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Biogeosciences Discussions

Interactive comment on "Are ammonia emissions from field-applied slurry substantially over-estimated in European emission inventories?" *by* J. Sintermann et al.

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Response to Referee 1:

We thank the referee for his careful reading and commenting of the manuscript. We appreciate the generally positive evaluation of the manuscript and acknowledge the many constructive critical comments for which our replies are listed below. For convenience, original referee comments are also included in italics. Individual responses start with '»'.

General comments The paper gives a very useful overview of the methods used to measure ammonia emissions from field-applied manure and raises important ques-

tions concerning the interpretation of the results of experiments undertaken over the last 20 years. The paper is generally well-written, particularly regarding the details of the methodologies. However, the end of the paper (sections 3.6 and section 4) is weak and does not do justice to the work that has been undertaken.

»We will strengthen these two sections according to the specific wishes and hints given by the referees (see also details below and in the response to Referee 2).

Specific comments : The authors have examined the effect of plot size on the emissions measured yet many other variables recorded in Table A1. Is it not worthwhile examining the influence of some of these other variables?

»The core information of our analysis is the clear dependence on the used plot size. Existing studies (Sogaard et al., 2002; Lim et al., 2007) have thoroughly analyzed most of the (earlier) data given in Table 1. These investigations and e.g. the resulting ALFAM model (see Section 2.4) show expected dependence on meteorological and slurry characteristics. We would just duplicate these analyses but would likely find a less clear dependence and weaker correlations due to the systematic trend in the data exhibited in Fig. 2. New reference: Lim et al., Europ. J. Agronomy, 26, 425–434, 2007.

It would be valuable if the authors commented on the usefulness of the various methodological approaches for different objectives (e.g. are small-scale approaches adequate for comparing the relative efficiencies of different abatement measures?) and whether there is sufficient information to recommend that some techniques should be abandoned completely. If this is the purpose of the new series of measurements comparing emissions from medium and large scale plots proposed by the authors, it should be stated.

»We regard small-scale approaches using a dynamic chamber approach as useful in case the goal is to characterize relative efficiencies of different management options. However, it should be kept in mind that a dynamic chamber will always alter the environment and may consequently alter ammonia volatilization compared to the undisturbed environment. We strongly conclude that the determination of emission levels should be based on measurements which ideally do not change the characteristics of the ammonia exchange at the surface. At present not enough information is available to completely abandon a specific measurement technique. We expect that the proposed new intercomparison measurements will allow a better interpretation of recent and earlier observations. We will add a corresponding statement in the conclusions.

Do the authors consider that a better understanding of the mechanisms underlying the differences in measurements between plot sizes might allow the results of earlier experiments to be used in the derivation of emission factors in the future?

»From current knowledge, as discussed in Sections 3.1-3.4, we do not expect a strong difference between emissions from medium-scale plots with radius >20m and results determined on the field scale typical for agricultural practice. If this will be confirmed with a series of new measurements, the discrepancy reported in this study will remain unexplained and thus the earlier measurements cannot be corrected.

The comparison of the initial volatilisation rate using a Michaelis Menton and a mechanistic approach is interesting. However, for broadcast slurry that is not incorporated, the parameter of interest in the Michaelis Menton equation is Nmax, which is only partially related to the initial volatilisation rate. This should be mentioned.

» According to the Michaelis-Menten approach, the total cumulative loss Nmax is directly proportional to the corresponding initial value N(0) = Nmax/Km. It was a selection criterion for the data used in the ALFAM model that the temporal behavior of the reported emissions can be described with a Michaelis-Menten approach. Therefore the plausibility check for the initial flux represents a strong constraint also to the total emission. Especially a overestimation of the initial flux will have a strong relative impact on the total cumulative emission. We will add these remarks at the end of Section 3.5.

The authors state that current emission inventories need to be updated to reflect the findings of the new generation of field scale ammonia emission measurements. This *implies that there are sufficient data collected using the latest generation of measurements to achieve this objective. I do not think this is the case.*

» Generally, in our paper we clearly do not purport to provide a fully operational new EF methodology and definite EF values for the inventory community; our more modest objective was to point to a probable flaw in existing inventory methodologies, to get a debate initiated, to identify reasons for some observed discrepancies, and to advise on adequate measurement approaches and techniques to be used in further projects. In our concluding section we strongly recommend new series of measurements with the aim to test the hypothesis whether the size of the used plot scale has such a strong influence and in order to update the current emission factors. These series should add comprehensive, more reliable datasets, upon which updated inventory methodologies should be based. Currently such a series of new measurements has been started in Switzerland and we are positive that in the very near future a clearer answer is available here.

Deriving generalised emission factors from specific field experiments is not straightforward. The total cumulative ammonia emission from a particular slurry application depends on the chemical characteristics of the slurry (particularly pH, TAN and dry matter contents), meteorological conditions, crop cover, soil conditions (affects infiltration rates) and application technology (although only broadcast spreading is in focus here). Derivation of a European average emission factor needs to take into account both the mechanisms driving ammonia mission and the conditions under which slurry is applied in practice. The data collated by the authors might be representative of European practices and conditions but probably is not; most of the data appears to have been collected in northern and western Europe.

»We are perfectly aware that a a small number of specific field experiments cannot be upscaled directly to yield a generally applicable value. The specific characteristics of a slurry application inevitably will result in a very large range of emission factors as it can be clearly seen from the scatter of the values given in Figure 2. Ideally Tier 2 emission factors used for emission inventories should be based on process based models. But it has to be kept in mind that such models need to be calibrated and validated and will produce wrong factors in case experimental results with a systematic bias are used.

The reader should be warned that both good models and representative input data are necessary to obtain a representative emission factor for Europe. In section 2.4.1, the authors could point out that the negative relationship between Nmax and the TAN concentration in the slurry in the ALFAM model runs counter to our understanding of the mechanisms underlying ammonia volatilisation.

»Our phrasing in section 2.4.1 could be misleading. In the ALFAM model N(t) and Nmax are defined in a dimensionless way as a fraction of applied TAN. Therefore ammonia loss is implicitly linearly related to TAN and the mentioned dependence of -17% Nmax per increase of 1 gN kg-1 TAN just describes a (minor) deviation from the general linear dependence. We will clarify this point in the manuscript. We do agree with the reviewer that following a pure mechanistic understanding, at least the initial volatilization flux should increase linearly with increasing TAN (Eq. 9).

A proportion of the ALFAM dataset was reanalysed and an alternative model developed (see Lim et al (2007), Europ. J. Agronomy 26 425–434). This would be worth considering as an alternative to the original model.

»We will include the proposed reference in the introduction section. However, we are not convinced by the presented artificial neural network analysis as it seems to be heavily over-trained and consequently has no predictive capability.

I would question whether Fig 1 is necessary.

»We agree that this figure can be omitted.

The word ‘animal housing’ should be used instead of ‘stables’.

»We agree.

The authors state that 'it is assumed that the calculated emission levels, together with the modelled atmospheric chemistry and disposition, successfully predict the measured ambient concentrations'. There has been much discussion of the 'ammonia gap' between predicted and measured concentrations; if the situation is resolved then a scientific reference should be used here.

»We will clarify the statement with appropriate reference to literature.

The authors ignore slurry injection as an abatement technology.

»We will add this abatement technique.

The term 'sticky' is commonly used amongst practitioners to describe the tendency of ammonia molecules to temporarily bind to solid or liquid surfaces within sampling lines. I think it is acceptable to use this shorthand term, provided it is explained when first used.

»We will add a corresponding explanation.

Interactive comment on Biogeosciences Discuss., 8, 10069, 2011.