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**ESTIMATING AMMONIA EMISSION FLOW BY THE IMPLEMENTATION OF REVERSE
MODELLING**

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Abstract: This study represents an air quality modelling application to experimental data in order to evaluate ammonia emission in atmosphere from agricultural activities. The objective of this study is to estimate ammonia emission flow in atmosphere caused by slurry application; two techniques are compared: band spreading on soil surface and injection (closed slot). Reverse modelling method, described in UNI-EN 15445, is applied by implementing two different dispersion models, AERMOD and WINDTRAX and ammonia losses are evaluated through linear regression between models output and field measurements of NH₃ concentration carried out by ARPA Lombardia in two farms located in Regione Lombardia. Both the models implemented identify the trend of measured data and calculate a lower emission flow during injection than during band spreading on soil surface. Correlation coefficient R² values indicate the accuracy of the procedure implemented. Nevertheless, the values obtained refer to meteorological conditions and NH₃ concentration data of specific cases of study.

Key words: *Reverse modelling, dispersion model, ammonia, manure management, emission flow*

INTRODUCTION

Air quality dispersion models allow to simulate pollutants' dispersion and reaction in atmosphere. Reverse modelling is a procedure widely applied in scientific literature which consists in the use of a local dispersion model to calibrate the value of emissions produced by specific sources or to determine the value of emission factors that can be also used in other contexts. In this study, part of a project required by Regione Lombardia, reverse modelling is applied to estimate emission flows in atmosphere caused by typical sources that characterised a farm, such as housing, storage and spreading areas, starting from experimental data. Agricultural sector has relevant effects on air quality and on ammonia emissions in atmosphere. The Regione Lombardia emission inventory, INEMAR, updated to 2017 (INEMAR, 2020), assumes that the 97% of regional ammonia emissions are due to fertilisation and manure management; the latter, in particular, accounts for the 86% of total emission in 2017. Moreover, NH₃, with NO_x, plays an important role in the formation of secondary fine particulate matter (Angelino et al., 2013). Main variables that influence ammonia emissions from agricultural sources refer to composition of food ration, livestock characteristics, the amount of excreted nitrogen, housing, storage, spreading and treatment technologies, type and quantity of manure and meteorological conditions.

MATERIALS AND METHODS

This study applies reverse modelling for the estimation of ammonia emission flows. Reverse modelling is a procedure defined by the European regulation, adopted in Italy, UNI EN 15445:2008 – Fugitive and diffusive emissions of common concern to industry sector – Qualification of fugitive dust sources by Reverse Dispersion Modelling. This standard illustrates a method to quantify the fugitive emission rates of diffuse fine and coarse dust sources of industrial plants or areas. The implementation of the procedure involves several steps. Firstly, emissive areas, measuring points and receptors are identified and georeferenced, a hypothetical value of emission flow is set and meteorological parameters are defined. These data are given in input to an air quality dispersion model that calculates ammonia concentration in each receptor. Finally, least squares regression between concentrations estimated by the model and measured concentrations is applied to obtain an optimized value of NH₃ emission flow. Air quality dispersion models used in this work are AERMOD (EPA, 2018) and Windtrax (Crenna, 2016a and Crenna,

2016b). Meteorological data given in input to the models are calculated every day by the air quality diagnostic modelling system used in ARPA Lombardia characterised by a domain of 236 x 244 km² set on Regione Lombardia and covered by a grid of 4 km step and 13 vertical levels between 10 and 6000 m. This system communicates with the database, owned by ARPA Lombardia, containing data collected by air quality and meteo-hydrological monitoring grid. The meteorological input is obtained by assimilating the hourly parameters, from a subset of stations of the local network and from radio sounding carried out in Linate through a mass-consistent model, to meteorological fields produced by the European Centre for Medium-Range Weather Forecast, ECMWF (Silibello et al., 2007). The atmospheric turbulence parameters are then estimated with the SurfPRO processor (Silibello et al., 2007).

Aermod

Aermod is a steady-state plume model, approved by US Environmental Protection Agency, that incorporates air dispersion based on planetary boundary layer turbulence structure and scaling concept, including treatment of both surface and elevated sources and both simple and complex terrain (<https://www.epa.gov/scram/air-quality-dispersion-modeling-preferred-and-recommended-models#aermod>). Main data provided in input to the model refers to georeferencing and emissive property of sources, location of measuring points and receptors grid, period of simulation and meteorological fields, obtained as illustrate in previous paragraph. Aermod gives in output hourly ammonia concentration in each receptor.

Windtrax

Windtrax (Crenna, 2016a) is an air quality dispersion model that simulates the transport of pollutants in the atmosphere surface layer and calculates emission rates or pollutants concentrations near to emissive sources. The model can be applied if surface around the sources and the sensors is flat and free of obstructions and if the maximum distance between them is one kilometre. Parameters required in input concern the characterization of emission sources, the location of receptors at measuring points and the definition of meteorological parameters, whose values are obtained as previously illustrated. Since data required by Windtrax are hourly average (the model is not time dependent, therefore a meteorological field including parameters representing all simulation period cannot be provided as input), as many simulations as the hours in simulation period are carried out, obtaining an emission flow for each hour of simulation.

Least square regression between NH₃ concentrations estimated by the models and concentrations detected in measuring points is implemented in order to estimate ammonia emission flow.

Emission flows deriving from Windtrax application are calculated implementing regression between Windtrax concentration and Aermod concentrations instead of measured data because Windtrax provides only hourly ammonia levels, while measured data refer to the whole simulation period.

Case study

In this work reverse modelling is applied to two different farms, located in Regione Lombardia, where ARPA Lombardia carried out a monitoring campaign to measure ammonia concentration during slurry application (D'Angelo et al., 2020). In the first one, a cattle farm, the main emission sources are the fields where slurry is spread. Four scenarios, corresponding to distribution period, are analysed. For the second farm, a swine breeding, a scenario, corresponding to injection carried out in May 2019, is simulated. In the farm and in the surrounding context, different emissive sources are identified (Figure 1): two housing areas hosting cattle (B1 and B2); a housing area hosting swine (P1); three storage (S1, S2 and S3); a spreading area (D1) and two linear sources, a highway (L2) and an extra-urban road (L1), divided into three sub-sources. In conclusion 5 scenarios are analysed: 1-cattle farm, injection, May 2018; 2-cattle farm, band spreading, May 2018; 3-cattle farm, injection, September 2018; 4-cattle farm, band spreading, September 2018; 5-swinr farm, injection, March 2019.

RISULTS AND DISCUSSION

In Table 1 ammonia emission flows obtained by the implementation of reverse modelling using Aermod as air quality dispersion model are shown. These values correspond to NH₃ concentrations, measured by

ARPA Lombardia during slurry application. The correlation coefficient R2, whose values are always major of 0.6, proves the accuracy of the procedure implemented.

Table 1. NH₃ emission flows obtained by reverse modelling using Aermod in five scenarios analysed.

Scenario	Farm	Emissive source	Emission flow ($\mu\text{g m}^{-2} \text{s}^{-1}$)	R2	Scenario	Farm	Emissive source	Emission flow ($\mu\text{g m}^{-2} \text{s}^{-1}$)	R2
1	Cattle	Field	40.20	0.63	5	Swine	Storage (S3)	37.64	0.81
2	Cattle	Field	232.00	0.73	5	Swine	Field (D1)	3.57	0.81
3	Cattle	Field	62.90	0.98	5	Swine	Road (L1-1)	16.67	0.81
4	Cattle	Field	110.00	0.65	5	Swine	Road (L1-2)	0.00	0.81
5	Swine	Cattle housing (B1)	80.64	0.81	5	Swine	Road (L1-3)	898.12	0.81
5	Swine	Cattle housing (B2)	186.4	0.81	5	Swine	Highway (L2-1)	32.22	0.81
5	Swine	Swine housing (P1)	74.44	0.81	5	Swine	Highway (L2-2)	63.44	0.81
5	Swine	Storage (S1)	75.38	0.81	5	Swine	Highway (L2-3)	0.00	0.81
5	Swine	Storage (S2)	26.24	0.81					

In Table 2 ammonia emission flows calculated by applying Windtrax in reverse modelling procedure referred to cattle farm are illustrated. These results are comparable and generally of the same size as values estimated by Aermod. Nevertheless, Windtrax appears less accurate than Aermod: R2 assumes some values less than 0.6, since it varies from a minimum of 0.27 to a maximum of 0.98. In this case, each scenario is associated with a variability range of R2 and not a single value because as many simulations are carried out as many hours of simulation period.

Emission flows related to slurry application, obtained by the implementation of the two models, are comparable to values reported in scientific literature. Carozzi et al. (2012) estimate an ammonia emission flow equal to $163 \mu\text{g m}^{-2} \text{s}^{-1}$ by using Windtrax in case of slurry band spreading on soil surface.

Table 2. NH₃ emission flow obtained by reverse modelling using Windtrax in four scenarios of cattle farm.

Scenario	Farm	Emissive source	Emission flow ($\mu\text{g m}^{-2} \text{s}^{-1}$)	R2	Scenario	Farm	Emissive source	Emission flow ($\mu\text{g m}^{-2} \text{s}^{-1}$)	R2
1	Cattle	Field	11.90	0.38-0.58	3	Cattle	Field	27.53	0.27-0.41
2	Cattle	Field	105.00	0.72-0.98	4	Cattle	Field	67.80	0.60-0.84

In Figure 1, maps illustrate, for different scenarios analysed, ammonia mean concentration on simulation period estimated by Aermod, when optimised emission flow is given in input to the model. Maps do not represent Windtrax outputs since it provides less accurate values. In the maps, higher values are detected in the areas above the identified emission sources and the plume follows the prevailing direction in which wind blows. Moreover, greater concentrations arise from band spreading on soil surface than injection. This trend is confirmed also by emission flows (Table 1 and Table 2): results detect a lower emission flow when slurry is applied by injection (in the order of 10^{-5} or $10^{-6} \text{g m}^{-2} \text{s}^{-1}$, compared to $10^{-4} \text{g m}^{-2} \text{s}^{-1}$ corresponding to band spreading). For example, in cattle farm, emission flow associated to injection is lower of 80% (in May 2018) and of 40% (in September 2018) than the one estimated during band spreading. This difference is confirmed by scientific literature: UN-ECE (2014) calculates a reduction of more than 60% in case of injection. In scenario 5, NH₃ estimated by the model is greater than the values calculated in scenario 1 and 3 because of plurality of emissive sources located in the farm and in the surrounding context.

Results calculated by the implementation of reverse modelling are affected by spatial disaggregation of emissive sources. In Figure 2 ammonia concentrations maps obtained by simulating two different scenarios of swine farm are shown: in the first one (map on the left) different emissive sources related to housing, storage and distribution areas are identified; while in the second one (map on the right) only one emissive source including all housing, storage and distribution areas, is considered. The comparison of two maps illustrates that ammonia concentration are distributed more uniformly throughout the territory when only one source is identified; while, when emissive sources are disaggregated, NH₃ is more concentrated over the located areas and reaches higher values. In both cases, however, the plume follows the prevailing direction in which wind blows. The graphic in Figure 2 compares ammonia concentrations estimated in two scenarios along the direction in which wind blows.

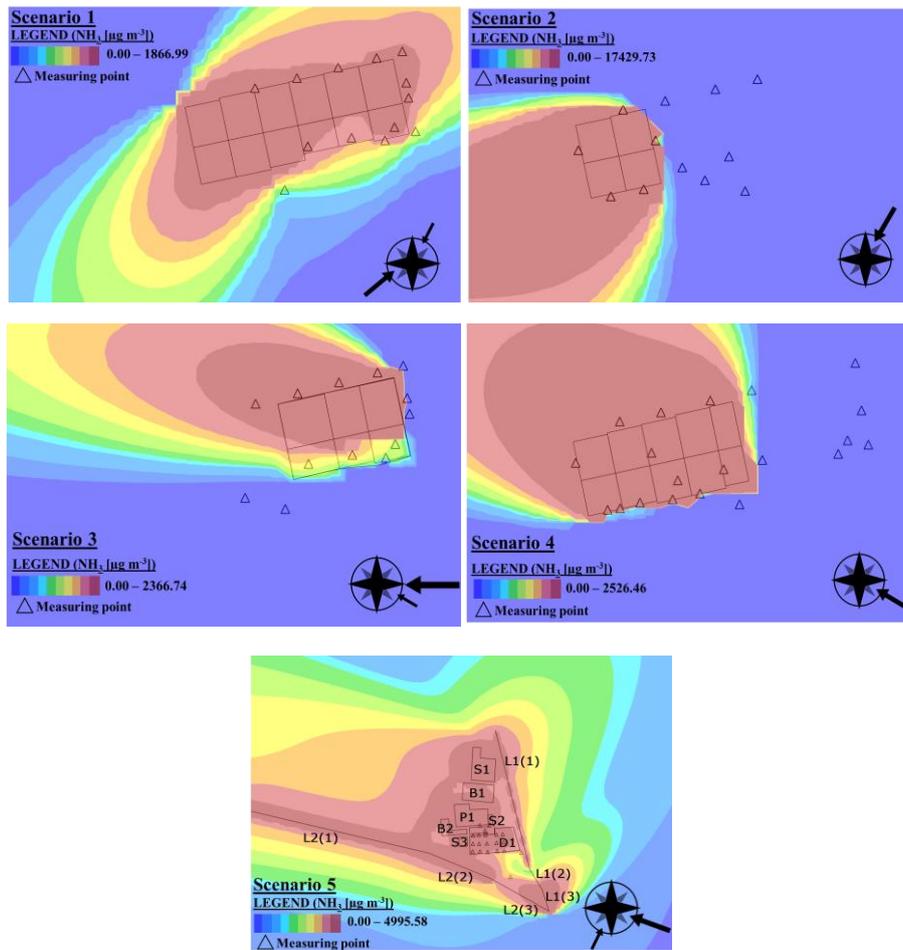


Figure 1. Average ammonia concentration on simulation period for different analysed scenarios. The black outlined areas represent emission sources.

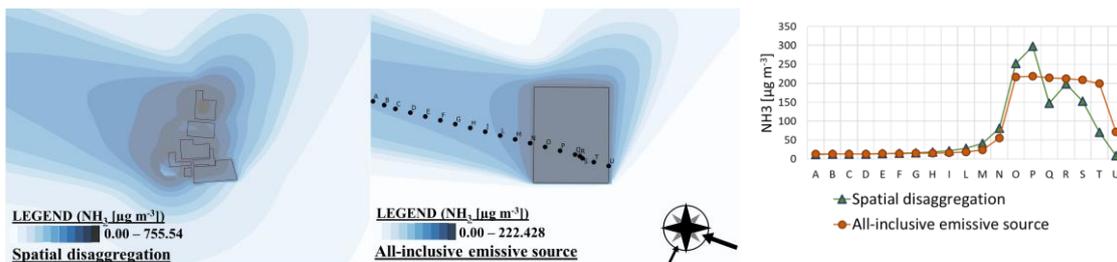


Figure 2. Ammonia concentration maps for different spatial disaggregation of emissive source areas.

CONCLUSION

Reverse modelling is a procedure that allows emissions to be quantified and to be allocated to their sources, starting from measured concentration data. In this work, this technique estimates emission flows comparable to values reported in scientific literature. Nevertheless, the results obtained refer to specific case of study and specific meteorological conditions of simulation period.

The main uncertainty in the implementation of reverse modelling occurs when a plurality of emission sources is identified, since this multiplicity involves an excessive number of parameters with respect to the number of observation (overfitting). In this case, air quality dispersion model can have difficulty in allocating emissions to specific sources.

Results of this study demonstrate efficiency of injection spreading technique in terms of ammonia emission. Nevertheless, in order to reduce ammonia emissions arising from agricultural sector, integrated interventions including all manure management phases must be considered because emission reduction in one phase can cause an increase in following steps. This phenomenon is demonstrated by the simulation of two different scenarios, referring to a swine farm, by using software BAT-Tool (CRPA, 2019), a model developed by LIFE PREPAIR project (Project PREPAIR – LIFE15 IPE IT013, <https://www.lifeprepare.eu/>) to calculate ammonia emissions from intensive pig and poultry livestock. The two cases study differ only in storage technique: without and with covering. BAT-Tool model estimates a reduction of 90% in ammonia emission arising from storage and a consequent increase of 12% in NH₃ level from slurry application when storage is covered if compared to without cover scenario.

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