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A volatile discourse – reviewing aspects of ammonia emissions, models and atmospheric concentrations in The Netherlands

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Abstract

In the Netherlands, there is a vigorous debate on ammonia emissions, atmospheric concentrations and deposition between stakeholders and research institutions. In this article, we scrutinise some aspects of the ammonia discourse. In particular, we want to improve the understanding of the methodology for handling experimentally determined ammonia emissions. We show that uncertainty in published results is substantial. This uncertainty is under- or even unreported, and as a result, data in national emission inventories are overconfident by a wide margin. Next, we demonstrate that the statistical handling of data on atmospheric ammonia concentrations to produce national yearly atmospheric averages is oversimplified and consequently atmospheric concentrations are substantially overestimated. Finally, we show that the much-discussed 'ammonia gap' – *either* the discrepancy between calculated and measured atmospheric ammonia concentrations *or* the difference observed between estimated NH_3 emission levels and those indicated by atmospheric measurements – is an expression of the widespread overconfidence placed in atmospheric modelling.

Keywords: Ammonia emission, manure application, ammonia emission modelling, Dutch Air Quality Monitoring Network, atmospheric ammonia concentrations, statistics

Introduction

Atmospheric ammonia (NH_3) concentrations in the Netherlands are reported to be amongst the highest in the world and are regarded as a hazard to biodiversity in natural ecosystems. Livestock are the largest contributor to ammonia emissions (PBL, 2016), and since 1993, major efforts have been made to reduce emissions. As a practical approach, the reduction of ammonia volatilization after manure application to farmland, regarded as the largest single emission source, has received much attention (Van Bruggen *et al*., 2011). In the 1990s, broadcast surface spreading made way for methods such as shallow and narrow band injection on grassland and deep placement on arable land (fallow). However, an evaluation of the scientific underpinning of the calculation of ammonia emission and deposition in the Netherlands stated that ammonia concentrations in the air 'have not decreased as much as expected since the introduction of mitigation measures. This has led stakeholders to question the effectiveness of the Dutch ammonia policy.' (Sutton *et al*., 2015) This is significant, as the Dutch agricultural community has invested much in these strategies, and is regarded internationally as environmentally innovative.

In this article, we analyse some parts of the scientific discourse on Dutch ammonia emissions. We take as our primary cue the article published by Huijsmans *et al*. (2016). Therein the focus is put on ammonia emissions from the application of cattle slurries to grassland. One of our goals was the reproduction of the presented results using the underlying data. We were motivated by both scientific curiosity and the desire to try to resolve a continuous dispute over the published results, which has implications for agricultural policies in the Netherlands.

We broaden our scope with a discussion on the much-used *Ryden and McNeill model* for fitting measured ammonia concentrations to emissions after manure application experiments (Ryden & McNeill, 1984). We also analyse the Dutch national data set of atmospheric ammonia concentrations as produced by the LML network (Landelijk Meetnet Luchtkwaliteit – Dutch Air Quality Monitoring Network).

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Reproducing ammonia emissions from manure application

In 2009, an overview of experimentally assessed emissions of ammonia (so-called emission factors) related to manure application was published (Huijsmans & Schils, 2009). The authors state that the emission factors, defined as the average total emission for each method as a percentage of total ammoniacal nitrogen (TAN) applied with the manure, are based on all available data, including the ranges in total emissions for each method. This amounts to a total number of observations of 199 on grassland and 58 on arable land, relating to various application techniques.

Unfortunately, the data sets used in Huijsmans & Schils (2009) are no longer available. Consequently, the results, republished by Huijsmans *et al.* (2016), cannot be reproduced. However, some experimental data sets gathered since 2010 have been made available, although these data do not contribute to those shown in Table 1. The results from *one* of these data sets are reproduced and discussed below (all other data sets received are mathematically treated in the same way). The results reproduced here can act as a template for other results derived from the unavailable data sets, as the same experimental techniques and mathematical model were used throughout the years (see e.g. Huijsmans, 2003). Consequently, we are partially able to explain the derivation of the emission factors and the uncertainties surrounding them, which are *not* or only obliquely reported.

Estimating ammonia emissions from manure application

In much of the Dutch policy-relevant work on the volatilization of NH_3 following manure application, the micrometeorological mass balance method was used (Denmead, 1983; Ryden & McNeill, 1984; Huijsmans, 2003). Although other methods are available, these will not be reviewed here because of our focus on Dutch emissions.

Ryden & McNeill (1984) offered a quasi-physical model to represent NH_3 flux over a manured plot. The following equation is proposed:

$$F = \frac{1}{x}\int_{z_0}^{z_p} \overline{uc}\, dz \tag{1}$$

where x is the fetch of the plot; z_0 and z_p are two heights where measurements take place; u is the instantaneous wind speed; c the instantaneous value of NH_3; and $\overline{uc}$ the time-averaged flux at height z.

This simple model states that, at a certain height, as either wind speed or NH_3 concentration increases, flux increases. This is a gross approximation at the boundary layer, where NH_3 volatilization involves many more factors other than just wind speed including temperature, soil chemistry (pH), soil moisture content, precipitation and TAN (e.g. Behera *et al.*, 2013). The solution to Equation (1) requires knowing the functional relationship between $\overline{uc}$ and z. Instead of a *physical* argument, Ryden and McNeill first assumed u and c to be (causally and probabilistically) independent. They next created two *empirical* functional relationships between $\bar{u}$ and $\bar{c}$ and height z. These relationships are themselves not derived from chemical or physical principles but were claimed to be observationally valid. The two equations are

$$\bar{u}(z) = D \ln z + E \tag{2a}$$

$$\bar{c}(z) = -A \ln z + B \tag{2b}$$

Equations (2a) and (2b) are substituted into Equation (1) for $\overline{uc}$, and the integral is then solved. The coefficients A, B, D, and E are unknown but estimated by ordinary linear regression between the natural log of height (z) and the observed wind speed and observed NH_3, the end result of which is

$$F = \frac{1}{x}\Big[-AD\,(z\,(\ln z)^2 - 2z\ln z + 2z) + (BD - AE)(z\,(\ln z - 1)) + EBz - \bar{c}_1 D\,(z\,(\ln z - 1)) - \bar{c}_1 E_x\Big]\Big|_{z_0}^{z_p} \tag{3}$$

where $\bar{c}_1$ is the ambient concentration of NH_3 on the windward side of the site, and z_0 and z_p are the height

Table 1 Emission factors (% total ammoniacal nitrogen [TAN] applied); mean and range for each slurry application method (Huijsmans & Schils, 2009)

Method	Experiments (number)	Total emissions (average based on all available data), %	Minimum, %	Maximum, %
Grassland				
Surface spreading	81	74	28	100
Narrow band	29	26	9	52
Shallow injection	89	16	1	63
Arable land				
Surface spreading	26	69	30	100
Surface incorporation	25	22	3	45
Deep placement	7	2	1	3

differences. Equation (3) shows the explicit dependence on the regression coefficients *A*, *B*, *D*, and *E*. The point estimates of these coefficients are input into the equation to produce *F*. However, as the coefficients are not known with certainty, *F* cannot be known with certainty either.

Just as there are sources of uncertainty in the relationships of windspeed and NH_3 concentration with height, there are methods to account for uncertainty in *F*. A crucial one is the physical model itself and in the posited empirical regression relationships, which we will not attempt to fathom here. Other physical–chemical models that describe NH_3 concentrations after manure application are in use, as are other statistical relationships and experimental set-ups. We have not compared those with the model of Ryden & McNeill (1984). What we can do is to incorporate parameter uncertainty in Equation (3). To do this, the confidence intervals and the central estimates for the parameters *A*, *B*, *D* and *E* are input into Equation (3) to produce a 95% confidence interval and central estimate of *F*. Below we will focus on this parameter uncertainty as a means of determining the quality of the fit of the assumed statistical relationships shown in Equation 2a and 2b. With the original authors, we assume statistical independence of the four parameters and of *u* and *c*, which of course is a first approximation.

Incorporating uncertainty in NH_3 flux estimates

With this knowledge in hand, we reproduced the published ammonia emissions by Huijsmans & Hol (2012). The data sets we obtained did not include an estimate of measurement uncertainty, and hence, the results of our analyses are overconfident.

Table 2 reproduces Table 4 in Huijsmans & Hol (2012), which includes data from four different fields from two separate weeks, which we were able to reproduce without issue. However, estimates of uncertainty are now included.

For example, the central estimate of *F* as found in line 4 is 18.7 kg NH_3-N/ha. After incorporating parameter uncertainty, the 95% confidence interval is 14.8–42.0 kg NH_3-N/ha. It is clear from examining Table 2 that the uncertainty in the regression coefficients produces sizeable uncertainties in F. We could not discover uncertainty measures used by authors employing the model of Ryden & McNeill (1984). The uncertainties we present assume the validity of that model, which itself is only a rough approximation of the actual physics and chemistry. We only provide the uncertainty inherent in the output of the model of Ryden & McNeill (1984), which thus is still an underestimation of the total uncertainty. The uncertainty in *F* obviously is bounded at the lower end by 0, and the empirical functional relationship is logarithmic, which explains the asymmetry in the confidence intervals. The flux *F* itself is also limited to the total amount of TAN applied and that from the ambient air. In every case, the upper bounds of the confidence intervals are about two-times the central estimate, while the lower bounds are about 10–20% of the central value.

Week 16 data of Table 2 for line 4 are shown in Figure 1. For early periods (after manuring), Equation 2b does a poor job at modelling the data. The data are below the model for most of the range. In later periods, the approximation is better, but as most of the NH_3 contribution comes from earlier periods; overall the model of Ryden & McNeill (1984) will overestimate values. Our analysis reveals this to be typical for all the data we examined. Similar plots were made (not shown) for windspeed and height, showing better fits between Equation 2a and measured data, but also showing the need for uncertainty intervals. An interesting problem with the model regression is seen in Periods 1 and 2, which are the most important. At certain values of the log height, there are predictions of negative NH_3 values, which of course are physically impossible (Figure 1; red arrows). The assumption of independence of the parameters for calculating the intervals in Table 2 is open to question. As we are interested in the predictive uncertainty of *F* and given the shortcomings of the regression models shown as Equation 2a and 2b detailed in Figure 1, this is not unreasonable. Of course, the only way to

Table 2 Reproduction of Table 4 of Huijsmans & Hol (2012) including confidence intervals around central estimates of *F*

Week	*N*	Application[a]	Ammonia emission kg NH_3-N/ha Mean [95% CI]	Ammonia emission % NH_4-N of total N applied Mean [95% CI]
16	1	On potato ridges + incorporation 1	28.7 [22.2, 50.9]	13.9 [10.8, 24.8]
	2	On potato ridges + incorporation 2	23.9 [18.9, 47.1]	14.9 [11.7, 29.3]
	3	In slits 1	29.7 [23.2, 69.1]	18.7 [14.5, 43.4]
	4	In slits 2	18.7 [14.8, 42.0]	11.8 [9.32, 26.5]
17	5	On potato ridges + incorporation 1	15.6 [13.1, 23.1]	10.0 [8.44, 14.9]
	6	On potato ridges + incorporation 2	25.5 [19.3, 151.0]	16.2 [12.3, 96.1]
	7	In slits 1	37.1 [28.6, 88.2]	24.2 [18.7, 57.5]
	8	In slits 2	25.6 [19.6, 60.9]	17.3 [13.2, 41.2]

[a]The experiment consisted of two series of measurements executed with two different manuring techniques. The experiment was carried out twice in weeks 16 and 17.

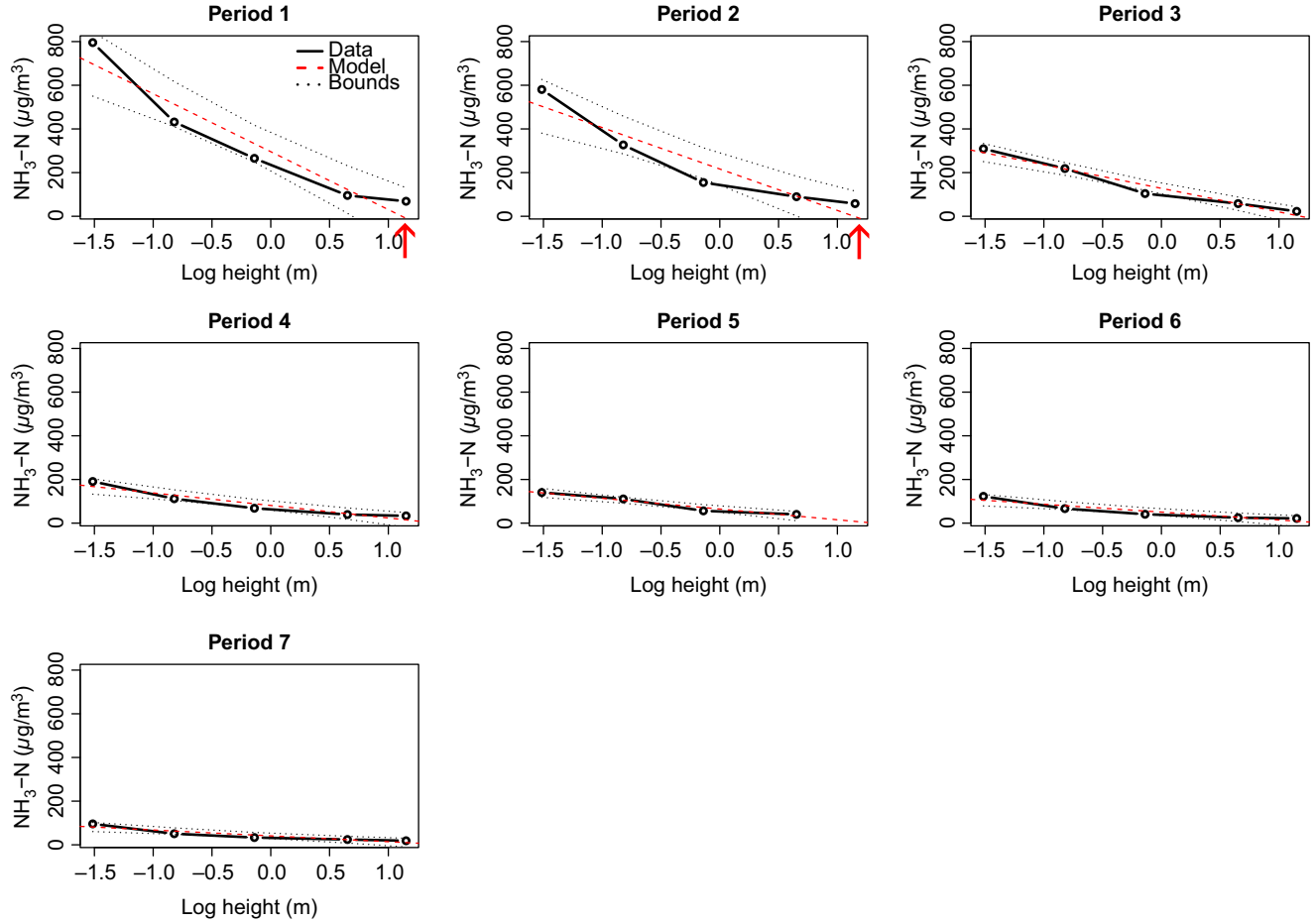

Figure 1 Log-linear empirical relationship between height and mean NH_3 concentration. (The small black circles indicate the actual data; data were measured at seven periods (there were data missing in period 5 at height 3.16 m). Note the log scale for height. The dashed red line represents Equation 2b, and the dotted black lines are the 95% uncertainty bounds to 2b.)

confirm these or any assumptions is to verify (probabilistic) predictions of F with actual measurement. The above has implications for the emission calculations that are made on a national scale and the connection with actual atmospheric concentrations as measured by the LML network.

Ammonia in the Dutch atmosphere

Measurement of atmospheric concentrations of ammonia helps to link the emission of ammonia with its deposition. We agree with Erisman *et al.* (1998) that in 'the causal relation emission-concentration deposition effects, the concentration and deposition observations might serve as a test of the effectiveness of measures and the temporal development of emissions.' Sutton *et al.* (2003) noted that '[c]orrect interpretation of adequate atmospheric measurements is essential, since monitoring data provide the only means to evaluate trends in regional NH_3 emissions.' The latter quote underscores the fundamental and (more or less) exclusive empirical quality of atmospheric concentrations above and beyond computed estimates of ammonia emissions and deposition.

Atmospheric ammonia concentrations have been measured since 1993 with the LML network. Ammonia is measured at stations Vredepeel (S131), Huijbergen (S235), De Zilk (S444), Wieringerwerf (S538), Leiduin (S540), Zegveld (S633), Eibergen (S722), Lunteren (S734), Wekerom (S738), Witteveen (S928) and Valthermond (S929); however, not all stations have complete data sets either from 1993 or through to 2014, the final year for which we received data.

The individual data sets are aggregated to national atmospheric averages to produce a mean of all the data from all stations. The importance of these atmospheric concentrations lies in the fact that ammonia deposition calculations are calibrated with the data gathered through the LML stations (see e.g. PBL, 2010). So, these empirical data connect emission estimates and deposition calculations and are thus highly important. The national averages seem to indicate a decline between 1994 and 2004 from 10 to 8 $\mu g/m^3$, and thereafter, concentrations remain at roughly the

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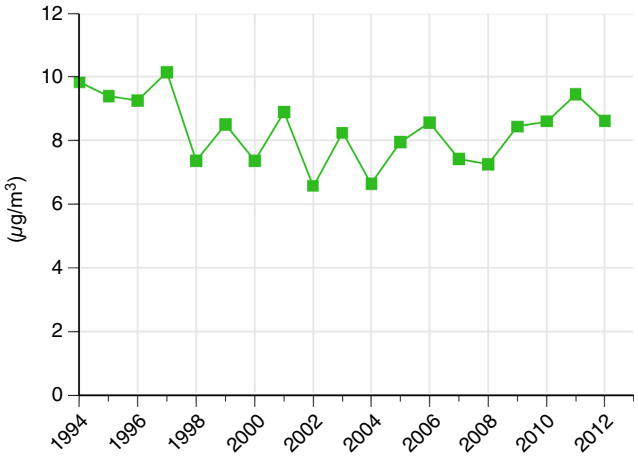

Figure 2 Annual national average ammonia concentrations from 1994 to 2012 (RIVM, 2013).

same level (Figure 2). To better understand the apparent trend, we conducted a number of analyses. First, it is noted in Figure 3 that the hourly LML data are highly variable, both within and across stations. This persistent variability makes summarizing the data, such as with a yearly national average, problematical. Indeed, all one-number summaries of data as complex as this necessarily omit crucial details and this cautions against drawing overconfident countrywide conclusions.

As is evident from Figure 3, levels of NH_3 are high at some stations (S131, S633 and S738), moderate at others (S538, S722 and S734) and low at the rest (S235, S444, S540, S928 and S929). Records are incomplete at four stations (S540, S734, S928 and to some extent S929).

Figure 4 illustrates the difficulties of ascertaining (national) trends. A popular method to define a trend is to fit a routine linear regression on the value of interest (here NH_3) at the starting point of a time series to the endpoint,

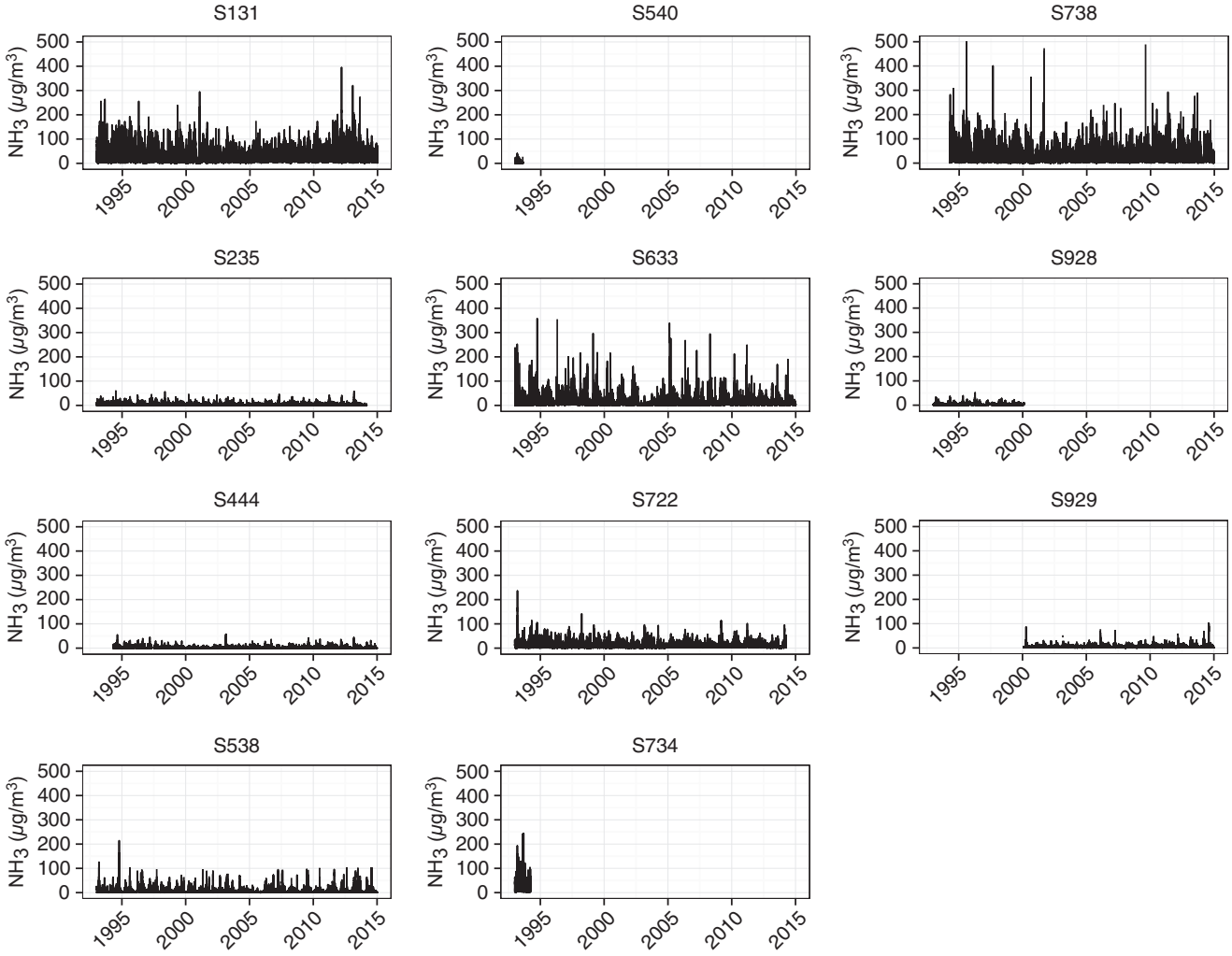

Figure 3 Times series plots of atmospheric ammonia concentration for each LML station.

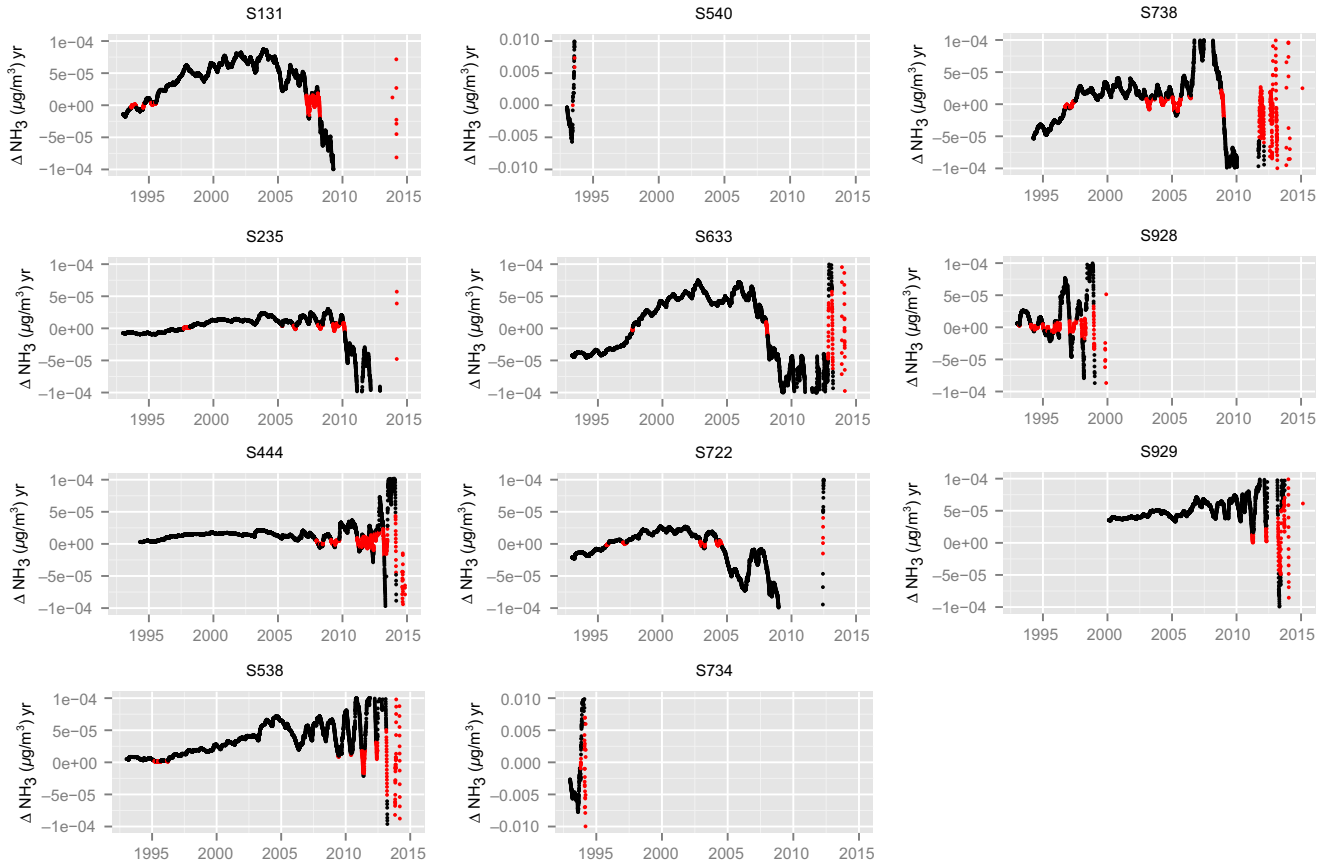

Figure 4 Trend analyses for all LML stations. (Points which are 'statistically significant' at 95% are black; those which are not 'significant' are red.)

using the date as the regressor. We performed a full trend analysis of all the data of all stations.

Each point is the result of a different regression with a different start date, but the same end date (the last date of data for each station). The start date is stepped along the series, starting with the first date available, then the second (excluding the first), then the third (excluding the first two dates) and so on. This approach harbours a bias that is frequently overlooked. The start and end dates of the data series are arbitrary, and merely by choosing different dates, trends oscillate between positive and negative, and become significant or non-significant by the simple change of a date. Some of this is due to the spiked nature of the series and some due to the inherent variability of ammonia concentrations. It is clear that no countrywide trend signal is evinced, and neither is there any clear indication of a consistent trend at any station (in contrast to findings by Van Zanten *et al.*, 2017).

Furthermore, every station records highly transient spikes in NH_3 concentrations (Figure 5). Each subplot shows a histogram of NH_3 concentrations at each individual LML station. The data are grouped into 'bins' of NH_3 concentrations, and the frequency of observation for each 'bin' (count) is reported. Those points that occur with a frequency <1 in a 1000 (0.1%) are shown with '+' signs, to highlight the large transient values. Clearly, none of the plots have a normal distribution, and as a result, summary measures like the mean and standard deviation can be misleading (Galton, 1907).

With data that is symmetrical or even roughly 'bell-shaped', a mean can be a useful one-number summary as an expression of the average behaviour of the system. In cases of high skew, which is evinced here, the median is preferred, because it gives a more accurate summary of data tendency and system behaviour. When data are symmetric, as when using normal distributions to characterize uncertainty (which is *not* defensible here), the mean and median coincide or are very close. Thus, there is often little reason to prefer the mean, unless there is some physical or causal reason. That causal reason does not exist here. For instance, some statistical models that represent ammonia concentrations using normal distributions inappropriately characterize measurements as a 'global' mean plus departures from that mean as if some physical process 'wants' to 'return' ammonia levels to that mean. This makes no physical sense, as the many causes for actual NH_3 concentrations are not

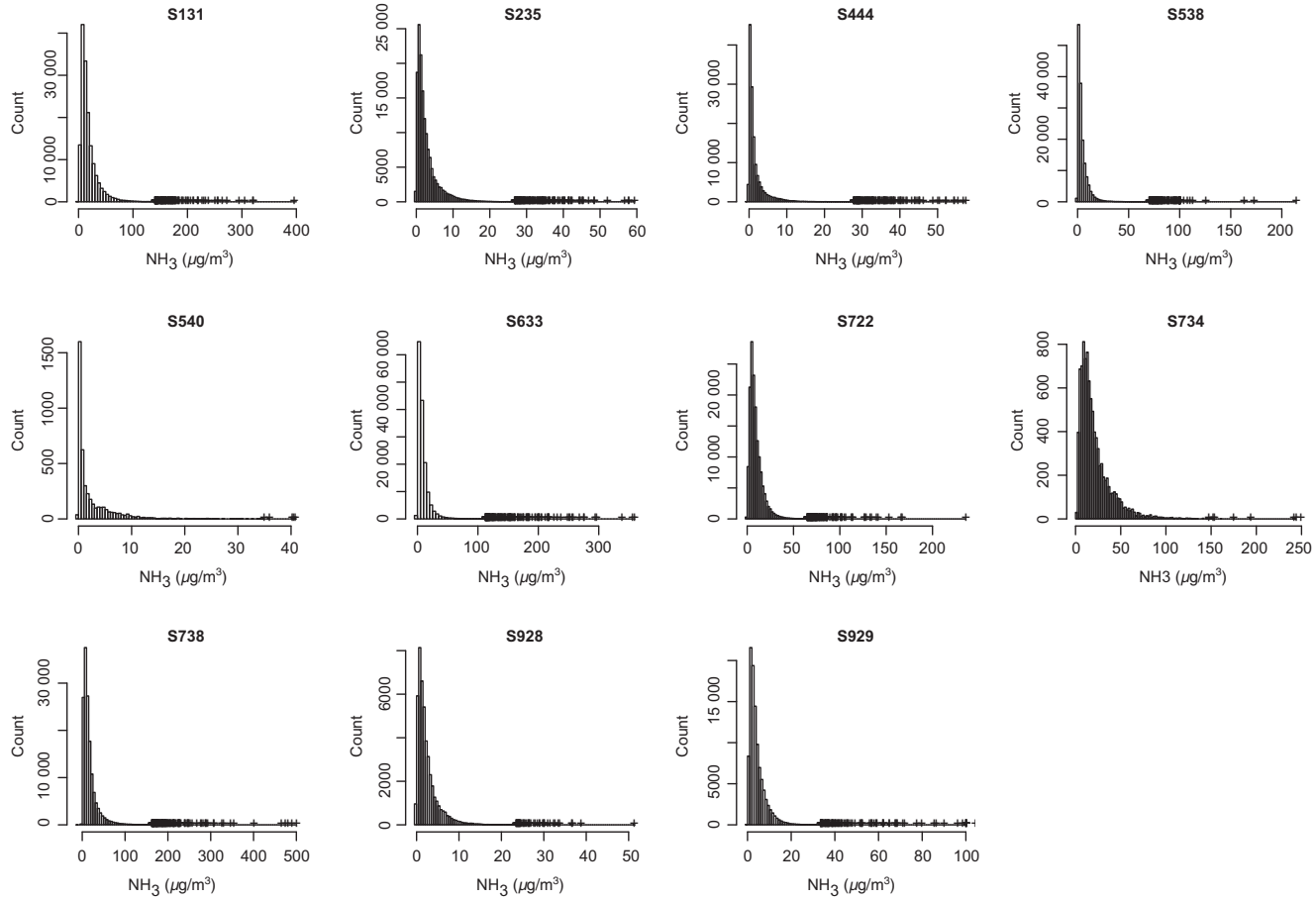

Figure 5 Histogram of atmospheric NH_3 concentrations at each LML station.

Table 3 Mean and median of all hourly measurement values per LML station and ratios of mean to median

Name	Mean $\mu g/m^3$	Median $\mu g/m^3$	Mean/median ratio
S131	18.37	13.13	1.40
S235	2.84	1.76	1.61
S444	1.83	0.82	2.23
S538	4.73	2.84	1.67
S540	2.67	0.86	3.10
S633	9.01	6.10	1.48
S722	9.48	7.45	1.27
S734	21.65	15.99	1.35
S738	16.86	11.56	1.46
S928	2.46	1.63	1.51
S929	4.35	3.19	1.36

'restorative' in this way. Instead, factors such as soil chemistry, fertilizer application timing, wind entrainment, precipitation, atmospheric chemistry and other mechanisms cause levels to continuously change, and often to swing wildly and to spike, as the plots in fact show. All these aspects are good reasons to choose the median as the most accurate representation of the system's behaviour.

The mean is highly sensitive to spikes in data. Table 3 summarizes mean and median for each LML station. The mean/median ratio shows that the mean in this skewed data is always higher than the median, with the smallest 'boost' in the average being 27% (at S722). When the data series is short, as it is at S540, the disparity is even larger, with the mean being 310% larger than the median. Use of the mean is therefore very misleading (Figure 6).

The wide heterogeneity of the LML data, both in measurement and topography (station location and local land use and so on), firmly argues against the use of an annual nationwide mean across the LML stations (see RIVM, 2013). This erroneously gives equal weight to each station in the mean. Because of this, for instance, a station that is on average 'up wind' will have more influence over national totals than another that is downwind or near the border of the country and which therefore cannot contribute much to national agricultural emissions (Sutton *et al.*, 2015).

To drive the point home with respect to the difficulty of national yearly averages, a representative scatterplot of

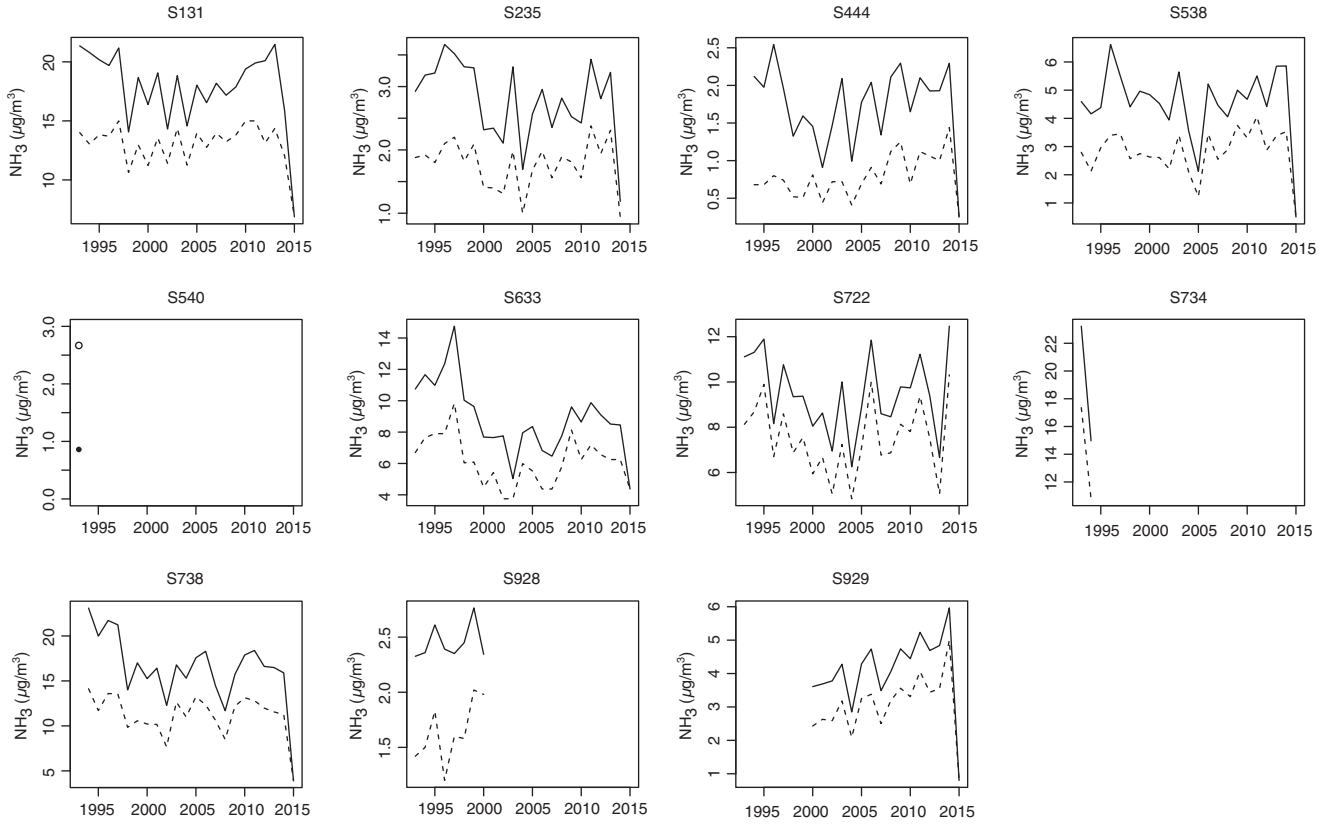

Figure 6 Monthly mean (solid) and monthly median (dotted) atmospheric ammonia concentrations for all LML stations.

hourly NH_3 values for two stations, S444 and S538, is given in Figure 7. This and the other plots we have produced (not shown) reveal that hourly NH_3 concentrations at one station are not well correlated with NH_3 concentrations at other stations. This implies that the causes of NH_3 in the atmosphere are localized and not countrywide. There is some

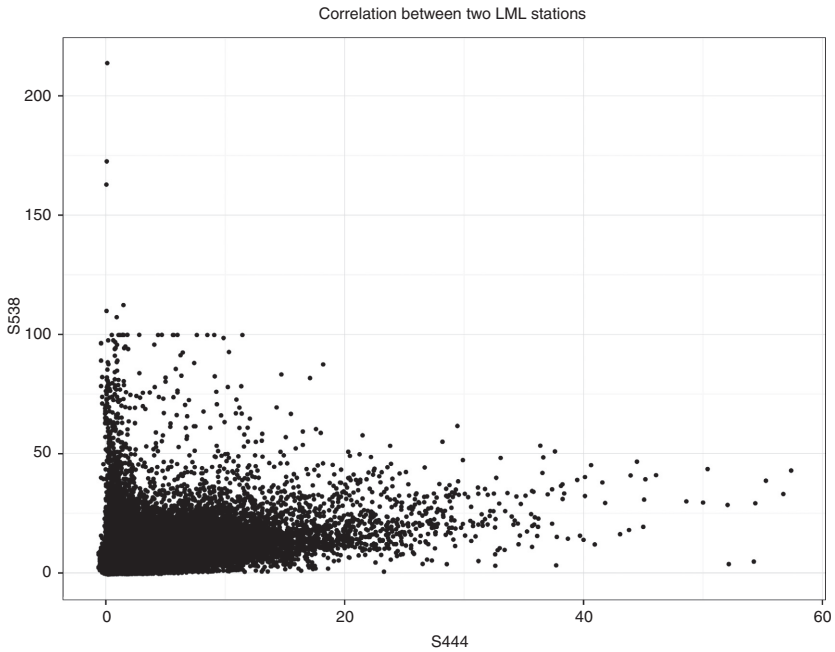

Figure 7 A representative scatterplot of NH_3 concentrations for two stations, S444 and S538.

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unsurprising evidence of a seasonal cycle embedded in the variability at each station. This localization argues against a countrywide single-number annual summary. Obviously, correlation necessarily increases between stations when data are aggregated to, say, monthly averages. This is purely the result of smoothing the data and does not in any way indicate increases in causal relationships or 'links' between sets of data (Briggs, 2016).

Unravelling emission and concentration estimates

In the years after the LML network became operational, it was noticed that atmospheric ammonia concentrations did not greatly change. That is unexpected because several emission reduction measures had been put in place and emissions in this period were estimated to have decreased by 35%. Later, this discrepancy between calculated emissions and the LML concentration measurements was dubbed the 'ammonia gap'. Erisman *et al.* (2001) defined this gap as 'the difference observed between the estimated NH_3 emission levels in the Netherlands and those indicated by atmospheric measurements'. Sutton *et al.* (2003) take Erisman *et al.*'s definition to its logical next step. They state that the 'gap' is 'the lack of a detectable reduction in NH_3 concentrations following the implementation of abatement measures in 1993.' Here, a downward signal is expected in the atmospheric ammonia concentrations as a result of abatement measures, which nevertheless does not show. That missing signal is referred to as the 'gap'. Conversely, in 1995, the RIVM issued a report in which an evaluation was presented of calculations of atmospheric ammonia concentrations in the Netherlands related to measurements (RIVM, 1995). With the aid of annual averaged values, modelled ammonia concentrations that are based on national emission estimates were 27% lower than the measured concentrations. This discrepancy persisted despite adding 16% to estimated national emissions. Although the report does not mention the 'ammonia gap' as such, the first outlines thereof seem to emerge with the discrepancy defined as the difference between averages of measured and modelled atmospheric ammonia concentrations. In line with this, De Ruiter *et al.* (2006) defined the 'ammonia gap' as the difference between the modelled atmospheric ammonia concentrations (with the operational priority substances (OPS) model) and the measured concentrations, the former being substantially *lower* (by some 25 to 30%) than the latter (see Velders *et al.*, 2010). So, the first definition of the 'ammonia gap' focuses on the ostensible difference between estimated ammonia emissions (which ostensibly change) and the measured atmospheric ammonia concentrations (which do not change), whereas the second definition revolves around the discrepancy between modelled and empirically measured atmospheric ammonia concentrations. It seems that considering the first definition, the 'gap' is now bigger than ever.

There is a rich scientific literature on the Dutch ammonia emissions. This gives the opportunity to see how the emission calculations changed but also how views evolved. In the 1980s, estimates of the total ammonia emissions in the Netherlands were in the order of 200 kt/year (Erisman, 1989). Between 1990 and 2016 ammonia emissions estimates roughly doubled with most of the increase occurring around 1990. The biggest change in historical emissions was due to increases in emission factors for manure application (see Van Bruggen *et al.*, 2011). In the nineties, an emission factor of 50% of applied TAN for surface spreading was assumed (Van Bruggen *et al.*, 2011, p. 107). Following the work of

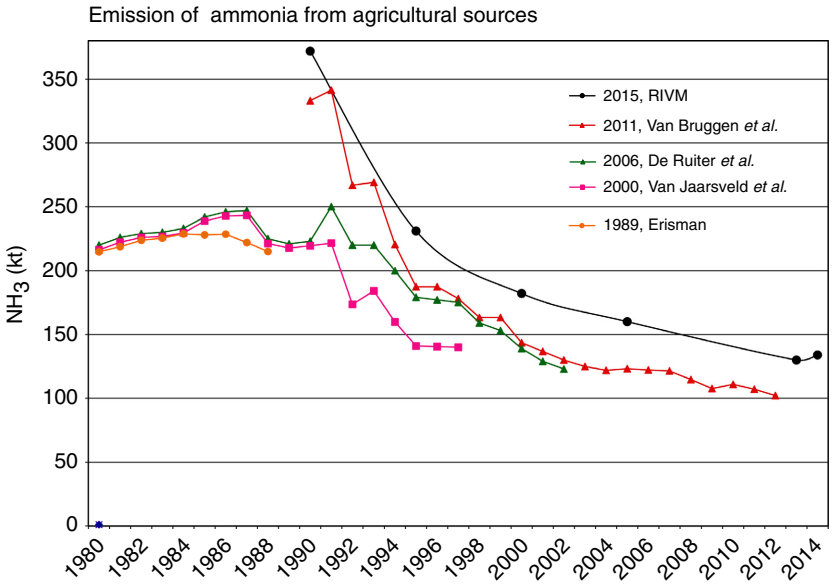

Figure 8 Different estimates (with references) of ammonia emission trends.

Table 4 Manure application techniques and emission factors as specified in the NEMA model

Manure application, techniques and emission factors	Share, %	Emission Factor (EF), %
Grassland – slurry		
Shallow injection	61	19.0
Slit coulter	13	22.5
Narrow band	25	26.0
Surface spreading	1	74.0
Cropland (uncropped) – slurry[a]		
Deep placement	71	2.0
Shallow injection	9	24.0
Slit coulter	9	30.0
Narrow band	7	36.0
Direct incorporation	4	22.0
2-pass incorporation	0	46.0
Surface spreading	0	69.0

[a]Manuring before crops were planted.

Huijsmans (2003), the emission factor for 1990 was changed to 74%, as surface spreading of manure was the predominant method at the time. As low-emission techniques for manure application were introduced from 1991 onwards (see Table 1 for examples), the estimated emissions have decreased considerably according to the updated calculations (Figure 8).

Given the substantial uncertainties relating to (nationwide) emissions, expressed in our discussion of the use of the model of Ryden & McNeill (1984), and all the complex processes that take place in the atmosphere, it is not surprising that trends in emissions and atmospheric concentrations differ or that model calculations are at odds with measured concentrations. Nevertheless, in terms of emission factors, overconfidence is inbuilt in the national emission inventories. The National Emission Model for Agriculture (NEMA) model is used to calculate agricultural emissions to the atmosphere in the Netherlands, with ammonia emission factors are simply specified as one decimal numerals (Table 4).

Coining the term 'ammonia gap' suggests that discrepancies between either the calculated atmospheric ammonia concentrations and actual measurements *or* changing emissions and measured (roughly) unchanging atmospheric concentrations are somehow physically real: this evidently is a false notion. More precisely, the gap has been 'reified into reality'. Reification is a widespread and classical fallacy dubbed by Alfred North Whitehead (1925) as the 'fallacy of misplaced concreteness'. Reification is making something which is hypothetical or abstract physically real. Abstractions in science are quite common and necessary; however, trouble arises when we start to think of abstractions as if they were concrete realities themselves – thereby 'reifying' them. This predicament is intensified when we think of the abstractions as somehow *more real* than the concrete realities from which they have been abstracted (see Briggs, 2016). Using the mean averages for the LML data and even aggregating the data to a national scale exacerbates this reification. Reducing the temporally and spatially highly variable atmospheric concentrations to one annual number produces over-certainty, or at least masks the substantial uncertainty present. The median as the correct and thereby more accurate expression of the complex data lowers local atmospheric ammonia concentrations substantially (see Table 3 and Figure 6).

Concluding discussion

We are aware that we have put large question marks over seemingly settled aspects of the ammonia discourse in the

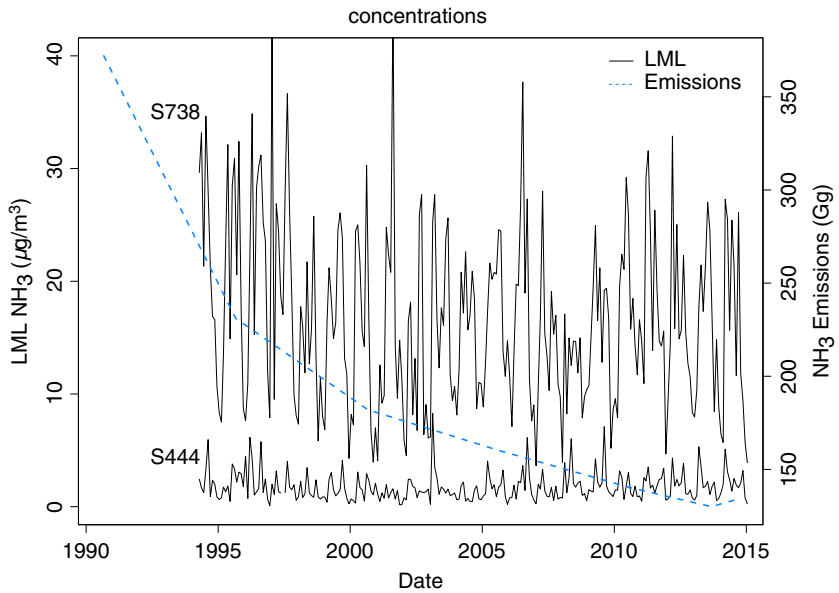

Figure 9 Estimated ammonia emissions (Gg) since 1990 (RIVM, 2015) and ammonia concentrations at two LML stations, one high and one low measurement; median values between 1993 and 2014.

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Netherlands. Firstly, the non-availability of the experimental data that underpin current agricultural policies and form a key part of the paper of Huijsmans *et al*. (2016) is worrying. Reproducibility is a crucial epistemic value that nowadays is at the forefront of scientific discussion (e.g. Ioannidis, 2005; Horton, 2015; Peng, 2015; Browman, 2016; Munafò *et al*., 2017).

Secondly, we have shown that the uncertainty in fitting experimental emission data is substantial, and yet, model-generated confidence intervals for the central emission estimates are not reported (Table 2). Emission factors derived from experiment and modelling (Ryden & McNeill, 1984) are published and used with an amount of overconfidence and accuracy that is not appropriate for the actual data gathering and subsequent statistical work (see e.g. Vonk *et al*., 2016).

Changing manure application from broadcast spreading to shallow injection, for instance, is likely to have resulted in emission reduction in the Netherlands, but by how much remains unknown. Our analysis of one data set gives enough insight into the model uncertainty to seriously question the NEMA model in which emission factors from experimental research are applied to national emission inventories with a precision of one decimal place. Even our own analysis is likely to be fraught with overconfidence, as for instance, measurement uncertainty is unknown. As a result, emission factors in the NEMA model carry sizeable uncertainties that are not made explicit and should carry over to the emission totals produced by the model. Indeed, Sutton *et al*. (2015) remark that no overall synthesis in the uncertainty in the trend of all contributions to Dutch ammonia emissions has been conducted.

We would assert that the uncertainty in the output of the equation given in Ryden & McNeill (1984) at least would result in emission uncertainties that would perhaps result in overlap between the different manure application techniques (see Table 1). Thus, the widely reported dividing lines between emissions from different manure application techniques are likely blurred. In some ways, this is expressed in Figure 9 where the ammonia emission estimates since 1990 are overlaid with LML-median value series for two stations, one with high and the other with low atmospheric ammonia concentrations. The 1990 value for estimated ammonia emissions is 373 kt (RIVM, 2015) or 372 kt (RIVM, 2016). Clearly, there is no correlation between estimated emissions and atmospheric median concentrations. The picture is similar for all LML stations:

Thirdly, current official output from the LML database leaves much to be desired. Two things are quite clear from our analyses: (i) median values should be the default when summarizing the highly skewed LML data set as it best describes the average system behaviour; (ii) no countrywide trend signal is evinced from the hourly LML data, and neither is there any consistent trend at any station. Use of the mean gives a misleading picture and suggests 'average' atmospheric ammonia values that are simply too high (Galton, 1907). Ammonia concentrations in the Dutch atmosphere are substantially lower than reported in the official outputs, resulting, we venture, in lower subsequent deposition. Actual ammonia emissions could be lower as well, although this was explicitly not researched here. With the reduction of data to a one-number annual summary, much useful information is being discarded.

Finally, the extensive research efforts over the past thirty years in the Netherlands have provided much valuable data on the agricultural impact on the local and national environment and beyond. However, lack of data transparency, oversimplified statistical procedures, and the resulting spurious accuracy of published and applied emission results dilutes the usefulness of these research efforts. This has created an environment in which results used for regulatory purposes seemingly can be employed without the usual *and* compulsory scientific provisos. This needs to change and we have provided some tools to make that change happen.

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