all upwind locations: $F > 25.6$, $p < 0.001$).

Peak monthly mean concentrations for the monitoring locations at other possible local sources of ammonia were considerably lower than at the poultry unit, with a maximum of 3.23 µg m^{-3} at the sewage works (mean ± s.d. = 1.94±0.73 µg m^{-3}) and 1.90 µg m^{-3} (mean ± s.d. = 1.42±0.25 µg m^{-3}) at the livestock farm. Temporal patterns of emissions from these minor sources differed from that of the poultry unit, and ammonia concentrations were weakly (sewage works: $F = 5.65$, $p = 0.036$) or not at all (livestock farm: $F = 0.29$, $p = 0.617$) correlated with those at the poultry unit. Ammonia concentrations within the Natura site were only correlated with emissions from the poultry unit, they were not correlated with those from the sewage works or nearby livestock farm. This suggests that these small-scale sources locally influence the ammonia concentrations in their immediate vicinity, but did not influence the Natura site.

Calculating additional ammonia concentrations from poultry unit
Assuming a background ammonia concentration of 0.60 µg m^{-3} as an annual mean, and that all excess ammonia above this level derived from the poultry unit (see methods), excess ammonia concentrations were calculated. The annual mean ammonia concentration at Site D was 1.2 µg m^{-3} suggesting that the poultry unit contributed an additional 0.6 µg m^{-3} averaged over the year, causing exceedance of the critical level of 1 µg m^{-3} inside the Natura site.

Calculating total nitrogen deposition load
Deposition from dry gaseous ammonia concentrations at background levels was 3.43 kg N ha^{-1}yr^{-1}, while deposition from dry gaseous nitrogen dioxide, and from wet inorganic N in bulk deposition were 0.67 and 3.60 kg N ha^{-1}yr^{-1} respectively, giving a total background

deposition of 7.7 kg N ha^{-1}yr^{-1}. Deposition loads attributable to the excess ammonia concentration from the poultry unit were very high (Table 1), ranging from 33 to 340 kg N ha^{-1}yr^{-1} within 300 m of the unit, and 3.40 kg N ha^{-1}yr^{-1} at monitoring Site D inside the Natura site boundary. In total, the poultry unit contributed an additional 44 % on top of the background, bringing total deposition within the Natura site to 11.1 kg N ha^{-1}yr^{-1}, clearly exceeding the lower end of the critical load range for Fixed dunes with herbaceous vegetation ("grey dunes") of 8 – 15 kg N ha^{-1}yr^{-1}. In the background deposition, reduced N comprises 65 % of the total, including dry and wet deposition components (N from NH_3 and NH_4). With inputs of ammonia from the poultry unit, the proportion of reduced nitrogen rises to 76 % of the total at Site D.

Impacts of nitrogen on growth of dune species

Experimental exposure of sand dune species mixtures to nitrogen deposition in various forms showed significant effects of nitrogen on plant growth and tissue chemistry. When exposed to wet N deposition as NH_4NO_3, *F. rubra* showed a significant effect of added nitrogen, with higher biomass at deposition levels of 14 kg N ha^{-1}yr^{-1} and above than at 6 kg N ha^{-1}yr^{-1} (Figure 4b), similar to levels of deposition observed at the Natura site. Growth of the other species showed no significant N effect. However, at all nitrogen levels, the biomass was dominated by two relatively nitrophilous species: *D. glomerata* and *P. lanceolata*, with *F. rubra* third and the four other forb species having much lower biomass. Total above-ground biomass showed no significant response to N (Figure 4a), due to high variability between replicates.

In the ammonia fumigation experiment, the combined biomass of the two harvests (Figure 5a) showed a significant increase between ammonia concentrations of 0.4 – 4.3 µg m^{-3} and again between concentrations 4.3 – 22 µg m^{-3}, dose equivalents of 2 - 24 kg N ha^{-1}yr^{-1} and 24 - 126 kg N ha^{-1}yr^{-1} respectively, again showing significant increases in biomass within the deposition range observed at the Natura site. Growth increases appeared to saturate above 22 µg m^{-3}. When biomass was analysed by species (Figure 5b), both *P. lanceolata* and *F. rubra* showed significant biomass increases between 0.4 – 4.3 µg m^{-3}, while *D. glomerata*, and *F. rubra* showed a significant biomass increase from 4.3 – 22 µg m^{-3}. The other species showed no significant effects of ammonia fumigation. All species showed significant increases in

tissue N concentration (Table 2) with each successive treatment, except *D. glomerata* which showed significant increases in tissue N at all but the lowest ammonia treatment.

In the *in situ* phytometer experiment, the combined biomass from both harvests showed a decline with distance from the poultry unit (Figure 6a). Biomass was highest at site A at the poultry unit itself, then declined significantly to sites C (300 m distance) and D (800 m distance, inside the Natura site) which were not different from each other. Biomass then declined again at site E, further into the site. When analysed by species (Figure 6b), biomass of the dominant species *D. glomerata*, showed the same pattern as that of total biomass, i.e. a significant decline with distance from the poultry unit. Biomass of *P. lanceolata* was high but rather variable, being highest at sites A and D. Biomass of *F. rubra* and the other species showed no significant effect of distance from the poultry unit. Five of the six species showed significant changes in tissue N (Table 3): *D. glomerata* and *A. millefolia* showed a significant drop in tissue N at sites D and E inside the Natura site, compared with sites A and C. *F. rubra* and *L. hispidus* showed successive drops from A to C and again to D and E inside the Natura site. Tissue N in *C. nigra* did not differ between sites C, D and E, but was lower at these sites than at the poultry unit (site A).

Estimated nitrogen accumulation in soils

The additional N deposition attributed to the poultry unit of 3.4 kg N ha^{-1}yr^{-1} at site D (Table 1), is calculated to result in an additional 157 kg N ha^{-1} in the soil over a 50-year period which, assuming a constant C:N ratio will result in an additional 2652 kg C ha^{-1} over the same period. When compared with current carbon stocks in the soil of 29,100 kg C ha^{-1} (data from Jones et al., 2004), this represents a 9.1 % increase in the current soil C pool over the 50 year period.

Discussion

This study has shown that emission peaks from an ammonia point source can be detected upwind to a distance of 2.8 km. The low background ammonia concentrations of 0.60 µg m^{-3} are similar to concentrations of 0.62 µg m^{-3} measured at another west coast dune system (Jones et al., 2005), reflecting the relatively small area of improved agricultural land and low livestock densities along much of the west UK coast. This background is much lower than suggested by other UK studies. Often it is assumed that the lowest concentration measured,

usually upwind, represents the background (e.g. Cape et al., 2009). Alternatively, a reference background concentration is assumed based on monitoring network sites in rural locations (NETCEN, 2006). Both of these approaches may be inappropriate for defining background concentrations, due partly to local agricultural sources around rural sites (Dragosits et al., 2002; Paoli et al., 2010), but also due to lack of appreciation of possible episodic upwind effects of large point sources of ammonia, which can be considerable as shown in this study.

This study also shows that the elevated ammonia concentrations upwind can contribute to exceedance of the critical level of 1 $\mu g\ m^{-3}$ for sensitive bryophytes and lichens, and to exceedance of the critical load for total N deposition at distances up to 800 m upwind. Due to the decline of ammonia with distance from source, deposition is highest within the parts of the Natura site nearest the poultry unit. These areas are also the most sensitive to both ammonia and N deposition because they comprise the older grassland and wetland communities on de-calcified soils, to which the lower part of the critical load range 8 - 11 kg N $ha^{-1}yr^{-1}$ should be applied (Bobbink and Hettelingh, 2011). The grasslands are lichen-rich, which are sensitive to direct toxicity effects. The vegetation and soils are particularly susceptible to eutrophication as the soils lie within the pH range of 4.5 – 6.5 (Jones et al., 2004) where phosphorus availability should be at a maximum (Kooijman and Besse, 2002).

The experimental manipulations presented here show that dune vegetation has the potential to respond to levels of total N deposition and to ammonia concentrations observed within the Natura site, but also that this growth response can occur under field conditions. Increases in graminoid biomass have been observed in other dune mesocosm experiments at loads of 20 kg N $ha^{-1}yr^{-1}$ (van den Berg et al., 2005), but the experiments in this study showed significant growth effects below 14 kg N $ha^{-1}yr^{-1}$ in controlled wet deposition experiments and below 11 kg N $ha^{-1}yr^{-1}$ deposition in the field phytometer study. These biomass changes at relatively low levels of N supply are likely to reflect the start of species composition shifts observed in a wide range of other semi-natural habitats at higher levels of N deposition (Heil and Diemont, 1983; Phoenix et al., 2012). Often these large-scale community shifts are triggered by other stresses such as drought and climatic effects, herbivory or pathogen attack (Heil and Diemont, 1993; Power et al.,1998; Strengbom et al., 2002). The increased tissue N concentrations in the ammonia fumigation experiment and the gradient study showed that all

species were utilising the ammonia as a nitrogen source. Elevated tissue N content may be a catalyst for some secondary effects such as increased herbivory.

Ammonia from poultry units is known to cause changes in species composition and tissue chemistry in woodland flora, with nitrophiles replacing oligotrophic species (Pitcairn et al., 2002; 2009), but this is the first time ammonia impacts have been indicated for dry dune species. However, there is mounting evidence of total N deposition impacts on dune grasslands: Positive relationships of N with biomass of dune vegetation in semi-fixed 'yellow-dune' grassland have been reported from gradient studies in the UK (Jones et al., 2004), but are harder to detect in established fixed 'grey-dune' grassland as these are often grazed by rabbits or livestock. Nonetheless, gradient studies have shown that species composition of these older dune grasslands was reduced by nitrogen deposition at loads between 10 and 20 kg N ha^{-1}yr^{-1} (Jones et al., 2004), and at lower deposition loads in acidic dune systems in the Baltic (Remke et al., 2009). Although not tested in this study, effects on lichen communities at the ammonia concentrations recorded within the Natura site are also likely. Negative effects on lichens have been shown at ammonia concentrations of 1-2 µg m^{-3} (Sutton et al., 2009) in the UK, and shifts in lichen community composition from oligotrophic to mesotrophic species occurred at deposition loads of 3 to 9 kg N ha^{-1}yr^{-1} in the USA (Geiser et al., 2010).

This study predicts long-term effects on soil N and C pools as a result of future N emissions from the poultry unit. Experimental studies have shown elevated N pools in dune vegetation, primarily in the moss biomass (Plassmann et al., 2009). Over longer timescales, as the moss layer decomposes, this nitrogen will subsequently enter the soil N pool, with implications for long-term rates of soil development (Jones et al., 2008). This will almost certainly lead to thicker organic soil layers, driving successional development and facilitating the dominance of competitive species at the expense of typical sand dune species. It is therefore likely that the poultry unit has already had an effect on vegetation and soils within the Natura site over the 38 years since 1972 that it has been operating. Due to complex topographical variation within the sand dune Natura site and gradients of soil age which co-vary with distance from the poultry unit, it is not possible to definitively detect changes in vegetation and soils within the site. However, this should not be taken as evidence of no impact, and there are indications

that lichen assemblages are less species rich in the de-calcified dune grasslands closest to the poultry unit (Woods, 2009).

These findings have major implications for management of sites of conservation interest in close proximity to intensive animal units. Firstly, it should be not be assumed that an upwind location will avoid adverse impacts. Secondly, off-site mechanisms should be considered to reduce the local impact of existing point-source emissions by techniques such as shelter-belts which can capture up to 15 % of ammonia emissions (Loubet et al., 2009; Theobald et al., 2001), in addition to measures to reduce emissions such as altered animal husbandry methods. Care should also be taken to avoid pollution swapping, i.e. the transfer of atmospheric pollution to soil or water eutrophication as a result of such mitigation measures. Modelling of ammonia concentration fields and wider landscape planning are useful tools to mitigate potential ammonia impacts on sensitive ecosystems (Cellier et al., 2012).

Summary:
In conclusion, this study has established that ammonia emissions from an intensive poultry unit can be detected up to 2.8 km upwind, and that these emissions contribute to exceedance of critical levels of ammonia and exceedance of critical loads of total N. Nitrogen exposure experiments show that plants take up the ammonia as a nutrient, and respond to both ammonia levels and nitrogen deposition loads at the ammonia concentrations observed in the Natura site by increasing biomass. Therefore the observed ammonia contribution from the poultry unit is likely to be affecting plant growth within the Natura site, and having long term effects on soils.

References

Bobbink, R., Hettelingh, J.P. (Eds), 2011. Review and revision of empirical critical loads and dose-response relationships. Proceedings of an expert workshop, Noordwijkerhout, 23-25 June 2010. ISBN: 978-90-6960-251-6.
http://www.rivm.nl/bibliotheek/rapporten/680359002.pdf

Bobbink, R., Hicks, K., Galloway, J., Spranger, T., Alkemade, R., Ashmore, M., Bustamante, M., Cinderby, S., Davidson, E., Dentener, F., Emmett, B., Erisman, J.W., Fenn, M., Gilliam, F., Nordin, A., Pardo, L. De Vries, W., 2010. Global assessment of nitrogen deposition effects on terrestrial plant diversity: a synthesis. Ecological Applications 20, 30-59.

Boxman, A.W., Van der Ven, P.J.M., Roelofs, J.G.M., 1997. Ecosystem recovery after a decrease in nitrogen input to a Scots pine stand at Ysselsteyn, the Netherlands. Forest Ecology and Management 101, 155–163.

Cape, J.N., van der Eerden, L.J., Sheppard, L.J., Leith, I.D., Sutton, M.A., 2009. Evidence for changing the critical level for ammonia. Environmental Pollution 157, 1033-1037.

Curreli, A., Wallace, H., Freeman, C., Hollingham, M., Stratford, C., Johnson, H., Jones, L., 2013. Eco-hydrological requirements of dune slack vegetation and the implications of climate change. Science of the Total Environment 443, 910-919.

Dragosits, U., Theobald, M.R., Place, C.J., Lord, E., Webb, J., Hill, J., ApSimon, H.M., Sutton, M.A., 2002. Ammonia emission, deposition and impact assessment at the field scale: a case study of sub-grid spatial variability. Environmental Pollution 117, 147–158.

Emmett, B.A., Reynolds, B., Chamberlain, P.M., Rowe, E., Spurgeon, D., Brittain, S.A., Frogbrook, Z., Hughes, S., Lawlor, A.J., Poskitt, J., Potter, E., Robinson, D.A., Scott, A., Wood, C. & Woods, C., 2010. Countryside Survey: Soils report in 2007. pp. 192. NERC/Centre for Ecology & Hydrology.

Erisman, J.W., Otjes, R., Hensen, A., Jongejan, P., van den Bulk, P., Khlystov, A., Möls, H., Slanina, S., 2001. Instrument development and application in studies and monitoring of ambient ammonia. Atmospheric Environment 35, 1913– 1922.

Everard, M., Jones, L., Watts, B., 2010. Have we neglected the societal importance of sand dunes? – An Ecosystem Services perspective. Aquatic Conservation: Marine and Freshwater Ecosystems 20, 476-487.

Ford, H., Garbutt, A., Jones, D.L., Jones, L., 2012. Impacts of grazing abandonment on ecosystem service provision: Coastal grassland as a model system. Agriculture, Ecosystems and Environment 162, 108– 115.

Fowler, D., O'Donoghue, M., Muller, J.B.A., Smith,R.I., Dragosits, U., Skiba, U., Sutton, M.A. and Brimblecombe, P., 2004. A chronology of nitrogen deposition in the UK between 1900 and 2000. Water Air Soil Pollution Focus 4, 9-23.

Geiser, L.H., Jovan, S.E., Glavich, D.A., Porter, M.K., 2010. Lichen-based critical loads for atmospheric nitrogen deposition in Western Oregon and Washington Forests, USA. Environmental Pollution 158, 2412-2421.

Grootjans, A.P., Adema, E.B., Bekker, R.M., Lammerts, E.J., 2004. Why coastal dune slacks sustain a high biodiversity. In: Martinez M.L., Psuty N.P., (Eds). Coastal Dunes: Ecology and Conservation. Ecological Studies. Springer, Berlin. pp. 85-101.

Heij, G.J., Schneider, T., (Eds.), 1991. Acidification Research in the Netherlands: Final Report of the Dutch Priority Programme on Acidification. Elsevier, Amsterdam.

Heil, G.W., Diemont, W.H., 1983. Raised nutrient levels change heathland into grassland. Vegetatio 53, 113-120.

Jones, M.L.M., 2005. Nitrogen deposition in upland grasslands: Critical loads, management and recovery. PhD Thesis, University of Sheffield.

Jones, M.L.M., Angus, S., Cooper, A., Doody, P., Everard, M., Garbutt, A., Gilchrist, P., Hansom, G., Nicholls, R., Pye, K., Ravenscroft, N., Rees, S., Rhind, P. & Whitehouse, A., 2011. Coastal Margins [chapter 11]. In: UK National Ecosystem Assessment. Understanding nature's value to society. Technical Report. UNEP-WCMC, Cambridge, 411-457.

Jones, M.L.M., 2010. Impacts of an intensive poultry unit on a sand dune SAC. Report to Countryside Council for Wales, July 2010. CCW Contract No: 941.

Jones, M.L.M., Pilkington, M.G., Healey, M., Norris, D.A., Brittain, S.A., Tang, Y.S., Reynolds, B., 2005. Determining a nitrogen budget for Merthyr Mawr sand dune system. Final report for Countryside Council for Wales. CEH Project No: C02352NEW, CCW Contract No: FC 72-02-59. May 2005.

Jones, M.L.M., Sowerby, A., Williams, D.L., Jones, R.E., 2008. Factors controlling soil development in sand dunes: evidence from a coastal dune soil chronosequence. Plant and Soil 307, 219-234.

Jones, M.L.M., Wallace, H.L., Norris, D., Brittain, S.A., Haria, S., Jones, R.E., Rhind, P.M., Reynolds, B.R., Emmett, B.A., 2004. Changes in vegetation and soil characteristics in coastal sand dunes along a gradient of atmospheric nitrogen deposition. Plant Biology 6, 598-605.

Jones, M.R., 2006. Ammonia deposition to semi-natural vegetation. PhD Thesis, Edinburgh University.

Kooijman, A.M., Besse, M., 2002. The higher availability of N and P in lime-poor than in lime- rich coastal dunes in the Netherlands. Journal of Ecology 90, 394-403.

Krupa, S.V., 2003. Effects of atmospheric ammonia NH3 on terrestrial vegetation: a review. Environmental Pollution 124, 179-221.

Loubet, B., Asman, W.A., Theobald, M.R. Hertel, O., Tang, S.Y., Robin, P., Hassouna, M., Dammgen, U., Genermont, S., Cellier, P., Sutton, M.A., 2009. Ammonia deposition near hot spots: processes, models and monitoring methods. In: Sutton, M.A., Reis, S., Baker, S.M.H., (Eds). Atmospheric Ammonia: Detecting Emission Changes and Environmental Impacts – Results of an Expert Workshop under the Convention on Long-range Transboundary Air Pollution, Springer, New York.

NEGTAP, 2001. Transboundary air pollution: acidification, eutrophication and ground-level ozone in the UK, National Expert Group on Transboundary Air Pollution, Edinburgh.

NETCEN, 2006. A Diffusion Tube Survey in the Vicinity of the Newborough Poultry Farm 21 January 2005 to 20 July 2005. Report for the Environment Agency.

Paoli, L., Pirintsos, S.A., Kotzabasis, K., Pisani, T., Navakoudis, E., Loppi, S., 2010. Effects of ammonia from livestock farming on lichen photosynthesis. Environmental Pollution 158, 2258-2265.

Phoenix, G.K., Emmett, B.A., Britton, A.J., Caporn, S.J.M., Dise, N.B., Helliwell, R., Jones, L., Leake, J.R., Leith, I.D., Sheppard, L.J., Sowerby, A., Pilkington, M.G., Rowe, E.C., Ashmore, M.R., Power, S.A., 2012. Impacts of atmospheric nitrogen deposition: responses of multiple plant and soil parameters across contrasting ecosystems in long-term field experiments. Gobal Change Biology 184, 1197-1215.

Pitcairn, C.E.R., Leith, I.D., van Dijk, N., Sheppard, L.J., Sutton, M.A., Fowler, D., 2009. The application of transects to assess the effects of ammonia on woodland groundflora. In: Sutton, M.A., Reis, S., Baker, S.M.H., (Eds.) Atmospheric Ammonia: Detecting emission changes and environmental impacts. Results of an Expert Workshop under the Convention on Long-range Transboundary Air Pollution. Springer, New York. Pp 59-69.

Pitcairn, C.E.R., Skiba, U.M., Sutton, M.A., Fowler, D., Munro, R., Kennedy, V., 2002. Defining the spatial impacts of poultry farm ammonia emissions on species composition of adjacent woodland groundflora using Ellenberg Nitrogen Index, nitrous oxide and nitric oxide emissions and foliar nitrogen as marker variables. Environmental Pollution 119, 9-21.

Plassmann, K., Edwards-Jones, G., Jones, M.L.M., 2009. The effects of low levels of nitrogen deposition and grazing on dune grassland. Science of the Total Environment 407, 1391-1404.

Plassmann, K., Jones, M.L.M., and Edwards-Jones, G., 2010. Effects of long-term grazing management on sand dune vegetation of high conservation interest. Applied Vegetation Science 13, 100-112.

Power, S.A., Ashmore, M.R., Cousins, D.A., Sheppard, L.J., 1998. Effects of nitrogen addition on the stress sensitivity of Calluna vulgaris. New Phytologist 138, 663-673.

Remke, E., Brouwer, E., Kooijman, A., Blindow, A., Roelofs, J.G.M., 2009. Low atmospheric nitrogen loads lead to grass encroachment in coastal dunes, but only on acid soils. Ecosystems 12: 1173–1188.

Remke, E., Brouwer, E., Kooijman, A.M., Blindow, I., Esselink, H., Roelefs, J.G.M., 2009. Even low to medium nitrogen deposition impacts vegetation of dry, coastal dunes around the Baltic Sea. Environmental Pollution 157, 792-800.

RoTAP, 2012 Review of Transboundary Air Pollution: Acidification, Eutrophication, Ground Level Ozone and Heavy Metals in the UK. Contract Report to the Department for Environment, Food and Rural Affairs. Centre for Ecology & Hydrology.

Rowe, E.C., Jones, M.L.M., Henrys, P.A., Smart, S.M., Tipping, E., Mills, R.T.E., Evans, C.D., 2011. Predicting effects of N pollutant load on plant species based on a dynamic soil eutrophication indicator. Countryside Council for Wales. Science Report 977., p. 39.

Sala, O.E., Chapin, F.S., Armesto, J.J., Berlow, E., Bloomfield, J., Dirzo, R., Huber-Sanwald, E., Huenneke, L.F., Jackson, R.B., Kinzig, A., Leemans, R., Lodge, D.M., Mooney, H.A., Oesterheld, M., Poff, N.L., Sykes, M.T., Walker, B.H., Walker, M., Wall, D.H., 2000. Biodiversity - Global biodiversity scenarios for the year 2100. Science, 287, 1770-1774.

Sheppard, L.J., Leith, I.D., Crossley, A., Van Dijk, N., Fowler, D., Sutton, M.A., Woods, C., 2008. Stress responses of *Calluna vulgaris* to reduced and oxidised N applied under `real world conditions'. Environmental Pollution 154 3, 404-413.

Sheppard, L.J.; Leith, I.D.; Crossley, A.; van Dijk, N.; Cape, J. N.; Fowler, D.; Sutton, M.A., 2009. Long-term cumulative exposure exacerbates the effects of atmospheric ammonia on an ombrotrophic bog: implications for critical levels. In: Sutton, M.A., Reis, S., Baker, S.M.H., (Eds.) Atmospheric Ammonia: Detecting emission changes and environmental impacts. Results of an Expert Workshop under the Convention on Long-range Transboundary Air Pollution. Springer, 49-58.

Sheppard, V. 2003. Study of Ambient Air Quality at Newborough, 19th June 2003 to 15th October 2003. Monitoring and Assessment Process. Environment Agency Report M&A/CCM/TR/2003/009/M.

Stevens, C.J., Dise, N.B., Mountford, J.O. & Gowing, D.J., 2004. Impact of nitrogen deposition on the species richness of grasslands. Science, 303, 1876-1879.

Strengbom, J., Nordin, A., Näsholm, T. and Ericson, L., 2002. Parasitic fungus mediates change in nitrogen-exposed boreal forest vegetation. Journal of Ecology 90, 61-67.

Sutton, M.A., Wolseley, P.A., Leith, I.D., van Dijk, N., Tang, S.Y., James, P.W., Theobald, M.R., Whitfield, C., 2009. Estimation of the ammonia critical level for epiphytic lichens based on observations at farm, landscape and national scales. In: Sutton, M.A., Reis, S., Baker, S.M.H., (Eds.) Atmospheric Ammonia: Detecting emission changes and environmental impacts. Results of an Expert Workshop under the Convention on Long-range Transboundary Air Pollution. Springer, New York, pp 71-86.

Tang, Y.S., Cape, J.N., Sutton, M.A., 2001. Development and types of passive diffusion samplers for monitoring atmospheric nitrogen dioxide and ammonia concentrations., Proceedings of the International Symposium on Passive Sampling of Gaseous Air Pollutants in Ecological Effects Research. The Scientific World, pp. 513-529.

Theobald, M.R., Milford, C., Hargreaves, K.J. Sheppard, L.J., Nemitz, E., Tang, Y.S, Phillips, V.R., Sneath, R., McCartney, L., Harvey, F.J., Leith, I.D., Cape, J.N., Fowler, D., Sutton, M.A., 2001. Potential for ammonia recapture by farm woodlands: design and application of a new experimental facility. The Scientific World, 1 S2, 791 –801.

van den Berg, L.J.L., Tomassen, H.B.M., Roelofs, J.G.M., Bobbink, R., 2005. Effects of nitrogen enrichment on coastal dune grassland: A mesocosm study. Environmental Pollution 138, 77-85.

Vestreng, V., Nziachristos, L., Semb, A., Isaksen, I.S.A. and Tarrason, L., 2008. Evolution of NOx emissions in Europe with focus on road transport control measures. Atmospheric Chemistry and Physics Discussions, 8, 10697–10747.

Woods, R.G., 2009. Impact of ammonia emissions from local sources. Survey Report by Plantlife to Countryside Council for Wales, May 2009.

Figure Legends

Figure 1. Map showing study context, including sampling points for gaseous ammonia along upwind transect (A-F) and smaller point sources the sewage works (to the NW) and small livestock farm (to the SE). Annual wind rose shows 10-yr average wind direction and frequency (1994-2003) for RAF Valley, a nearby coastal meteorological station.

Figure 2. Pattern of mean annual ammonia concentration ($\mu g\ m^{-3}$) with distance upwind (-) and downwind (+) from the poultry unit (m), measured in two studies. Black squares - data from this study (12 months, December to the following November); open diamonds – bias-corrected diffusion tube data from NETCEN (2006) for comparison (7 months, January to July).

Figure 3. Temporal pattern of gaseous ammonia concentrations for sampling locations upwind of the poultry unit (a) at sites A to F, and (b) showing greater resolution for sites D to F within the Natura site boundary. See Figure 1 for location details. Thick horizontal line shows critical level (annual mean) of $1\ \mu g\ m^{-3}$.

Figure 4. Dry weight biomass in wet nitrogen deposition (NH_4NO_3) experiment using mesocosms of sand dune species, showing a) total biomass and b) biomass by species. Differing letters denote significant differences between treatments for *F. Rubra* ($p<0.05$), all other spp. and total biomass non significant, (ANOVA, N=2). Bars represent ± 1 s.e.

Figure 5. Dry weight biomass in ammonia fumigation experiment using mesocosms of sand dune species, showing a) total biomass and b) biomass by species. Differing letters (*a, a, A, .a etc.*) denote significant differences between treatments for total biomass ($p<0.001$) and for *D. Glomerata* ($p<0.001$)*, P. lanceolata* ($p<0.001$)*, F. Rubra* ($p<0.001$) respectively, all other spp. non significant, (ANOVA, N=2). Bars represent ± 1 s.e.

Figure 6. Dry weight biomass for the in situ ammonia gradient phytometer experiment using mesocosms of sand dune species at sites A, C, D, E (for distances upwind refer to Table 1), showing a) total biomass and b) biomass by species. Differing letters (*a, a, A, .a etc.*) denote significant differences between treatments for total biomass and for *D. Glomerata* ($p<0.001$)*,

P. lanceolata (p<0.01)*, C. nigra* respectively (p<0.05), all other spp. non significant, (ANOVA, N=4). Bars represent ± 1 s.e.

Table 1. Nitrogen deposition (kg N ha^{-1}yr^{-1}) at locations upwind of the poultry unit. STW = Sewage Treatment Works. Background N deposition, excluding any dry ammonia contribution, comprises 4.27 kg N ha^{-1}yr^{-1} from dry nitrogen dioxide deposition and bulk wet deposition of ammonium and nitrate (see Results).

Location	Distance upwind (m)	N dep from dry NH$_3$	Total N dep incl. all N sources	Excess N dep from poultry unit
A	0	343.3	347.6	339.9
B	100	84.3	88.6	80.9
C	300	36.2	40.5	32.8
D	800	6.8	11.1	3.4
E	2000	4.0	8.3	0.6
F	2800	5.4	9.7	1.9
STW		11.1	15.4	Not estimated

Table 2. Tissue N concentrations (%) of six dune grassland species exposed to ammonia at four concentrations (equivalent N deposition in brackets) in open top chambers. For each species, different letters denote significant differences between treatments, (ANOVA, N=2). Standard error calculated using pooled variance across treatments.

NH$_3$ treatment, μg m^{-3} (kg N ha^{-1} yr^{-1})	0.4 (2)	4.3 (24)	22 (126)	35 (200)	Pooled SE
Dactylis glomerata	0.98 [a]	0.81 [a]	2.00 [b]	2.80 [c]	0.11
Plantago lanceolata	0.76 [a]	1.21 [b]	1.78 [c]	2.55 [d]	0.05
Festuca rubra	0.81 [a]	1.15 [b]	2.42 [c]	3.31 [d]	0.08
Centaurea nigra	0.74 [a]	1.10 [b]	1.87 [c]	3.02 [d]	0.08
Achillea millefolium	0.89 [a]	1.27 [b]	1.87 [c]	2.46 [d]	0.05
Leontodon hispidus	1.06 [a]	1.49 [b]	2.38 [c]	3.53 [d]	0.14

Table 3. Tissue N concentrations (%) of six dune grassland species in phytometers placed at increasing distance (m) upwind of the poultry unit. For each species, different letters denote significant differences between treatments, (ANOVA, N=4). Standard error calculated using pooled variance across treatments.

	Site A (0 m)	Site C (300 m)	Site D (800 m)	Site E (2000 m)	Pooled SE
Dactylis glomerata	1.42 [a]	1.26 [a]	0.87 [b]	0.97 [b]	0.11
Plantago lanceolata	0.89 [a]	0.77 [b]	0.47 [c]	0.76 [b]	0.03
Festuca rubra	1.73 [a]	1.16 [b]	0.79 [c]	0.79 [c]	0.07
Centaurea nigra	1.30 [a]	0.97 [b]	0.81 [c]	0.77 [c]	0.09
Achillea millefolium	1.39 [a]	1.36 [a]	0.79 [b]	0.77 [b]	0.08
Leontodon hispidus	1.67 [a]	1.44 [b]	1.00 [c]	1.07 [c]	0.08

Figure 1.
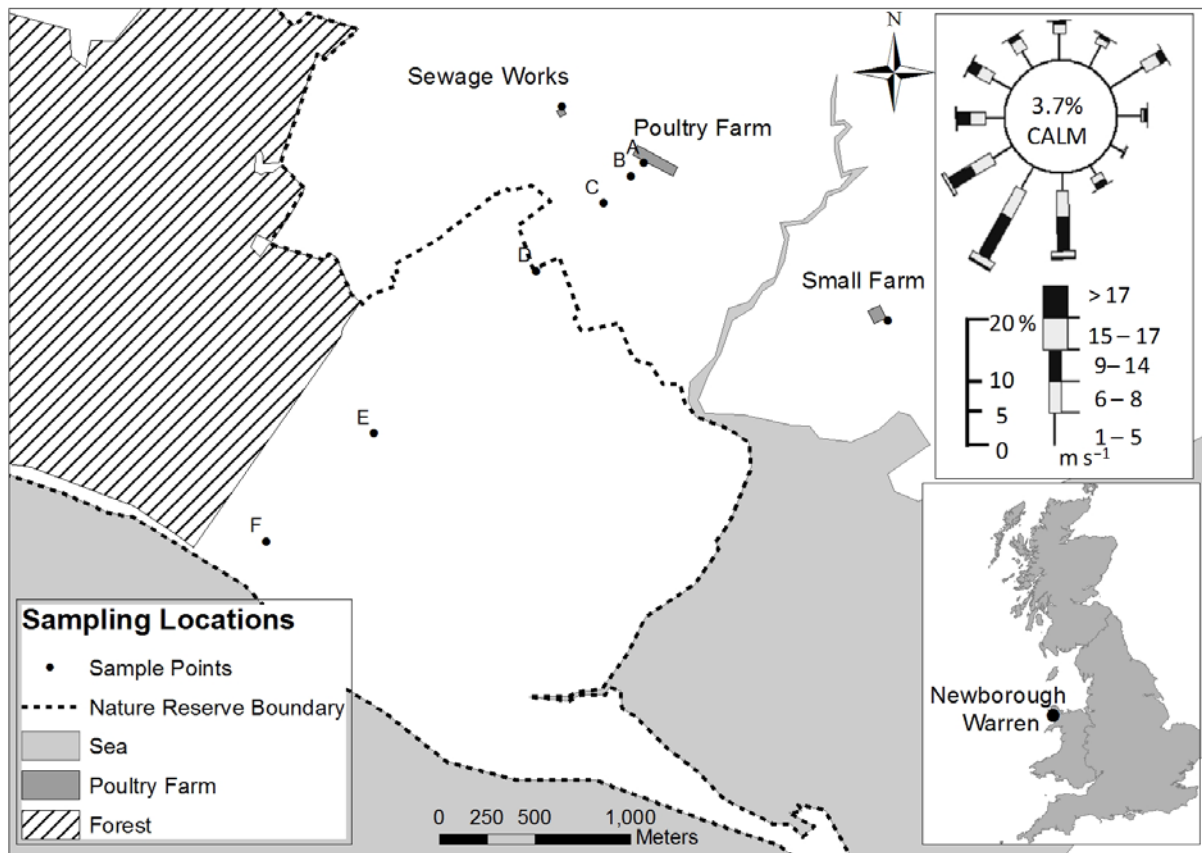
Figure 2.

Figure 3.

Figure 4.

a)

b)

Figure 5.

Figure 6.

a)

b)