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Modeling and measurements of ammonia from poultry operations: Their emissions, transport, and deposition in the Chesapeake Bay

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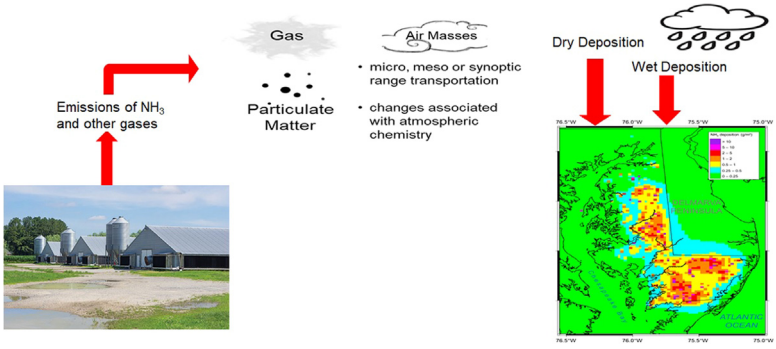
HIGHLIGHTS

- Ammonia deposition to the Chesapeake Bay from Poultry CAFOs.
- AERMOD Modeling study.
- Sensitivity of ammonia deposition to deposition velocity.
- Measurements of atmospheric ammonia on Delmarva Peninsula.

GRAPHICAL ABSTRACT



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ABSTRACT

The goal of this study is to determine how much ammonia/nitrogen is being deposited to the Maryland Eastern Shore land and the Chesapeake Bay from poultry operations on Maryland's Eastern Shore. We simulated the fate of ammonia/nitrogen emitted (using emission factors from the U.S. EPA in conjunction with Carnegie-Mellon University) from 603 poultry facilities using the air quality model, AERMOD. The model domain was approximately 134 km by 230 km (and covers the full land area of Maryland's Eastern Shore), with a horizontal resolution of 2 km by 2 km. Ammonia concentration observations were made at 23 sites across Maryland's Eastern Shore during two periods (September and October 2017) in order to calibrate the model. An ammonia deposition velocity of 2.4 cm/sec was selected based on the sensitivity analysis of results for the simulation of a large poultry facility, and this value fell within the range of measurements reported in the scientific literature downwind of Concentrated Animal Feeding Operations (CAFOs). The ammonia deposition velocity of 2.4 cm/s leads to an estimated total annual ammonia deposition of 11,100 Megagrams/year (10,600 Mg/yr deposition to land, and 508 Mg/yr deposition to water (1 Mg = 1,000,000 g = 1.1023 US Tons)). In addition, model simulations indicate that ~72.4% of ammonia emissions from poultry animal feeding operations would be deposited within the modeling domain. However, this deposited ammonia/nitrogen may be transported through waterways from the land mass and ground water to the Chesapeake Bay. A comprehensive sensitivity analysis of the assumed ammonia deposition velocity (ranging from 0.15 to 3.0 cm/s) on estimated ammonia annual deposition is provided. Using the lower limit of an ammonia deposition velocity of 0.15 cm/s gives much smaller estimated total annual ammonia deposition of 2,040 Mg/yr (1,880 Mg/yr deposition to land and 163 Mg/yr deposition to water).

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1. Introduction and background

The Chesapeake Bay is the nation's largest estuary, but like many waterbodies in the U.S. it is plagued with poor water quality due to excess loads of nitrogen and phosphorus (Sheeder et al., 2002; U.S. EPA, 2010, Da et al., 2018). These excess nutrients cause algal blooms that reduce water clarity, contribute to the Bay's dead zone i.e. areas in the Bay and its tidal rivers with insufficient levels of dissolved oxygen, and drinking water concerns (Boesch et al., 2001; US EPA, 2011; Linker et al., 2013; Beachley et al., 2019; and Walker and Beachley, 2019). Because of these water quality problems, in 2010 the U.S. Environmental Protection Agency (EPA) worked with the six Bay states and the District of Columbia to develop the Chesapeake Bay Total Maximum Daily Load that requires significant reductions in nitrogen, phosphorus and sediment in order to meet water quality standards (US EPA, 2010). Roughly one-third of the nitrogen entering the Bay and its tidal rivers comes from atmospheric deposition, and recent estimates indicate roughly one-half of this is due to ammonia (Paerl et al., 2002: Linker et al., 2013). The main source of this ammonia is animal operations (Aneja et al., 2001; Bittman and Mikkelsen, 2009; Battye et al, 2017).

Although agricultural production is widespread throughout the Chesapeake watershed, there are three major animal production regions with the greatest concentrations of animals: the Lower Susquehanna River in Pennsylvania, the Shenandoah Valley in Virginia and West Virginia, and the Delmarva Peninsula in Delaware, Maryland, and Virginia (Fig. 1). The Delmarva Peninsula is dominated by integrated poultry (mostly broilers) production (Fig. 3).

To determine transport, dispersion, and deposition of emitted ammonia requires air quality modeling. Emitted pollutants in the atmosphere are transported by winds and dispersed by turbulent fluctuations in all directions (Aneja et al., 2001; National Research Council, 2003; Yao et al., 2018). Energy exchanges at the earth's surface influence the planetary boundary layer (PBL) height and turbulent exchanges of momentum, heat and mass (pollutants), thus carrying the pollutants to large horizontal distances and spreading them through the depth of the PBL (Arya, 1999). Model simulated ground -level concentrations (GLC) and deposition of ammonia are analyzed to evaluate their impacts to sensitive ecosystems such as the waterways and the Chesapeake Bay. The objective of this study is to estimate the deposition of ammonia/nitrogen (the "nitrogen" signifies that the parameter is expressed based on mass of N) to the Chesapeake Bay and adjacent lands from poultry animal feeding operations (AFOs) located on the Maryland Eastern Shore (yellow region in Fig. 1).

1.1. Emission factors

The U.S. EPA has been working with Carnegie-Mellon University (CMU) to develop NH_3 emission factors that take into account local meteorological conditions at the county level (these are referred to as EPA/CMU emission factors). More recently, EPA and CMU have produced a Farm Emission Model (FEM), which takes into account meteorological conditions and potential emission control practices, such as the addition of aluminum sulfate to poultry waste (McQuilling and Adams, 2015). Each emission factor or emission model covers three components that contribute to the total emission factor: (1) Confinement refers to the emission from animals residing inside of a Confined Animal Feeding Operation (CAFO) and emission from the waste produced within the contained area; (2) Storage refers to the emission of ammonia/nitrogen from the storage of the waste removed from the CAFO; (3) Land application, as implied, is the emission of ammonia/nitrogen after waste is applied to a field as fertilizer. The total emission factor is the sum of confinement, storage, and land application. We assume that farms in the study region generally do not use waste management amendments, such as aluminum sulfate, and that the farms store and apply waste in the vicinity of the original confinement area.

Ammonia emission factors are subject to considerable variability and uncertainty. Previous studies give emission factors as high as 0.789 kg NH_3 per bird per year (Gates et al., 2005), and as low as 0.035 kg NH_3 per bird per year (Burns et al., 2007). We have adopted an emission factor of 0.20 kg NH_3 per animal per year which is the average annual emission factor developed by CMU and EPA for the counties in the study region. This factor is a composite of emission factors for broiling chickens and laying chickens, with broilers accounting for about 90% of poultry emissions in the region. McQuilling and Adams (2015) have calculated the mean fractional error of the CMU/EPA FEM currently used to estimate animal emission factors, at 69% based on comparison with measurements at broiler operations.

1.2. Fate of atmospheric Ammonia/Nitrogen

At the earth's surface, NH_x (=ammonia (NH_3) + ammonium (NH_4^+)) has a range of beneficial and detrimental consequences for humans and the environment (Tomich et al., 2016; Battye et al., 2017). For example, nitrogen fertilizers have had a beneficial effect on agriculture globally by increasing crop yields. However, the high loading of reactive nitrogen (reactive nitrogen includes all biologically active, chemically reactive, and radiatively active nitrogen compounds in the atmosphere and biosphere of the earth, in contrast to non-reactive gaseous dinitrogen (N_2)), has led to deleterious effects on the environment, such as acidification of soils, forest decline, decreased visibility from increased aerosol production, and elevated nitrogen (both ammonia/nitrogen and oxides of nitrogen (NO_x)) concentrations in ground and surface waters, possibly leading to enhanced eutrophication in downwind ecosystems (Asman et al., 1998; Aneja et al., 1998; Krupa, 2003; Baek and Aneja 2004). Thus, there is a need to study the NH_x deposition changes, spatial distribution, and transport of ammonia from agricultural sources (both crop and animal) to gain a better understanding of effective means to control or reduce excess amounts of ammonia and ammonium deposition.

Any atmospheric ammonia that is not dry deposited or scavenged by raindrops is converted into atmospheric ammonium (Seinfeld and Pandis, 2016). This is done through the interaction of gaseous ammonia with small water particles not large enough to effectively dissolve gaseous ammonia. The conversion of ammonia to atmospheric ammonium (NH_4^+) is important because the ammonium aerosol has a much longer lifetime than ammonia and is an alkaline species that is readily used in the process of $PM_{2.5}$ formation, especially in the presence of sulfuric acid and nitric acid (Jacobson, 1999; Baek and Aneja, 2004; Paulot and Jacob, 2014).

Dry deposition is another process which is important to understand the fate of atmospheric ammonia. Depending on an area's temperature, humidity, and precipitation, dry deposition may be the largest contributor to nitrogen deposition from ammonia releases (Duyzer, 1994). Dry deposition refers to the removal of atmospheric gases or particles without the presence of moisture in the atmosphere. Given that ammonia is highly soluble, it is important to consider dry deposition to both vegetation and to water bodies. Water bodies on which ammonia is deposited can cause dissolution of ammonia and lead to an additional nitrogen deposition mechanism (Larsen et al., 2001). With no natural surface resistance due to the solubility of the species, ammonia uptake by water bodies is efficient and is an important factor in areas where wetlands, rivers, lakes, or other large ocean bodies are present (Larsen et al., 2001). This fact coupled with the concentration

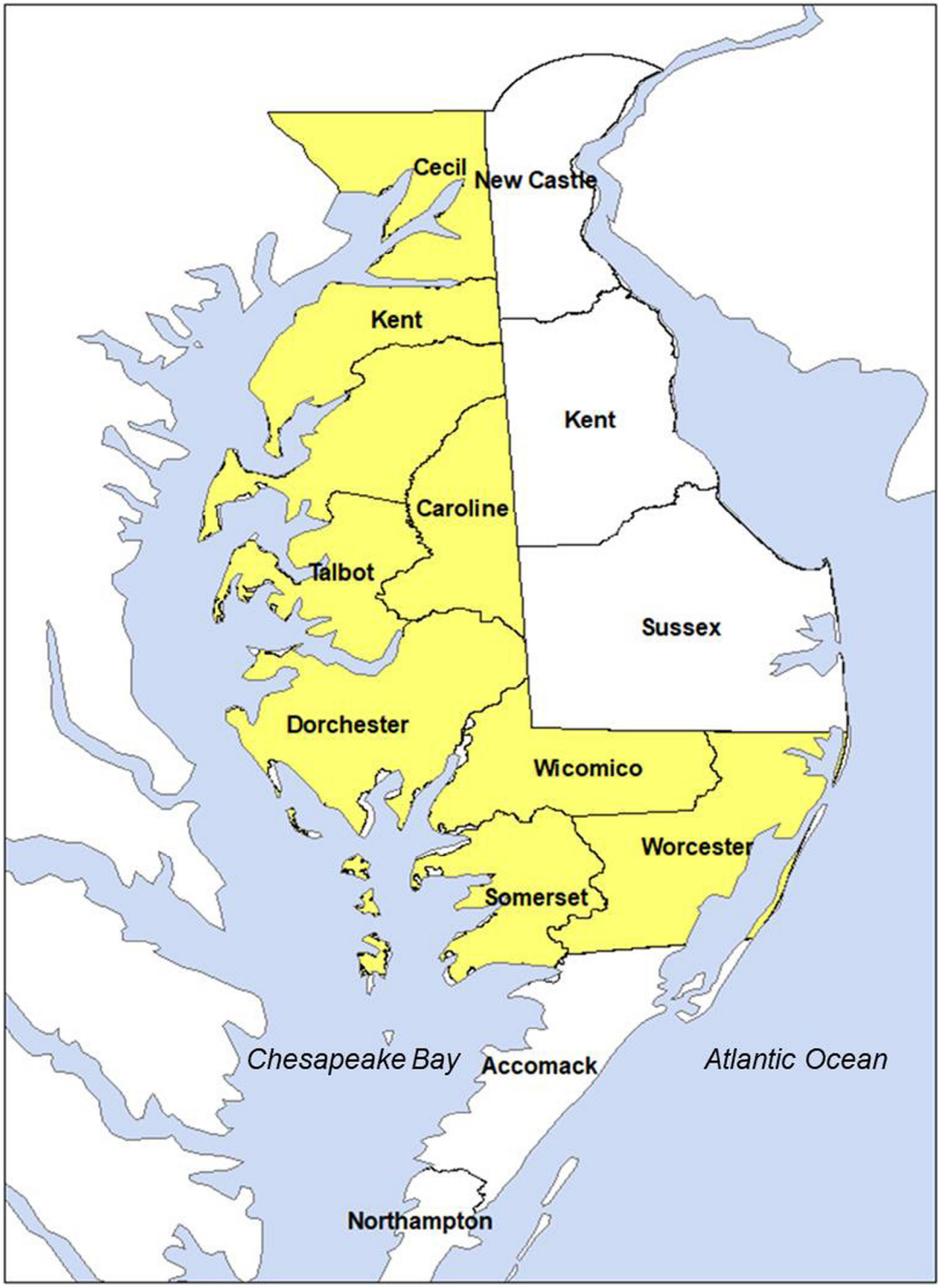

Fig. 1. Map of Maryland, Delaware, and Virginia on Delmarva Peninsula and the Chesapeake Bay. Counties shaded in yellow (only Maryland poultry AFOs) were used in the study. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

of emission sources on the Delmarva Peninsula makes dry deposition a vital topic of this study.

Deposition of ammonia/nitrogen to water and land surfaces with vegetation is expressed using a resistance model approach. When expressing deposition to vegetation, atmospheric gases encounter several factors (resistances) influencing their deposition fluxes. These are aerodynamic resistance (r_a), quasi-laminar resistance (r_b), and surface resistance (r_c). The resistance of gases to transport from the atmosphere to the surface is r_a. Once a gas molecule makes it to the surface for exchange, it must overcome resistances to molecular diffusion across the quasi-laminar boundary layer of air at the leaf surface (r_b) and uptake to the surface (canopy) itself. The r_c is determined by the characteristics of the surface (e.g., presence of moisture, acidity of the surface, leaf stom-

atal processes) to which the gas is depositing. There are separate resistances that make up r_c, which include water resistance, ground resistance, and foliar resistance. Typically, a vegetative canopy exists which involves additional complex resistances, but is usually referred to collectively as the canopy resistance (Seinfeld and Pandis, 2016).

In addition to the resistance model of dry deposition flux, the bi-directional flux of ammonia may have to be considered. When the concentration of ammonia in the atmosphere is higher than the ammonia compensation point at the surface, ammonia will deposit to the vegetation-soil system whereas when the compensation point of ammonia is higher in the soil and vegetation, ammonia will be emitted to the atmosphere (Pleim et al., 2013; Farquhar et al., 1980). In the scope of this study, bi-directional flux was not considered to be important.

1.3. Previous research

This study builds off initial research conducted on the Delmarva Peninsula in 2004 by Siefert et al. (2004). Siefert et al. used inverse modeling to determine the emission strength of the initial poultry operation, while the model used in this study infers the original strength of the emission source from Maryland AFO population data and emission factors from CMU/EPA. O'Shaughnessy and Altmaier (2011) also used inverse modeling using the American Meteorological Society (AMS)/U.S. EPA Regulatory Model (AERMOD). The objective of this study is to simulate the concentrations and deposition at points downwind and later in time assuming the initial strength with emission factors from the U.S. EPA/CMU. Unfortunately, due to the large differences in emission strength, the results of Siefert et al. (2004) and this study are expected to be too different to be compared.

Overall, few studies have attempted to apply AERMOD to horizontal scales of greater than 100 km. The main concern is the application of implied horizontal homogeneity assumption in similarity theories and relations used in AERMOD. However, these assumptions are likely to be valid over Delmarva Peninsula due to its flat terrain. In addition, AERMOD has not been used to simulate the dispersion of atmospheric ammonia, as compared to its applications to other compounds. Theobald et al. (2012) conducted a study utilizing AERMOD's dispersion calculations in the United Kingdom (U.K.) to model ammonia in a rural landscape, locally. That study found that AERMOD shows accuracy despite no inclusion of the bi-directional flux and land-cover data which would influence the transport distances.

It is also important to keep in mind that an important assumption of this study is that no waste management practices or environmental technologies are used to mitigate ammonia emissions throughout the modeling domain, and that the facilities are producing at maximum animal capacity at all times throughout the duration of the simulation. This will provide an upper-limit scenario for ammonia/nitrogen deposition and concentration values.

1.4. Dispersion modeling

AERMOD is EPA's preferred dispersion model for near-field applications, promulgated in 2005 and revised in 2017 (U.S. EPA, 2005, 2017). It is similar to other dispersion models in that they are designed to model the transport of certain chemicals. Initially, the U.S. Military began to experiment with dispersion modeling due to fear of chemical weapons (U.S. EPA, 2013). This led to scientists becoming aware of atmospheric dispersion. At first, gradient transport theories with constant and variable eddy diffusivities were proposed. More sophisticated statistical theories were developed by Taylor (1922). Both horizontal and vertical dispersion were later investigated using the Gaussian and non-Gaussian plume dispersion equations that are utilized in AERMOD (Arya, 1999).

AERMOD uses steady-state plume modeling to calculate concentrations and depositions with the goal of minimizing errors in model output due to small changes in input parameters (U.S. EPA, 2013). The horizontal and vertical concentration distributions are assumed to be Gaussian in the stable boundary layer (SBL) and unlike many dispersion models, it is assumed to be a bi-Gaussian probability density function following statistical concentration distributions in the convective boundary layer (CBL) (Deardorff and Willis, 1985; Briggs, 1993). The general form of concentration distribution in AERMOD within both the SBL and the CBL is:

$$C(x,y,z) = \frac{Q}{U} P_y(y,x) P_z(z,x) \tag{1}$$

where C is the average concentration, Q is emission strength, U is the average wind speed, and P_y and P_z are the probability density functions describing the concentration as a statistical expression away from the model centerline (Peters, 2015). Divisions occur between the CBL, SBL capping the CBL for pollutants emitted by near-surface sources, and the transition between the two. However, most time is spent in the CBL and final concentrations are determined by several forms of dispersion equations describing vertical dispersion, lateral dispersion, and natural centerline fade dispersion (U.S. EPA, 2013). While concentration calculations are at the forefront of the AERMOD formulation, deposition is the most important parameter discussed in this study. Chamberlain (1953) describes the simple deposition model used in AERMOD's formulation involving calculations of ground-level concentrations (GLC) due to a continuous point source:

$$C_0(x,y,0) = \frac{Q_x}{\pi \sigma_y \sigma_z U} \exp\left(-\frac{y^2}{2\sigma_y^2}\right) \exp\left(-\frac{H^2}{2\sigma_z^2}\right) \tag{2}$$

where Q_x is the depleted source strength downwind at a distance of $\times$ and governed by the mass equation:

$$\frac{\partial Q_x}{\partial x} = -\int_{-\infty}^{\infty} F_d(x,y)\partial y \tag{3}$$

And the dry deposition flux:

$$F_d(x,y,0) = v_d C_0(x,y,0) \tag{4}$$

This is termed as a source-depletion model and is a linear relationship allowing deposition to be calculated from GLC calculations with previous determination of SBL and CBL contributions within AERMOD calculations and a prescribed deposition velocity (v_d) (Cimorelli et al., 2005).

Several studies in the past have used AERMOD's dispersion capabilities. Many studies have modeled hydrogen sulfide (H_2S) emissions and dispersion using AERMOD. Some studies have used AERMOD to determine emission factors for better representation of emission from agricultural practices, using an inverse modeling approach. O'Shaughnessy and Altmaier (2011) found that AERMOD worked effectively when using inverse modeling, especially at distances of less than 6,000 m. Other studies have used AERMOD at local scales, but our literature search did not find any AERMOD-related studies that incorporated areas larger than 50 $\times$ 50 km^2. Attempting to apply AERMOD to larger domains makes this study unique. Other studies have successfully applied AERMOD to a local application of ammonia. Briggs, 1993 used AERMOD to determine deposition velocities under different seasons and stability conditions. Deposition velocities were modeled on a local scale and found that total deposition occurred within 2,500 m of the source. Theobald et al. (2012) compared AERMOD, Atmospheric Dispersion Modelling System (ADMS), Local Atmospheric Dispersion and Deposition (LADD), and the Operational Priority Substances model

(OPS-st) in terms of concentration within 1,000 m of a source. Input processes were varied throughout their study which found that for area and volume sources, AERMOD and OPS-st predicted larger concentrations for a case study. Overall AERMOD, ADMS, and OPS-st performed well within the range of acceptability criteria. Hanna et al. (2001) compared ADMS, AERMOD, and Industrial Source Complex (ISC3) at downwind distances of 10 to 20 km. A total of 6 sites were used in their comparison. AERMOD shows better performance at 3 of these 6 sites over ADMS and ISC3.

AERMOD utilizes three forms of meteorological data: (1) site-specific data, i.e. a local meteorological tower, (2) National Weather Service (NWS) or Federal Aviation Administration sites, or (3) prognostic meteorological data. The most readily available data is NWS data, which was used in this study.

AERMOD does not include a system for simulating the conversion of NH_3 to NH_4^+ or the formation of particulate matter from NH_4^+ salts. Therefore, we have only simulated atmospheric concentrations of NH_3 and dry deposition of gaseous NH_3. This neglects the formation of NH_4^+ particulate matter and the potential dry and wet deposition of NH_4^+ particulate matter, and may result in some overestimation of gaseous NH_3 concentration and NH_3 dry deposition. These effects are minor at close proximity to the emission sources, such as where monitor to model comparisons are made in this study.

1.5. Deposition velocities

Schrader and Brümmer (2014) reviewed published ammonia deposition velocities for various land use types and found annual mean values ranging from 0.1 to 1.8 cm/s for semi-natural, 0.4 to 3.0 cm/s for mixed forests, and 0.2 to 7.1 cm/s for agricultural sites. Deposition velocities span more than an order of magnitude within and across land use types. Phillips et al. (2004) conducted their study in an area similar to the Maryland Eastern Shore, which was described as a semi-natural site downwind of the North Carolina State University Research Farm in central North Carolina. Measurements were not taken at the facility but were taken downwind over turf grass. They did not take direct measurements at a CAFO, but several samplers were located downwind (~1 km) of large facilities and in open grass fields.

In general, semi-natural sites have a relatively low deposition velocity with many ranging from about 0.6–1.8 cm/s (Myles et al., 2011). Variation is due to the area of study and time of year. Most studies report deposition velocities during the fall season with some reporting annual means for comparison (Bajwa et al., 2008; Phillips et al., 2004; Myles et al., 2011). Phillips et al. measured a deposition velocity of 2.8 cm/s during the daytime in the fall. Stability, ground temperature, moisture, and other factors may also limit or amplify deposition velocities.

Our main area of interest in the literature review involves agricultural production. In areas downwind of agricultural soils, deposition velocities are expected to be lower than any other type of land types considered unless the measurements are taken downwind of the ammonia source or in areas of intensive agriculture (Schrader and Brummer (2014)). This is reflected in a study done by Myles et al. (2011) which reported a deposition velocity at 7.1 cm/s over a fertilized soil. Other studies such as Baek et al. (2006) found a deposition velocity of 6.3 cm/s downwind fetch of an ammonia source. Studies with deposition velocities below 1 cm/s are likely located within a few hundred to 1,000 m of an ammonia source or in soils with a high ammonia concentration (Bajwa et al., 2008). Theobald et al. (2012) used a deposition velocity of 0.15 cm/s in a study that compares local transport of ammonia within 1 km of a source using different dispersion models. Pleim et al. (2013) provide some reasoning for this with more in-depth analysis provided by Cooter et al. (2010) for agricultural soils specifically. The ammonia/nitrogen bi-directional flux can cause areas of low deposition velocities near ammonia/nitrogen sources. High concentrations tend to increase the surface resistance, which will decrease the effective dry deposition velocity and decrease the overall deposition to an area. Therefore, a constant deposition velocity may not capture the extent of ammonia transport near areas of high concentration such as downwind of intensive animal operations. This difference can be as high as a factor of 10 at the source and a factor of 2, 60 m downwind of the source (Jones et al., 2007). Furthermore, it would be an additional benefit to include variable deposition velocities based on land-use categories. Within the model formulation, the user is allowed to define land-use characteristics in relation to the source. With a large quantity of modeled sources and unknown land-use characteristics of each individual site, the same land-use is assumed for the entirety of the region. Defining land-use at each site individually will improve the quality of modeled transport and provide more detailed surface characteristics that are used in the model output calculation.

2. Methods

2.1. Measurements

Ambient ammonia concentration was measured at 23 sites on Maryland's Eastern Shore during two sampling events of two weeks each i.e. September 8 to 22, 2017 and September 22 to October 6, 2017 (Fig. 3). Data were used to calibrate AERMOD. The CEH Adapted Low-Cost Passive High Absorption (ALPHA) sampler (Fig. 2) (a passive sampler) was used to measure NH_3 in air. The sampler uses a phosphorus acid-coated filter, which serves to capture the ammonia/nitrogen for later analysis in the laboratory. A white PTFE (Teflon) membrane protects the filter whilst allowing gaseous ammonia to diffuse through for capture. The membrane is positioned facing downwards during sampling which prevents rainfall from adversely affecting the measurements. The membrane end of the sampler is sealed with a protective cap whilst not being exposed. The passive sampling system consists of replicate ALPHA samplers attached to a shelter on a pole or post approximately 2.0 m (6.5 ft) above ground. Replicate samplers are used to give a more reliable estimation of the air concentration of ammonia. All samplers were transported in sealed plastic containers for protection.

Phosphorus acid is suitable as an absorbent for temperate climates and is used to coat the ALPHA membranes for all samplers during this measurement campaign. Phosphorus acid is prepared prior to the day of use, but as close to the day of setup as possible. The phosphorus acid coating solution is 50 mL with 52.0 g of phosphorus acid dissolved in 100 mL of deionized water. Laboratory preparation of the coated filters continues with an 8-step process to apply the coating to the filter paper. Step 1 involves the transfer of the coating solution to a small capped bottle. Next, approximately 10 filters at a time are arranged on clean petri dishes. Working as quickly as possible, 40 mL of solution is applied to the center of each filter. After this, the petri dishes containing the filters are placed inside a desiccator. Next, a vacuum pump is connected to the desiccator until each filter appears visibly dry after about 3 min. The coated filters are then ready to be stored until deployment. CEH recommends that, for storage, each filter is stored inside a small petri dish that is sealed and placed inside grip-seal bags and then stored in an airtight container.

In addition to the samplers, 20 travel blanks and multiple laboratory blanks (8 during trip 1 and 6 during trip 2) were used during each trip. Samplers were analyzed after each trip. Unfortunately, 2 travel blanks were discarded during trip 2 due to handling errors in

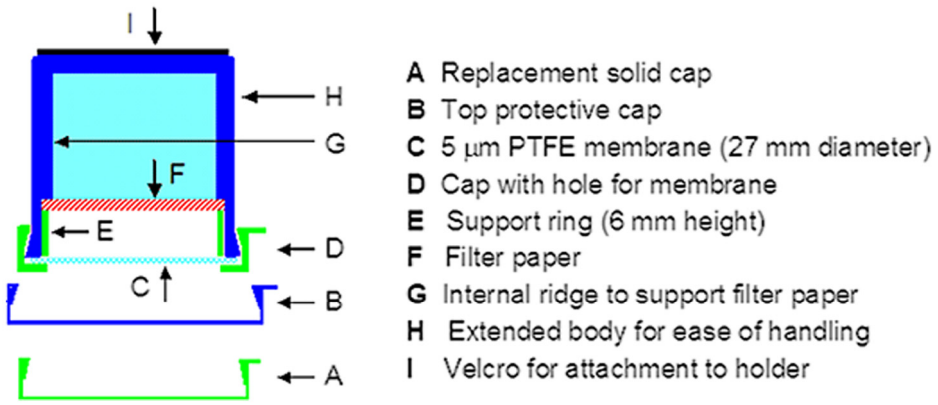

Fig. 2. Outline diagram of a single ALPHA Sampler with its components listed. (). Source: https://www.ceh.ac.uk/services/air-samplers

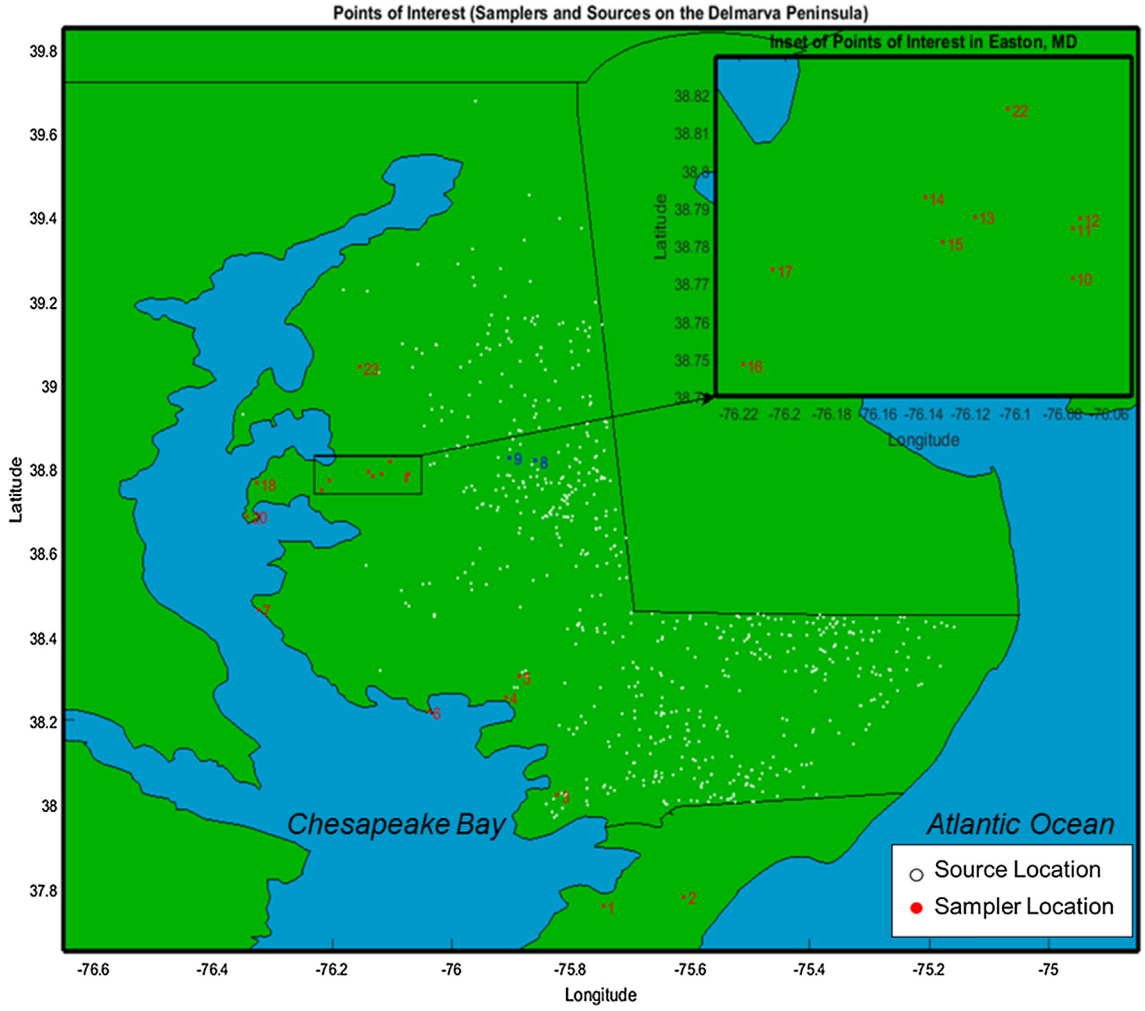

Fig. 3. Domain selected in AERMOD for the model simulation. The extent of the model domain is covered by the borders of the edges of the figure. Red locations are areas where samplers were placed. Their number is listed next to the sampler indicator. An inset is provided for an area near Denton and Easton, Maryland, where multiple samplers were deployed. White dots indicate the locations of poultry operations as of July 2017. There are 603 sources with data provided by MDE. The red star indicates the approximate location of from Salisbury-Ocean City regional airport (KSBY) which is also the station from which meteorology data was used. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

the field. The analysis of the samplers is a straightforward process recommended by the supplier of the samplers (Tang et al., 2001). Analysis is done with the assumptions implied in Fick's Law which states that the diffusion path length is the distance from the mouth of the sampler to the reaction surface at the end of the tube which can be shortened by disturbances such as turbulence. The total measured concentration is a function of this diffusion path length (L, m), the diffusivity of a pollutant (D; for NH_3 = 2.09 × 10^{-5} m^2

s^{-1} at 10), the area of the sampler's measuring surface (A, m^2), and t is time of exposure.

$$V = \frac{DAt}{L}(m^3) \quad (5)$$

From Eq. (5), the effective volume of the sampler is determined, and the final concentration (C) is calculated based on the mass of the pollutant (m_e, μg) minus the mass of pollutant found to be in the travel/laboratory blanks (m_b, μg).

$$C = \frac{(m_e - m_b)}{V}(\mu g m^{-3}) \quad (6)$$

Puchalski et al., (2011) studied passive ammonia monitoring in the United States by deploying passive samplers ALPHA and Radiello® in replicates to determine precision, and collocated with a reference method (annular denuders (URG Corp., Chapel Hill, NC)) to determine the accuracy. The accuracy of the ALPHA and Radiello® samplers, quantified as the median absolute relative percent difference compared to reference methods is 28% and 41% respectively; and the precision between duplicate samplers is 7% and 10% (triplicates) respectively.

2.2. Modeling procedures

AERMOD has distinct advantages over other dispersion models such as the Community Multiscale Air Quality (CMAQ) and ISC3. The CMAQ model does not allow for local scale calculation of concentrations and deposition. CMAQ is required for 3D calculation in as small as ~1 km horizontal spatial resolution which requires a tremendous amount of computing power. Using AERMOD allows us to increase the spatial resolution to as fine as 10 m, which is unprecedented in most air quality models. Additionally, we are able to use real-world meteorological data with AERMOD which reduces model error and increases the confidence in the model.

Uncertainty is introduced in AERMOD due to the spatial availability of meteorological data. Since there is only one meteorological station in our model, this station is used for the entire Maryland Eastern Shore along with Delmarva Virginia and southwestern Delaware, and is maintained by the National Weather Service (NWS). From this station, wind speed, wind direction, temperature, dew point, and pressure are used to define surface parameters for use within AERMOD. Additional limitations of AERMOD include a lack of 3D calculations. GLC and deposition are the only calculations available within AERMOD outputs. Having information on concentrations at a level just above ground-level would give us a more complete picture of ammonia scavenging and particulate matter (PM) formation, however, to an extent, wet deposition gives us the amount of ammonia converted to ammonium and used in PM formation (Walker et al., 2000). Another source of limitation is land-use data. AERMOD does not allow us to distinguish areas of wetland, water, agricultural land, urban areas, and deciduous forest in a deposition post-calculation at individual receptors. This can be done at each source in radial directions, but since the land type at each source location is unknown, the model is forced to view the entire domain as continuous agricultural land. The density of this vegetation is determined by the seasonal classification defined by the user using an assignment of 1 of 5 categories to each of the 12 months. Classifications are 1-summer (lush), 2-autumn (unharvested crops), 3-late autumn (harvested crops, frost, and sparse vegetation), 4-winter (continuous snow cover), and 5-transitional spring (partially green coverage). Elevation data less than 0 m gives the user information regarding areas of water. This limits our ability to calculate direct deposition to tributaries, water bodies, and wetlands and thus, calculated deposition in this study will be conservative.

2.3. Input data processing

AERMOD provides a terrain preprocessor created by the U.S. EPA for AERMOD called AERMAP (U.S. EPA, 2018), that provides information for each receptor and source defined by UTM coordinates in the input file. Elevation data is extracted from the United States Geological Service (USGS) and the National Elevation Dataset. Within the terrain preprocessor, AERMAP calculates both elevation and hill heights needed to account for terrain effects in the AERMOD simulation.

AERMOD also allows the user to process meteorology data from a single site in a program called AERMET. This is a limitation in several situations. AERMOD does not allow the user to input multiple meteorology stations, which causes issues for covering a large domain. Localized meteorological effects such as sea-land breeze will not be calculated within the preprocessor. These are important mesoscale meteorological features that could have a localized effect on certain coastal locations. Despite these limitations, synoptic scale weather patterns will certainly be captured by the model and are perhaps the most important meteorological feature for non-coastal locations which is where most of the sources reside. AERMET requires raw Meteorological Terminal Aviation Routine (METAR) files which limits the selection of locations to those with Automated Surface Observing System/Automated Weather Observing System (ASOS/AWOS) stations. Upper air data is obtained from the nearest site located on Wallops Island in Accomack County, Virginia.

Emission rates from each AFO were determined using an appropriate emission factor for broilers as determined by CMU/EPA emission factors. This includes confinement, storage, and land application components of the total emission factor. The emission factor reported was an average over the lifetime of the activity type. As the bird ages and grows larger, the bird emits more ammonia. Conversely, chicks in their early growth stages emit very little. Thus, an average emission factor was used for the broiler facility. Using CAFO population (activity) data from the United States Department of Agriculture (USDA) and facility specifications from the Maryland Department of the Environment (MDE), an emission rate for a ground-level area source (g m^{-2} s^{-1}) was calculated. Once this emission rate was calculated and applied to the model, AERMOD calculated concentrations and deposition based on the initial parameters of the ground-level source. Emissions were constant throughout the entire modeling period with no temporal variation. It is possible to vary the emissions based on time, but it is likely that timing of house "clean outs" and flock cycles vary operation by operation.

2.4. Single facility

To better understand the deposition distances experienced on a local scale, a single-facility simulation was performed. The goal of this analysis was to quantify how far deposition can occur over a single year from a single facility's emissions. The facility selected for this study is a large facility with 14 houses and a flock capacity of 530,000 birds assumed to be at full capacity year-round. The modeling domain was set with the center of the facility as the center of the domain. On each side of the facility, receptors were programmed to extend 5,000 m east, 5,000 m west, 5,000 m north, and 5,000 m south with a resolution of 100 m. Meteorology data from Salisbury, Maryland, during the year of 2017 was used in the single-facility simulation and the main simulation (described below). The single facility was treated as three sources defined as source clusters: east (containing 5 houses), central (containing 4 houses), and west (containing 5 houses). Google Earth was used to find the angle at which the houses are positioned for realistic simulations.

Deposition velocities were manually input into the model and varied from 0.15 cm/s to 3.0 cm/s to cover the range of possible deposition velocities in this study that would produce similar concentrations to the samplers. This range also accounts for land-use category and soil type. GLC and deposition were model outputs. For the sensitivity analysis, we were primarily concerned with concentration data and its comparison to a total sampler concentration which was found by combining both periods of concentration data. The model was run for 28 days during the sensitivity analysis and 365 days during the full-year deposition analysis. The deposition analysis was done after finding an appropriate deposition velocity for the model

Two comparison statistics were used in this study as suggested by Irwin (2013) and Yu et al. (2006). The statistics were the Mean Normalized Absolute Error (MNAE) and the Normalized Mean Bias (NMB). While MNAE is always positive, NMB can be positive or negative depending on whether the model over-predicts or under-predicts.

2.5. Main simulation

The main simulation shown in Fig. 3 covered the 134 km by 230 km domain. Initial resolution was set to 1 km × 1 km, however computational demands were too high, so a final resolution of 2 km × 2 km was used. A total of 603 CAFOs were modeled using Maryland AFO data provided by the Maryland Department of the Environment. As with the simulation of the single facility, AFOs were assumed to be at full-capacity year-round.

Concentrations were computed within the AERMOD program using the Gaussian plume equations and parameterizations of deposition. Wet and dry deposition were calculated separately. Dry deposition was calculated using dry deposition velocities analyzed from the sensitivity analysis. Wet deposition was calculated using the Henry's Law Constant and diffusivity of air determined by previous studies and those recommended by EPA in their regulatory user's guide for AERMOD (Sander, 2015; Wesely et al., 2002). This estimate can introduce a notable amount of error since the solubility of ammonia will vary temporally based on meteorological conditions and land-use.

3. Results

3.1. Sampling results

Samplers were deployed over two periods lasting two weeks each (September 8–22, 2017; and September 22 to October 6, 2017). The results were combined for model concentrations and are presented in Figs. 4 and 5 for the two separate periods. During the first period, ammonia concentration values ranged from 0.66 ppb to 4.75 ppb. These data show high spatial variability during the first trip with an average value of 1.80 ppb and a standard deviation of 1.22 ppb. The variability is believed to be due to localized sources. Thirteen samplers (#10–22) were placed in suburban/urban areas with high population density. This caused variations in sampled concentrations as an increase in sources due to the application of fertilizer to lawns and additional sources of nitrogen from pets. Some marine and biogenic sources could cause high values as well. Seven samplers (#1–7) were located in more remote areas, but these were also close to the coast. Meteorological factors such as sea breeze circulations, marine clouds, marine fog, and lower temperatures made areas near the coast and water bodies non-representative of a heavy agricultural area in terms of observed ammonia/nitrogen concentration. While all of these samplers give a good representation of the environment on a regional scale, two samplers (#8–9, Fig. 3) were placed in an area of high broiler population density with an observed ammonia concentration of 1.44 (±1sd 0.12 ppb) and 1.26 ppb (±1sd 0.12 ppb), respectively. We believe the results from these samplers serve as the most appropriate to use in our sensitivity analysis because the main source of measurable atmospheric ammonia was likely due to the nearby broiler AFOs. During trip 2, samplers 8 and 9 had similar results with an observed ammonia concentration of 1.65 (±1sd 0.012 ppb) and 1.63 ppb (±1sd 0.012 ppb), respectively. Spatial variability was less during the second interval measurement period with a mean sampler concentration of 1.48 ppb and standard deviation of 0.32 ppb. The similarity in the results from samplers placed in the same area give confidence to the measurements. The average ammonia concentration values for both trips for samplers 8 and 9 were 1.54 and 1.46 ppb (±1sd 0.06 ppb), respectively.

3.2. Sensitivity analysis results

Results of the sensitivity analysis are broken down into two statistical approaches, MNAE (Fig. 6) and NMB (Fig. 7). Fig. 8 provides a visual representation of the relationship between modeled and observed concentrations when the deposition velocity is prescribed as 1.0, 2.2, 2.4, and 2.6 cm/s. When looking at only the data points for sampler 8 and sampler 9, a deposition velocity of 2.4 cm/s shows that measured concentrations have a near 1:1 correlation with modeled concentrations. This area is indicated by a blue box in Fig. 8. The remaining 20 sites do not reside on the 1:1 line. This may be explained because the sites located along the coast are upwind (during the measurement period) of the broiler AFOs of interest (Figs. 4, 5, and 9). With a predominant southwest wind direction (Fig. 9), the source of ammonia concentration at these sites (Figs. 4 and 5) is from the Chesapeake Bay and mainland Virginia (Fig. 9B). Samplers at these sites will not capture the influence of broiler AFOs. However, these sites will represent the region's baseline ammonia concentration. Samplers 8 and 9 are the only two sites that capture the influence of ammonia concentrations from broiler AFOs. A more appropriate location for sampler locations would be in Sussex County, Delaware (Fig. 1) if a future sampling campaign was desired. A plot of MNAE is shown in Fig. 6 and shows the lack of accuracy in samplers excluding samplers 8 and 9. This is likely due to the variability of localized sources in the region that are not included in the model and the influence of predominant wind direction, discussed above. The second statistic to be employed in the analysis of modeled concentrations is NMB. Fig. 7 shows these results for each deposition velocity from 1.0 cm/s to 4.0 cm/s as suggested by a literature review of deposition velocities for semi-natural sites. For this metric, samplers 8 and 9 were the only samplers considered. A low deposition velocity leads to less deposition and higher ground level concentration (GLC). Alternatively, high deposition velocities lead to lower GLC as more deposition occurs. A NMB of ~0 is seen for both a deposition velocity of 2.4 cm/s and 2.5 cm/s. A deposition velocity of 2.4 cm/s has a slightly lower NMB and is supported by Phillips et al. (2004) as discussed in the introduction of this study. The results of MNAE also agree with this conclusion.

An ammonia deposition velocity of 2.4 cm/s gives an estimated total annual ammonia deposition of 11,100 Mg/yr (10,600 Mg/yr deposition to land, and 508 Mg/yr deposition to water (where 1 Mg = 1.1023 U.S. Tons). However, a much smaller ammonia deposition velocity of 0.15 cm/s gives an estimated total annual ammonia deposition of 2,040 Mg/yr (1,880 Mg/yr deposition to land, and 163 Mg/yr deposition to water). Using a deposition velocity of 2.4 cm/s, results from the AERMOD simulation show that 72.4% of nitrogen is deposited due to ammonia/nitrogen release from poultry CAFOs. A comprehensive sensitivity analysis of the

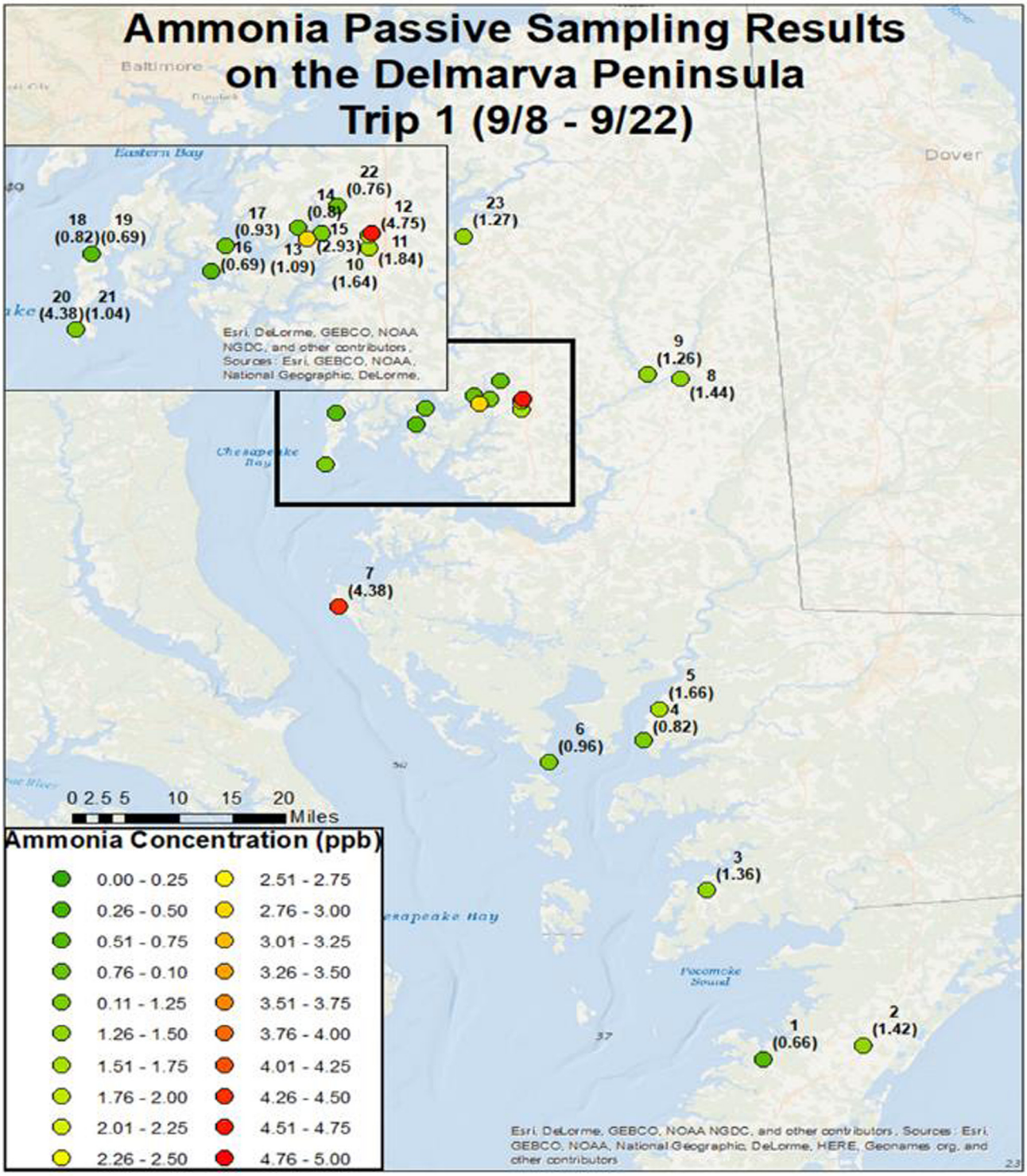

Fig. 4. Sampler results shown for various measurement sites on Delmarva Peninsula with corresponding concentration in units of parts per billion of ammonia/nitrogen (ppb). Colors indicate relative strength of the concentration for Trip 1 (September 8 to 22, 2017). These are combined in the final concentration calculation in the analysis from AERMOD.

effect of ammonia deposition velocity on estimated annual ammonia deposition is provided (Table 1) over the 2 km by 2 km modeling domain covering the Maryland Eastern Shore and Chesapeake Bay. These additional deposition velocities include 1.0 cm/s, 2.0 cm/s, and 3.0 cm/s.

3.3. Simulation results for a single facility

Annual averages were calculated for a single facility in central Maryland Eastern Shore using a modeled deposition velocity of 2.4 cm/s. For this single facility, multiple attributes were

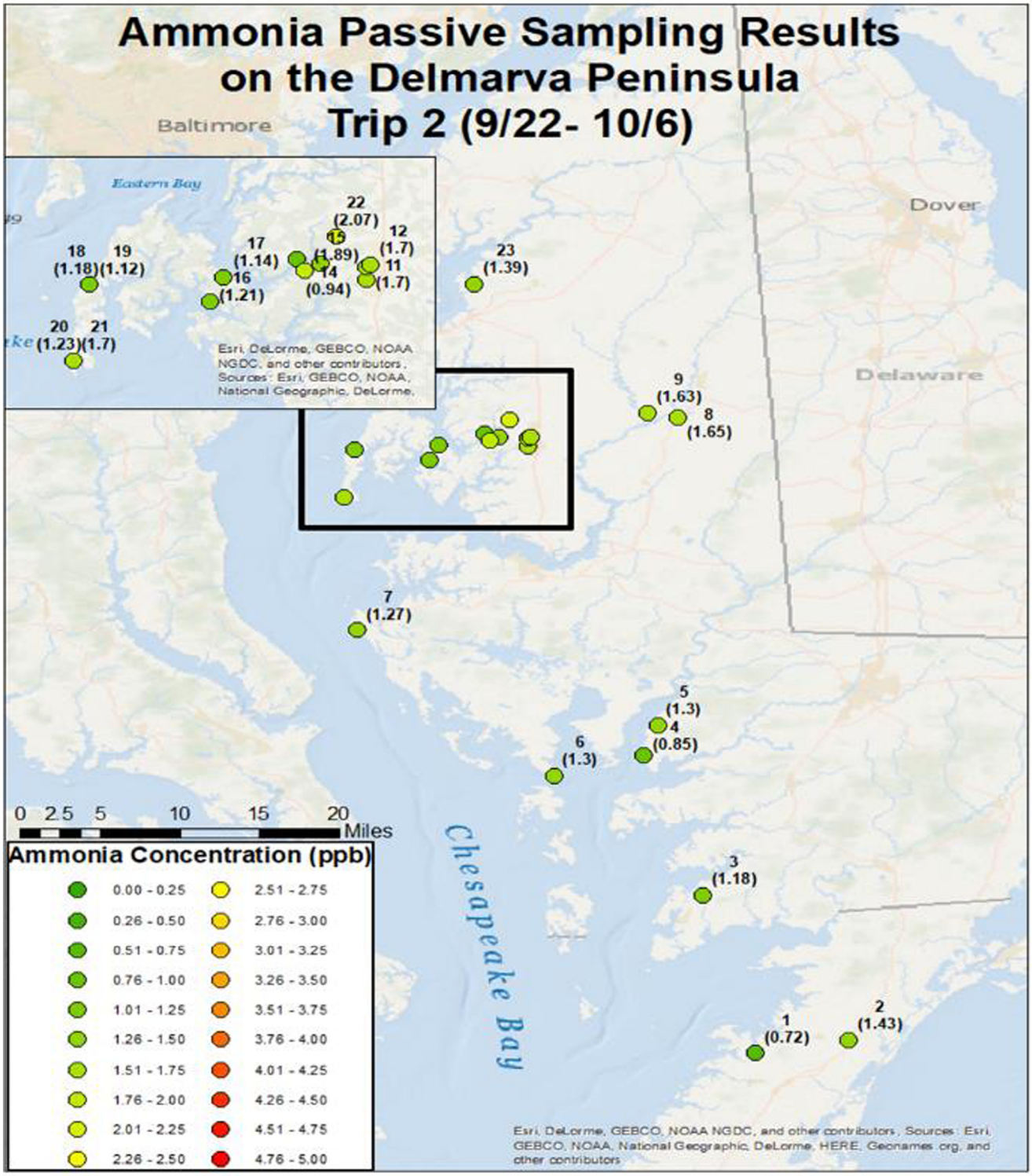

Fig. 5. Sampler results shown for various measurement sites on Delmarva Peninsula with corresponding concentration with units of parts per billion of ammonia/nitrogen (ppb). Colors indicate relative strength of the concentration for Trip 2 (September 22 to October 6, 2017). These are combined in the final concentration calculation in the analysis from AERMOD.

investigated to better understand deposition and concentration. The main area of investigation is deposition as a function of distance from the poultry facility. Results show that for the average meteorological conditions on the Maryland Eastern Shore, and a deposition velocity of 2.4 cm/s, homes and businesses within 2,500 m of the facility will experience average ammonia concentrations of 2.8 $\mu g\ m^{-3}$ (4.0 ppb). Under certain conditions, the short-term concentration can be much higher and above the threshold for human detection (which is approximately 5,000 ppb) of ammonia/nitrogen. While this has no known health effects, it is a significant nuisance for communities near poultry CAFOs (National Research Council, 2003). Concentrations quickly decline from this value to below 1.0 ppb beyond 2,500 m in either direction away from the source facility.

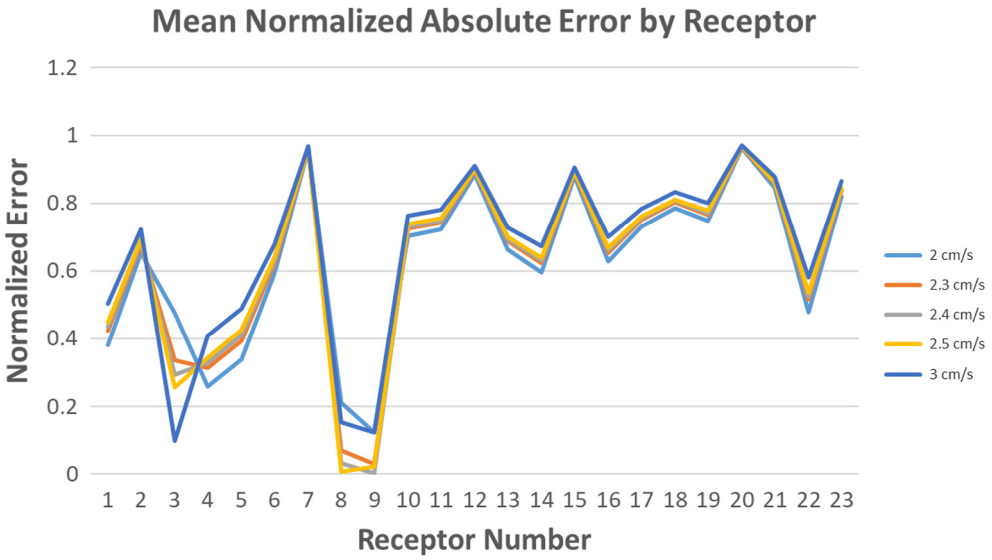

Fig. 6. Mean Normalized Absolute Error (MNAE) on the y-axis for the various receptors, i.e. samplers on the x-axis. Only deposition velocities of 2, 2.3, 2.4, 2.5, and 3 cm/s are considered in the MNAE calculation.

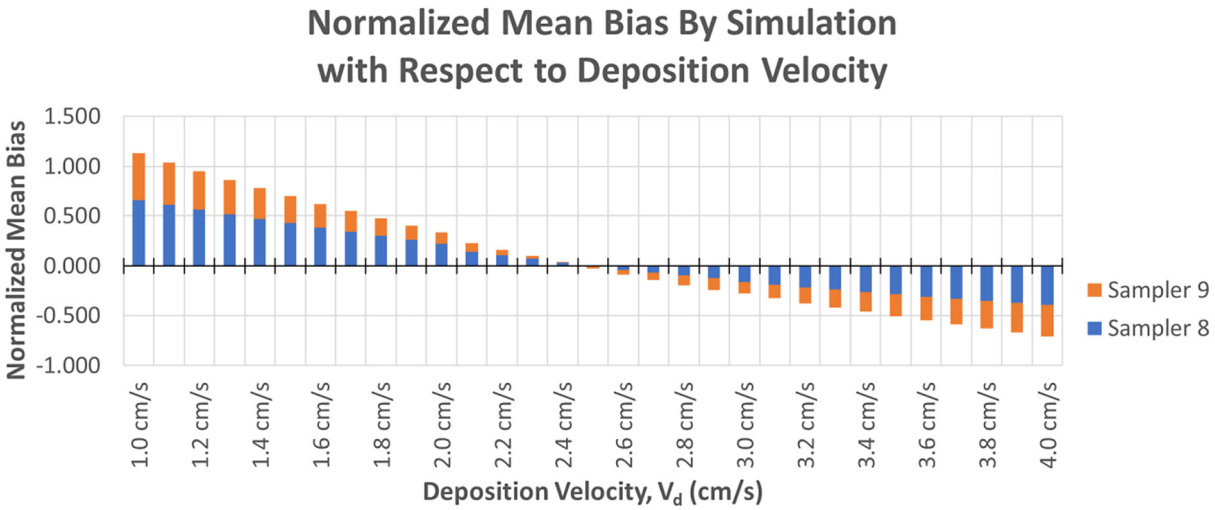

Fig. 7. Normalized Mean Bias (NMB) for deposition velocities 1.0 to 4.0 cm/sec considered during the sensitivity analysis portion of the study. Sampler 8 and sampler 9 were the only samplers considered in the NMB calculation due to other samplers' large error. Positive y-values indicate an over prediction of ammonia by the model while negative numbers indicate an under prediction by the model. The smaller the error, the better the results and a lack of bias in the model.

Approximately 40% of total emissions were found to be deposited within 2,500 m of an AFO source. Fig. 10 shows the cumulative ammonia deposition (% of emission) as a function of distance (m) from the source (for a deposition velocity of 2.4 cm/s) for a single poultry facility. Bajwa et al. (2008) found, on average, that approximately 9% of the total emissions from the source was deposited within 2,500 m of the source. Fig. 11 provides average annual ammonia deposition flux (g $m^{-2}yr^{-1}$) as a function of distance (m) from the source (for a deposition velocity of 2.4 cm/s) for a single poultry facility. Deposition fluxes decrease exponentially from the source as described by the Gaussian plume equation relating concentration and deposition described above in Eqs. (2) and (4). This is an expected result and a function of the model formulation. It is important to note that the model does not incorporate the ammonia bi-directional flux. The highest amount of deposition occurred immediately adjacent to the source where concentrations were at their highest. This is corroborated by Theobald et al. (2012) which found that concentrations will decrease to 1 μg m^{-3} or less at 1,000 m from a ground-level area source.

In the single facility simulation, total deposition within 50 km was found to be about 70% of the total emissions.

3.4. Simulation results for the larger domain

Concentration results (Fig. 12A, B) for a deposition velocity of 0.15 cm/s and 2.4 cm/s show an average ammonia concentration of 1.40 μg m^{-3} and 0.48 μg m^{-3} respectively across the entire modeling domain. As Fig. 12 (A, B) shows, the highest concentrations occur over the Eastern Shore with a minimum in concentration over the Chesapeake Bay. The amount of ammonia/nitrogen reaching the Bay waters is likely higher owing to the deposition to the landmass or other inland water bodies and subsequent transport into the Chesapeake Bay. Unfortunately, determining the deposition to rivers, streams, and tributaries would be very difficult without land-use satellite data. Furthermore, understanding how this ammonia/nitrogen is transported to the Bay waters itself is a separate issue as it is not advised to assume that all the nitrogen from ammonia/nitrogen deposited on land is ultimately transported to

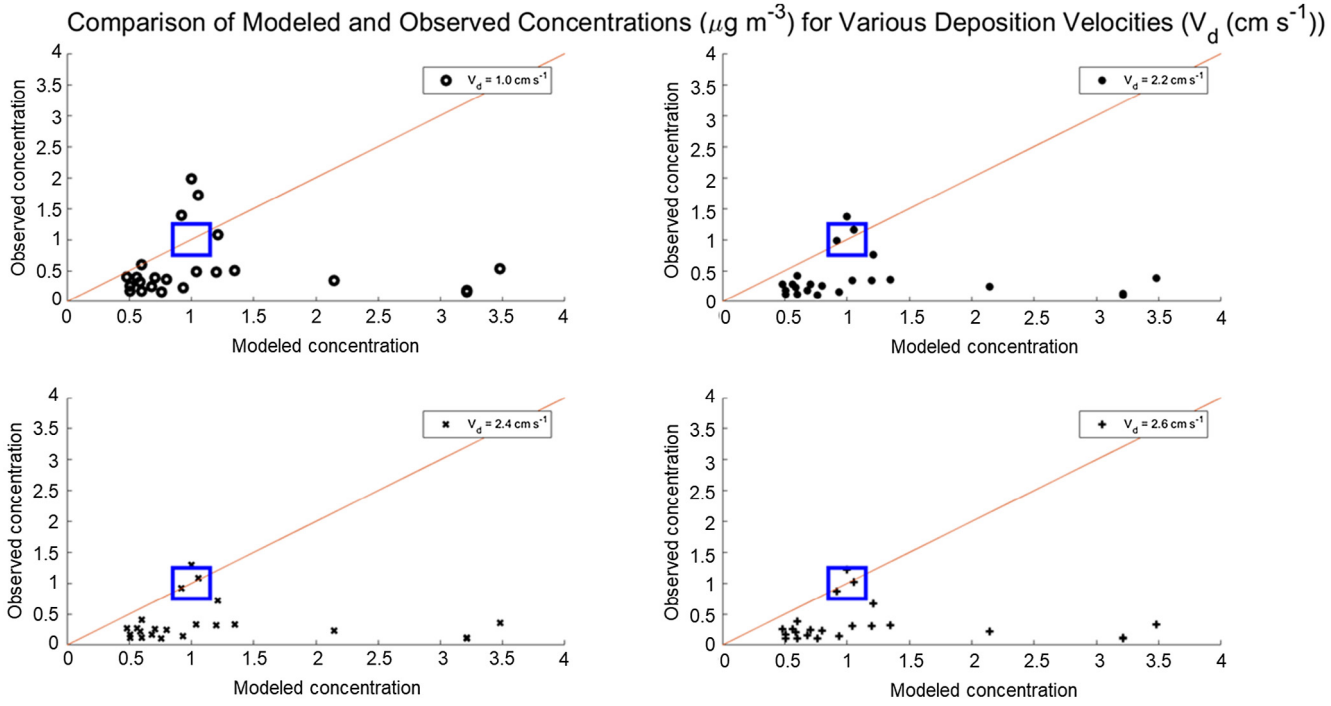

Fig. 8. Comparison of modeled concentration versus observed concentration for the sensitivity analysis. This plot includes all samplers in the model as well as the modeled concentration based on select deposition velocities (1.0, 2.2, 2.4, and 2.6 cm/sec). The 1:1 ratio line is plotted in red to provide the line of best correlation. The blue box represents samplers 8 and 9, i.e. high broiler population density and is the same for each graph. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.) Bajwa et al., 2008.

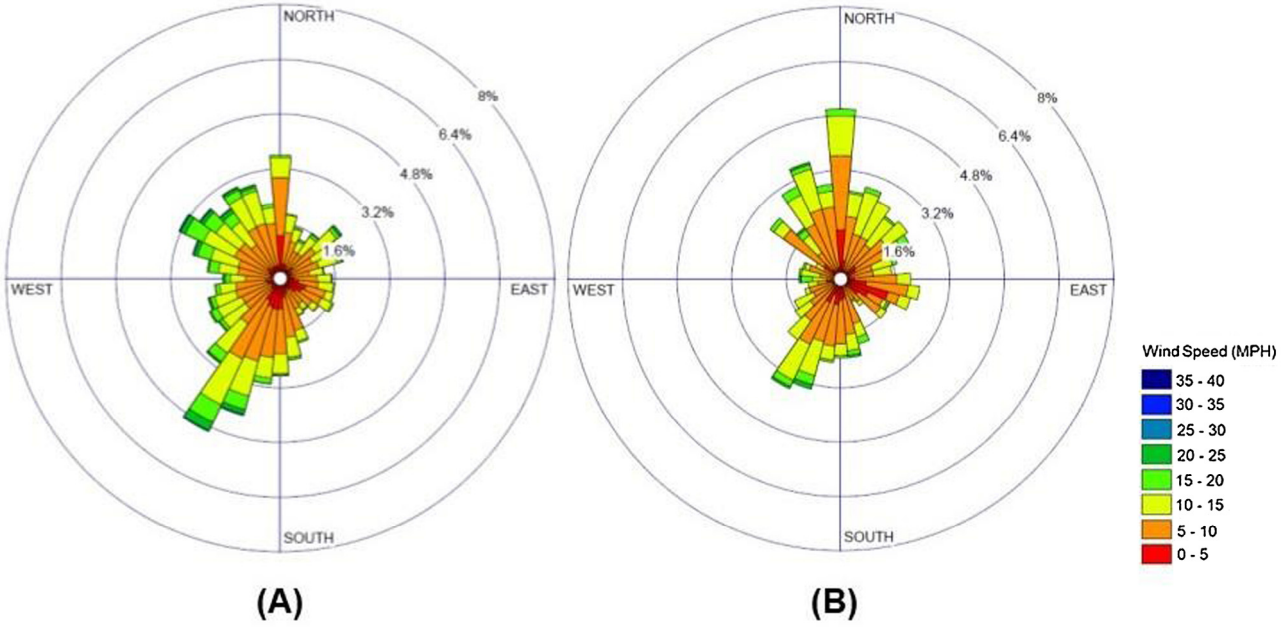

Fig. 9. Wind rose for wind speed and wind direction data used in all simulations associated with this study. Wind direction is noted by the spokes extending from the center of the wind rose. Wind speed is noted by the colors associated with each spoke in miles per hour (mph). Meteorology data is from Salisbury-Ocean City regional airport (KSBY) during (A) January 1 to December 31, 2017; (B) September 1 to October 31, 2017 (measurement period). (). Source: https://mrcc.illinois.edu/

the Chesapeake Bay (Nus and Kenna, 2012). Additionally, meteorological factors such as land-sea breeze would limit transport to the Bay in general. Winds will blow perpendicular to the shore during the day where temperature gradients between the land and the water occur (a common condition met in the area, but not measured in meteorology used in AERMOD simulations). This would protect Bay waters during appropriate atmospheric conditions. Winds from the southwest will enhance this push away from the Bay waters as strong southwesterly winds advect ammonia concentrations toward southwestern Delaware (Fig. 9).

Table 1
Total Estimated Deposition of ammonia to the modeling domain based on AERMOD simulations for a range of deposition velocities 0.15 cm/s to 3.00 cm/s. Column A indicates the simulated deposition velocity for a single AERMOD simulation. Column B indicates the estimated annual deposition in (Mg/yr) that includes deposition during calm conditions (this required an extrapolation of average deposition flux during hours with wind speeds equal to 0 cm/s). Column C indicates the estimated annual deposition as a fraction of emissions. Column D indicates the estimated annual deposition that occurred over the Bay waters (this can be viewed as direct deposition to the Bay water surface). It is assumed that any location within the modeled domain that has an elevation ≤ 0 m is the water surface of the Chesapeake Bay. Column E indicates the estimated annual deposition to the landmass (i.e. deposition to the modeling domain landmass other than the Bay). Column F gives the percent of estimated annual deposition that deposits directly to the Chesapeake Bay water surface based on column D and column B. Column F estimate does not include rivers, marshland, minor tributaries, or other water bodies, or ground water flow to the Bay.

A. Deposition velocity (cm/s)	B. Estimated annual deposition (within the modeling domain) (Mg/yr)	C. Deposition as a fraction of emissions (within the modelling domain) (%)	D. Estimated annual deposition to the Chesapeake Bay (Mg/yr)	E. Estimated annual deposition to the remainder (other than the Bay) of the modeling domain (Mg/yr)	F. Percentage of estimated annual deposition that deposits to the Chesapeake Bay (%)
0.15	2,040	13.4	163	1,880	7.97
1.00	7,400	48.4	401	7,000	5.42
2.00	10,260	67.0	486	9,770	4.73
2.40	11,100	72.4	508	10,600	4.58
3.00	12,100	79.2	531	11,600	4.37

(1 Mg = 10^6 g = 1.1023 U.S. Tons).

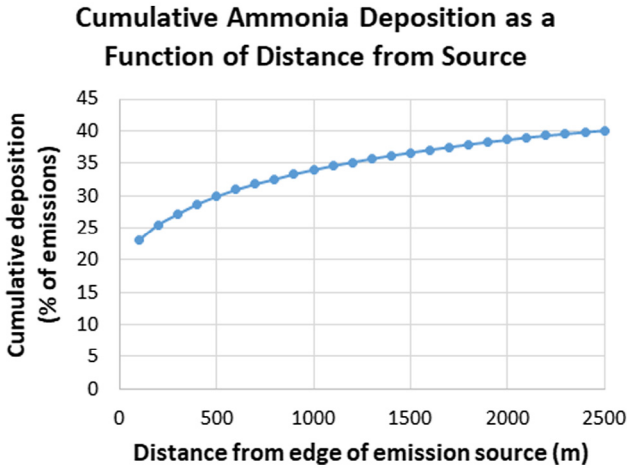

Fig. 10. Cumulative ammonia deposition (% of emission) as a function of distance (m) from a source (for a deposition velocity of 2.4 cm/s), for a single poultry facility.

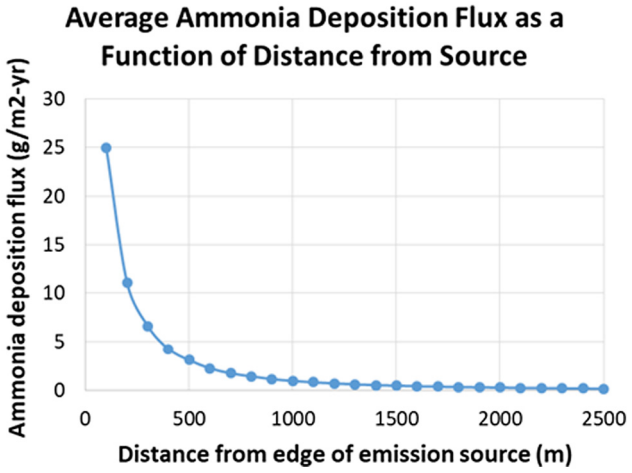

Fig. 11. Average annual ammonia deposition flux (g $m^{-2}yr^{-1}$) as a function of distance (m) from an AFO source (for a deposition velocity of 2.4 cm/s), for a single poultry facility.

AERMOD reports average deposition fluxes for each receptor within the modeling domain. The use of a constant Vd implies a linear relationship between flux and concentration.

Because of the linear relationship between deposition flux and concentration, the spatial patterns of deposition are similar to the spatial pattern of concentration. Using a deposition velocity of 0.15 cm/s and 2.4 cm/s (Fig. 12 C, D) provides annual average deposition flux (including both dry and wet deposition) over the course of a single year from poultry AFOs on the Maryland Eastern Shore. Deposition fluxes are calculated hourly and averaged over the entirety of the modeling period and reported as an average deposition flux. Average deposition fluxes show that throughout the year with meteorological observations and a deposition velocity of 0.15 cm/s, deposition over the modeling domain is calculated to be approximately 2,044 Mg (2,252 U.S. Tons); and for a deposition velocity of 2.4 cm/s, deposition over the modeling domain is calculated to be approximately 11,086 Mg (12,220 U.S. Tons). Overall emissions totaled to 15,345 Mg (16,914 U.S. Tons). Fig. 13 shows the impact of increasing deposition flux on the domain-wide deposition as a fraction of emissions. Using a deposition velocity of 2.4 cm/s, the fractional deposition was calculated as ~72% within the modeling domain. Moreover, modeling suggests that for a deposition velocity of 2.4 cm/sec for a single poultry facility, 30% of emissions will be deposited ~500 m distance and ~38% of emissions will be deposited ~2,000 m from the source (Fig. 10). Walker et al. (2008) found that about 10% of the emitted ammonia from a swine production facility deposited to the surface within about 500 m of the source. Fowler et al. (1998) found that about 3–10% of the locally emitted ammonia will deposit back locally. Asman (1998) incorporates much of the improvement in understanding of NH_3, emission, transport and deposition over the last two decades and shows that up to 60% of the NH_3, emitted from sources up to 3 m in height, may be deposited within ~2000 m of the source. Using a regional chemical transport model, Dennis et al. (2010) found that a fractional deposition of around 8–15% of total emissions will occur within 12 km of a source facility.

Linker et al. (2013) estimated roughly 2,830 Mg of ammonia/nitrogen was directly deposited to the Chesapeake Bay's tidal surface waters. By comparison, we estimated total ammonia deposition per year to the Chesapeake Bay is approximately ~508 Mg (418 Mg of nitrogen) using a deposition velocity of 2.4 cm/s. Worth noting is that our source inventory was limited to Maryland poultry AFOs and the mainstem of the Chesapeake Bay, whereas Linker et al. (2013) included all animal sources within the watershed and all tidal waters.

A comprehensive sensitivity analysis of ammonia deposition velocity on estimated annual ammonia deposition is provided (Table 1) over the regional modeling domain covering the Maryland Eastern Shore and Chesapeake Bay. This indicates that ~5% of the total deposition that occurs within the domain is depositing

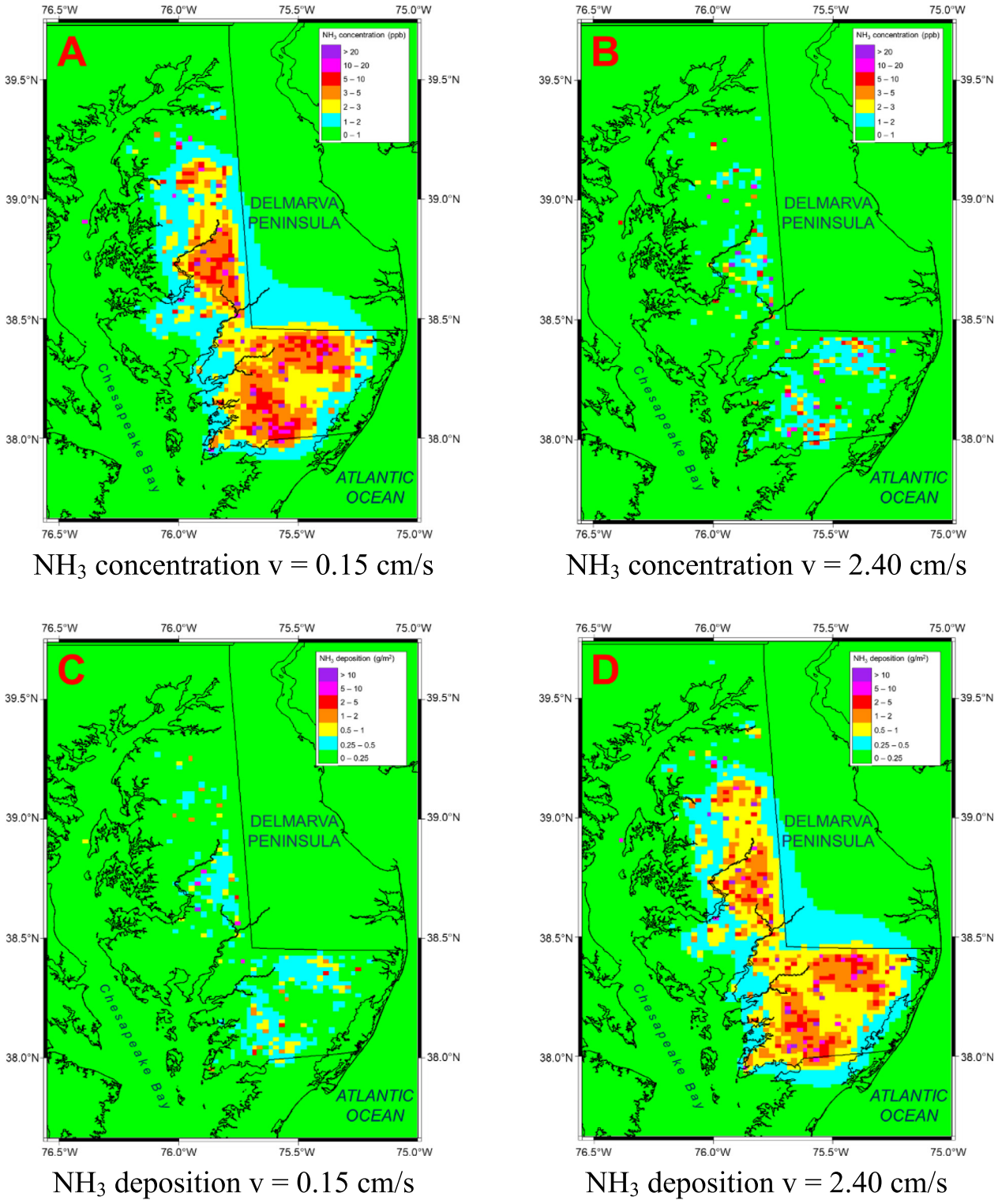

Fig. 12. Average ammonia concentration (A: deposition velocity of 0.15 cm/s; B: deposition velocity of 2.4 cm/s) over the Delmarva Peninsula (from only Maryland poultry AFOs) during 2017. The maximum ammonia concentration is 166 ppb (239 μg m^{-3}) for Vd 0.15 cm/s; and 148 ppb (216 μg m^{-3}) for Vd 2.4 cm/s. Annual deposition of ammonia (including both dry and wet forms of ammonia) over the Delmarva Peninsula (from only Maryland poultry AFOs) during the 2017 (C: 2044 × 10^6 g/year for Vd 0.15 cm/s); and D: 11,087 × 10^6 g/year for Vd 2.4 cm/s).

to the Chesapeake Bay waters directly. Moreover, this does not include the additional input from indirect deposition to rivers, streams, and groundwater which will likely transport to the Chesapeake Bay. It is important to note that all poultry houses are assumed to be at capacity year-round with constant emissions. Emission factors also introduce error into the model as they can vary based on waste management practice, weather, and poultry growth state. Finally, we did not assume any facilities were using waste amendments, such as aluminum sulfate, to control ammonia emissions.

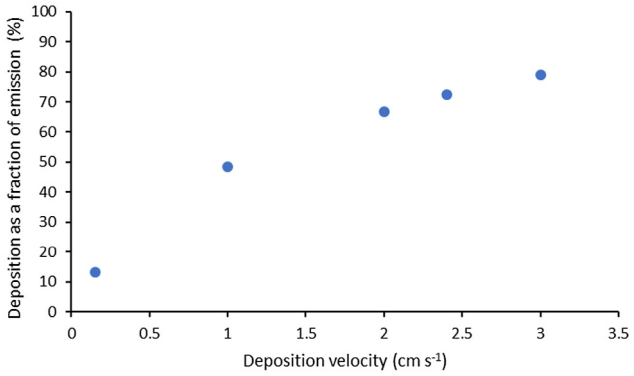

Fig. 13. Ammonia deposition as a fraction of emissions versus deposition velocity. (1 Mg = 1.1023 U.S. Tons).

Meteorological effects will have a significant impact on the deposition (both wet and dry) over the domain. The most critical of these meteorological parameters affecting atmospheric dispersion and deposition are wind speed, wind direction, and stability (Arya, 1999). Fig. 9 A shows the wind rose of the meteorology (wind speed and direction) used in the main simulation. A predominant wind from the southwest is seen approximately 5% of the time. This will transport ammonia away from the Chesapeake Bay, and cause higher concentrations to exist over the terrestrial surface northeast of the concentration of sources. This is similar to wind rose during the measurement period (Fig. 9 B). The second most common wind direction is from the north/northwest. More stable conditions at night will tend to increase concentrations at the surface and lead to more deposition at this time of day (Arya, 1999). Unstable conditions will allow the plume to disperse more effectively and lead to low concentrations (Arya, 1999).

4. Conclusions

This analysis is a combination of measurement and modeling of ammonia concentration/deposition to the Maryland Eastern Shore land and the Chesapeake Bay from poultry operations over the Delmarva Peninsula. The application of AERMOD to estimate fate and transport of ammonia from poultry operations has promise. The model was able to reliably predict ammonia concentrations from sites (samplers 8 and 9) that were closest to the source has proven to be accurate in predicting concentrations when validating with meteorology and sampling results close to the source (Figs. 3, 7 and 8). However, AERMOD's concentration predicting capability decreases when applied to a regional scale. AERMOD's ability to predict concentration drastically improved when considering sampled concentrations near the source cluster. Fig. 6 shows the mean bias applied to only samplers 8 and 9 and reports mean bias near 0 for a deposition velocity of 2.4 cm/sec. This is an encouraging result when applying the model to localized areas. Significantly higher mean biases in samplers at large distances from the source region are likely due to localized sources and the location of samplers being upwind of the largest cluster of poultry AFOs (Fig. 7).

Direct annual deposition to the Chesapeake Bay is estimated to range from ~163 Mg (180 U.S. Tons) for a deposition velocity of 0.15 cm/s to ~508 Mg (560 U.S. Tons) for a deposition velocity of 2.4 cm/s. These values, especially the estimate using the 2.4 cm/s deposition velocity, are within the range of Linker et al. (2013) who estimated roughly 2,830 Mg of nitrogen in the form of ammonia was directly deposited to the Chesapeake Bay's tidal surface waters. However, it is known that AERMOD is unable to calculate mesoscale meteorological features without being provided with appropriate weather data. Location of weather data used for this study was limited to an area in the center of the peninsula. In areas near the coast, sea breezes and other weather features of the marine environment will likely affect deposition calculations to the Bay. During the daytime, winds blowing inland will likely limit deposition to the Bay, but some conditions such as marine instabilities during the fall and early winter could exist to significantly increase deposition to the Bay surface. From this study, it is clear that direct deposition of ammonia/nitrogen to the Chesapeake Bay is less than the deposition to land, rivers and tributaries within the watershed.

A single facility analysis was performed using a deposition velocity of 2.4 cm/s; which was determined from a sensitivity analysis of measured concentrations in an attempt to determine transport distances of ammonia from broiler CAFOs (Fig. 10). We estimate that approximately 40% of the ammonia/nitrogen deposition occurs within 2,500 m of the source.

Overall, the emissions from poultry totaled to 15,345 Mg/yr. When using a deposition velocity of 2.4 cm/s, deposition over the modeling domain is calculated to be approximately 11,086 Mg/yr. This result is consistent with previous studies (Linker et al, 2013). However, it is interesting to compare and contrast these results for a lower deposition velocity e.g. deposition velocity of 0.15 cm/s. The deposition to the modeling domain is estimated to be around 2,044 Mg/yr. Indirect deposition due only to broiler CAFOs to the Chesapeake Bay remains unknown. Of the emitted ammonia, 13% is deposited back to the domain (using a deposition velocity of 0.15 cm/s); while ~72% is deposited back to the domain (using a deposition velocity of 2.4 cm/s). With nearly ~90% of the modeled deposition settling to the landmass, indirect deposition will clearly provide the largest proportion of deposition to the Chesapeake Bay from river transport. Unfortunately, AERMOD does not allow users to get a specific land-use data set to be used in the analysis phase of the output. Additionally, vegetation is an important consideration of this study. Dense forests will likely limit direct deposition to the Bay by taking up ammonia that would otherwise deposit to the water surface. These dense forests are near rivers and water bodies and may further limit deposition to the Chesapeake Bay. Moreover, use of Best Management Practice (BMP) of using aluminum sulfate in the poultry houses for reducing ammonia emissions was not accounted for.

Poultry CAFOs were assumed to be at capacity during the duration of the model simulation. This is not a realistic approach, since it is difficult to model the temporal emissions from a single facility for 603 separate facilities. Modeling scenarios, however, could be improved in several ways. For future research, it is suggested that the simulation is run for a single growing cycle rather than an entire annual rotation. Additionally, seasonal variation, particularly in deposition velocities, is an important variable to include. Second, we assumed all AFOs were at capacity during the model simulation. More realistic estimates would be achieved if simulations reflected the growing cycle of the birds. Third, an estimate of the number of facilities that use waste amendments to control ammonia would improve model accuracy. Lastly, additional monitoring data would allow for better model calibration. This should address seasonal changes in emissions as well as deposition velocities. Regardless of whether realistic estimates of deposition to the Chesapeake Bay can be produced exactly in a model environment, an increase in emission will lead to an increase in deposition. Therefore, it is increasingly important to understand the effects of ammonia/nitrogen-nitrogen deposition to the Chesapeake Bay area both as the DELMARVA Peninsula experiences growth and the construction of new sources of ammonia continues.

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