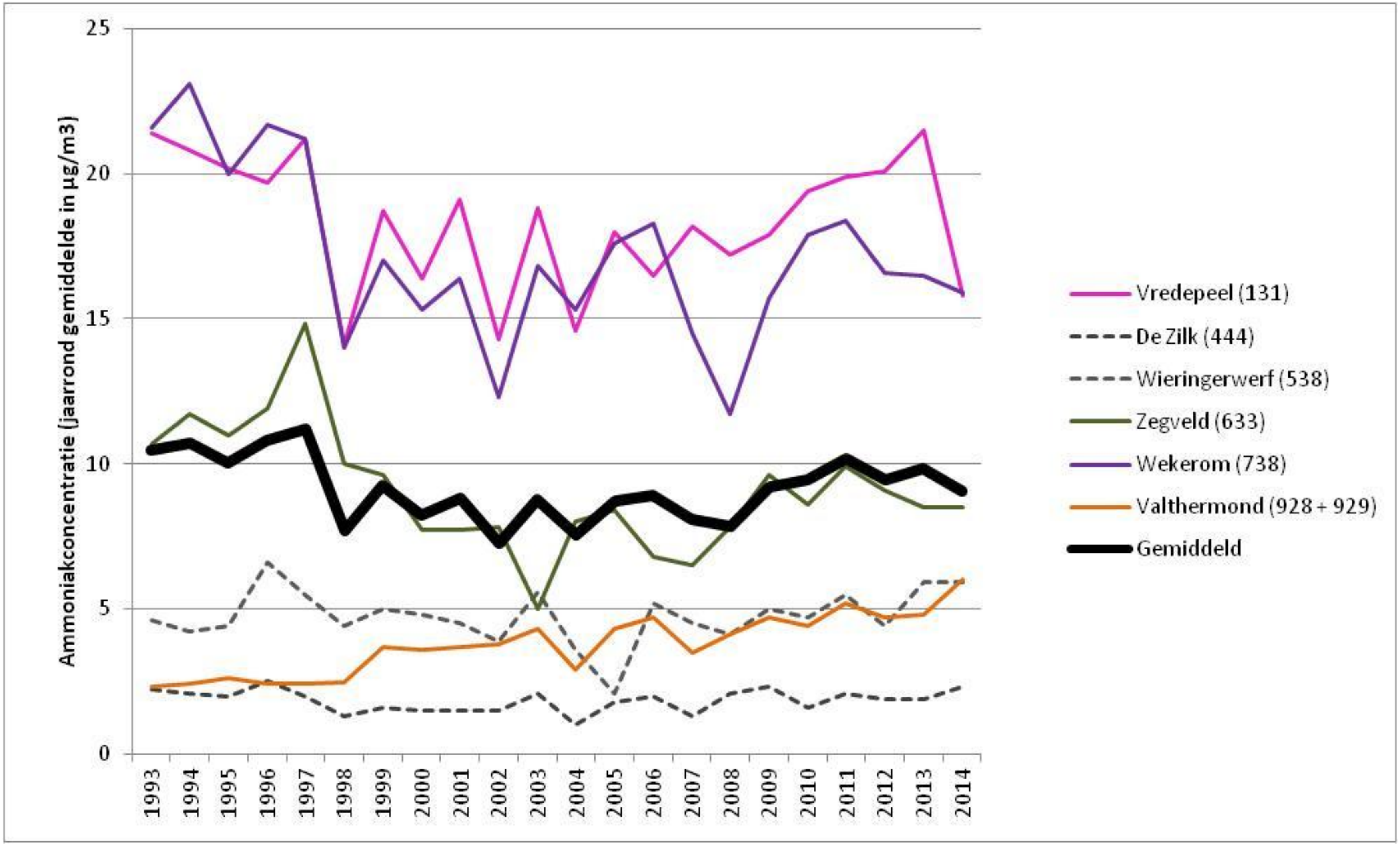
Ammoniak in Nederland

Een noordoostelijke spelbreker

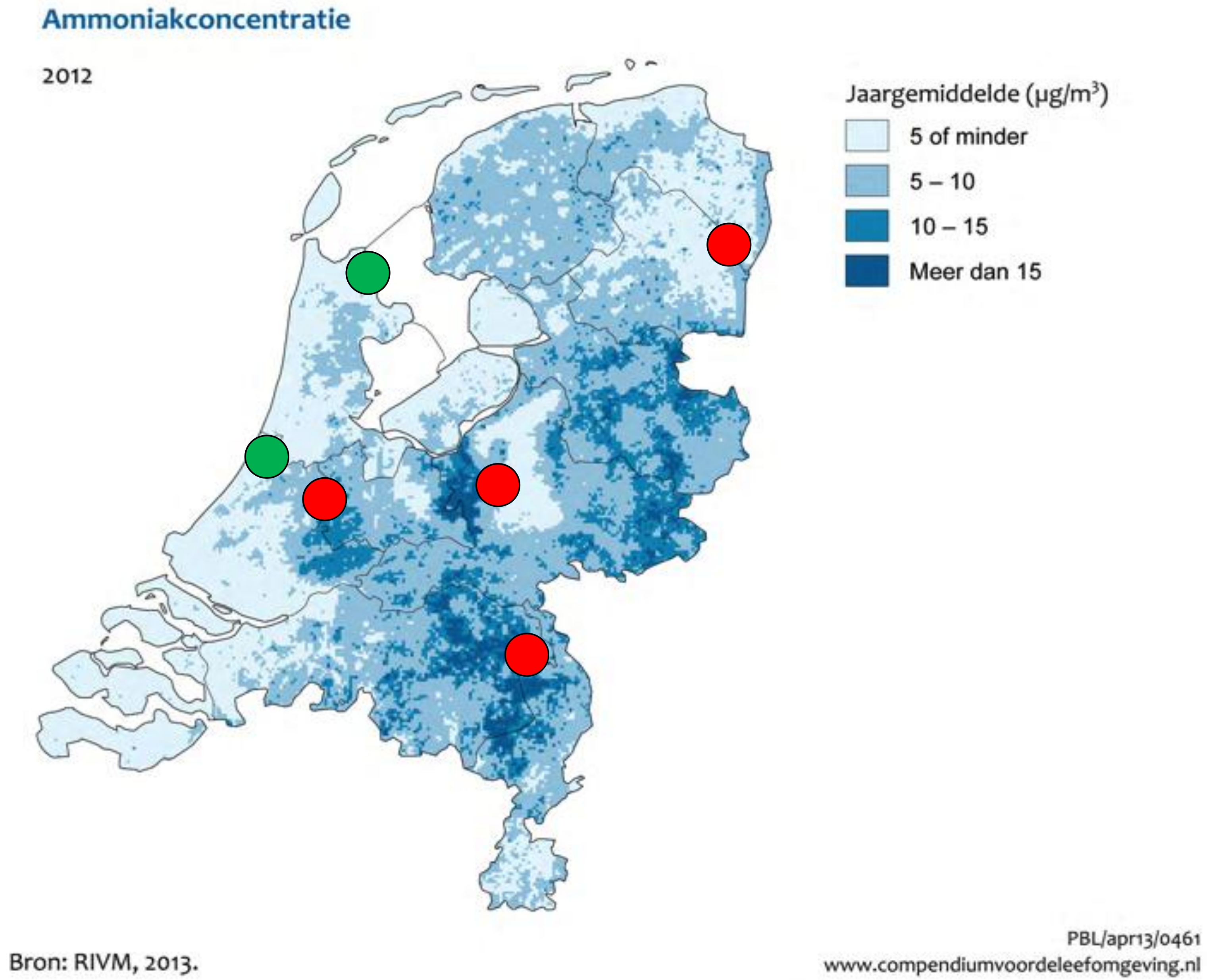
De impact van een foutief geplaatst meetstation op het Nederlandse ammoniakbeleid

Geesje R. Rotgers en Jaap C. Hanekamp

Ammoniak in NL: 6 stations (4 voor landbouw)

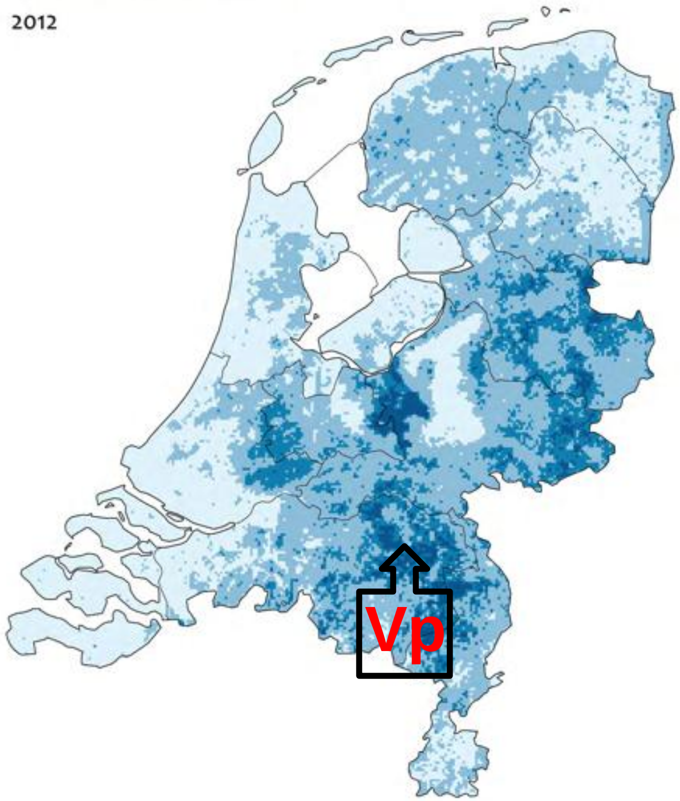


De 6 LML meetstations in Nederland



Ammoniakconcentratie

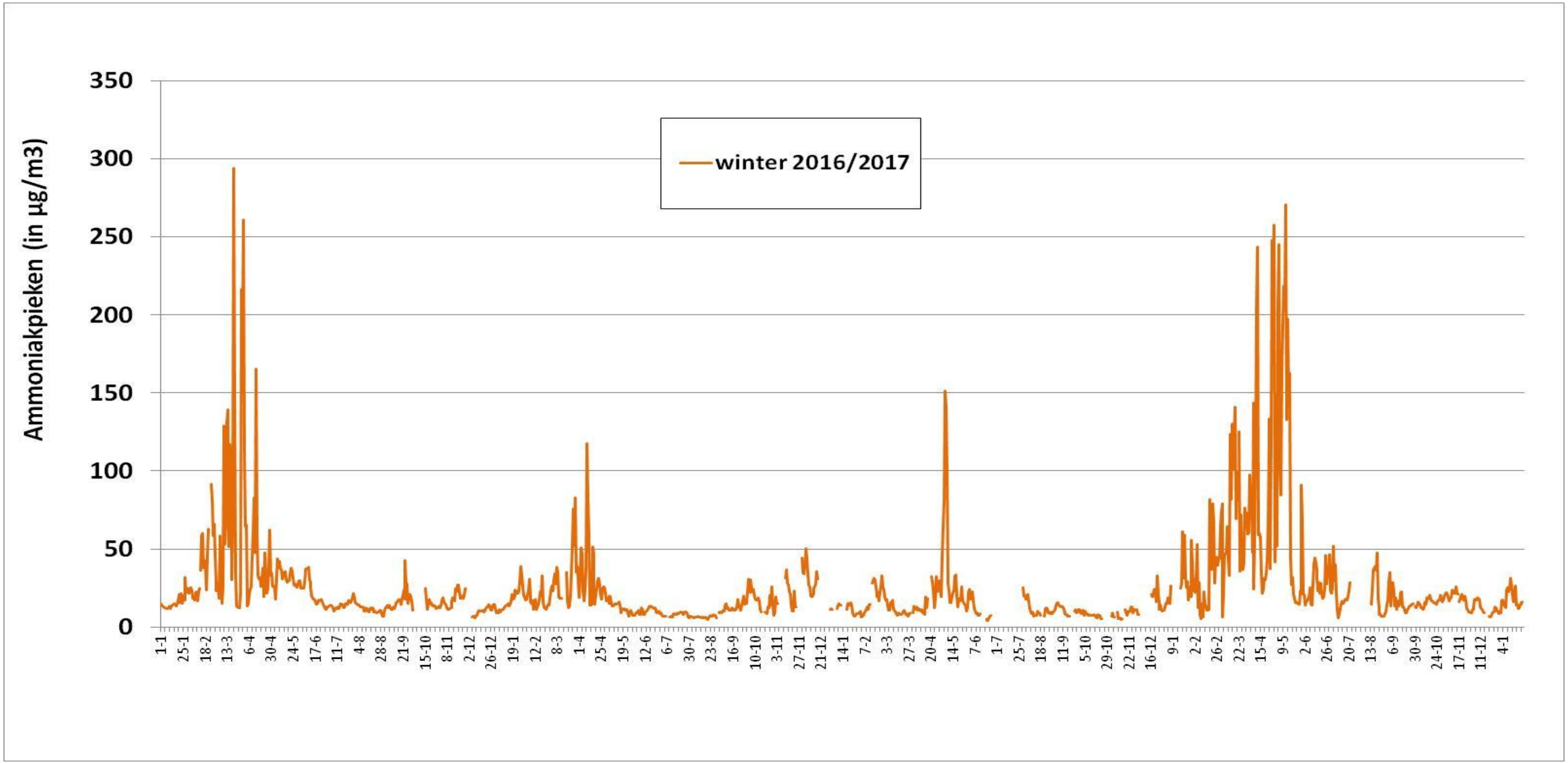
2012



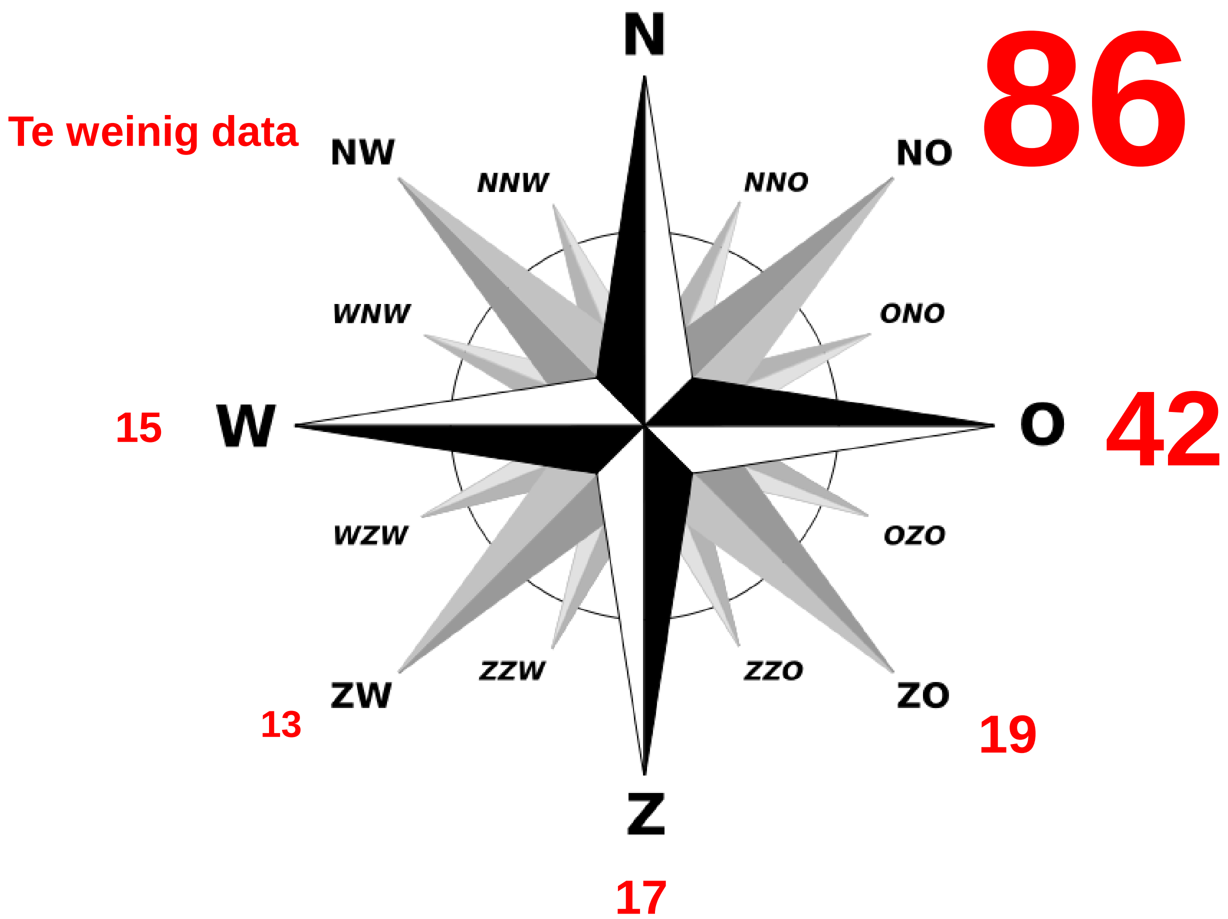
Jaargemiddelde (μg/m³)
- 5 of minder
- 5 – 10
- 10 – 15
- Meer dan 15

Bron: RIVM, 2013.

Een ‘rare’ bevinding op station Vredepeel: ‘mestpieken’ in de winter?

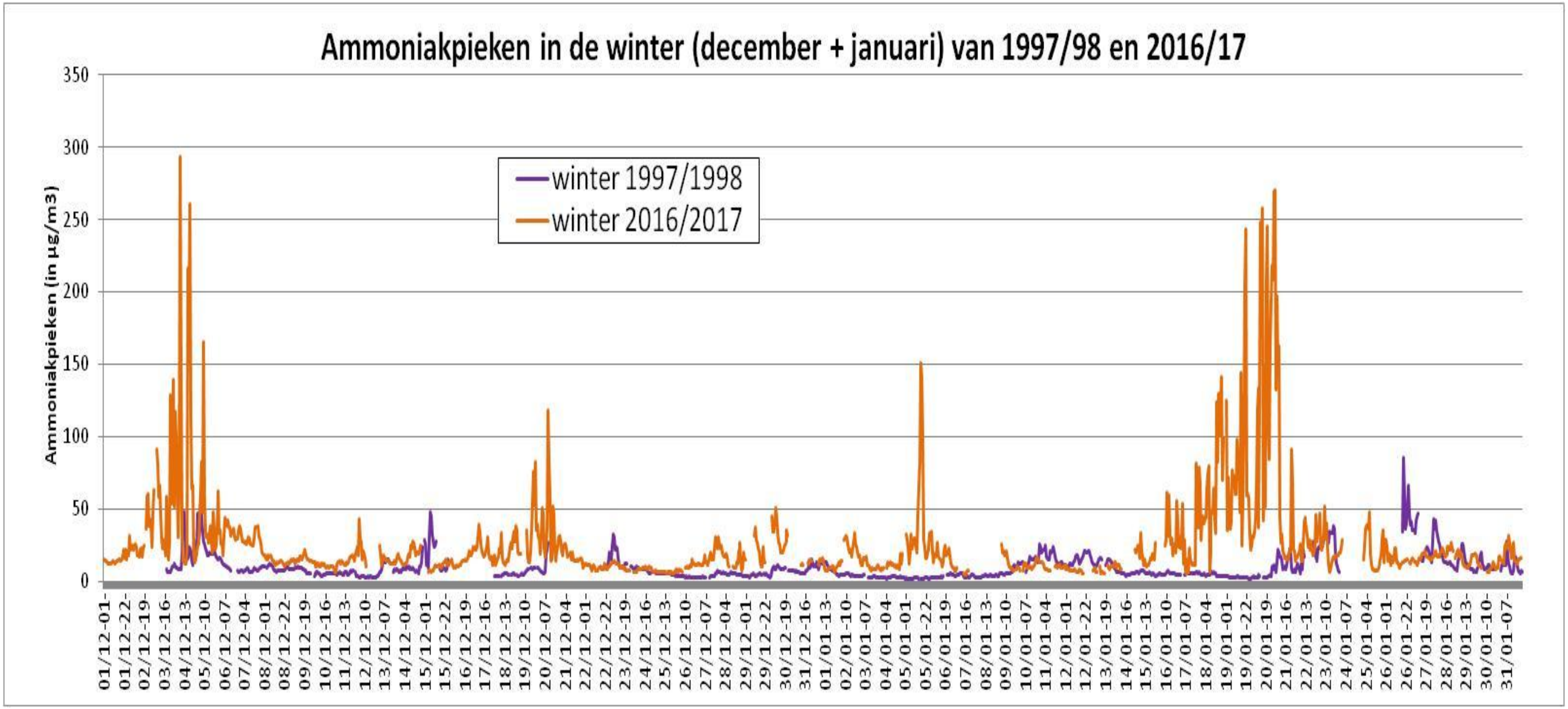


De gemiddelde concentraties ammoniak gemeten bij verschillende windrichtingen, in de periode 1 november 2016 t/m 31 maart 2017

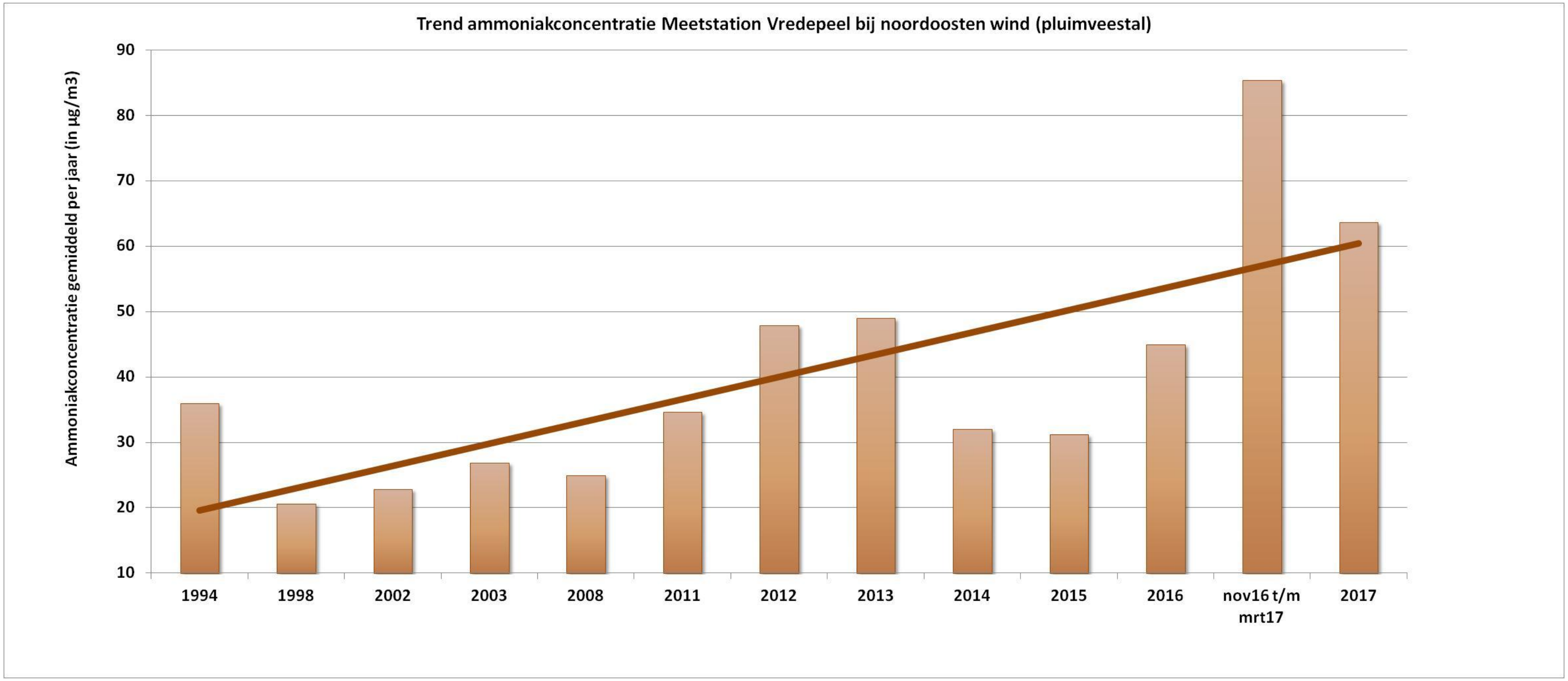


LML meetstation Vredepeel

Vergelijking van de hoogte van de ammoniakpieken in de winters 1997/1998 en 2016/2017, gemeten door meetstation Vredepeel



Trend van ammoniakconcentraties LML station Vredepeel bij NO wind



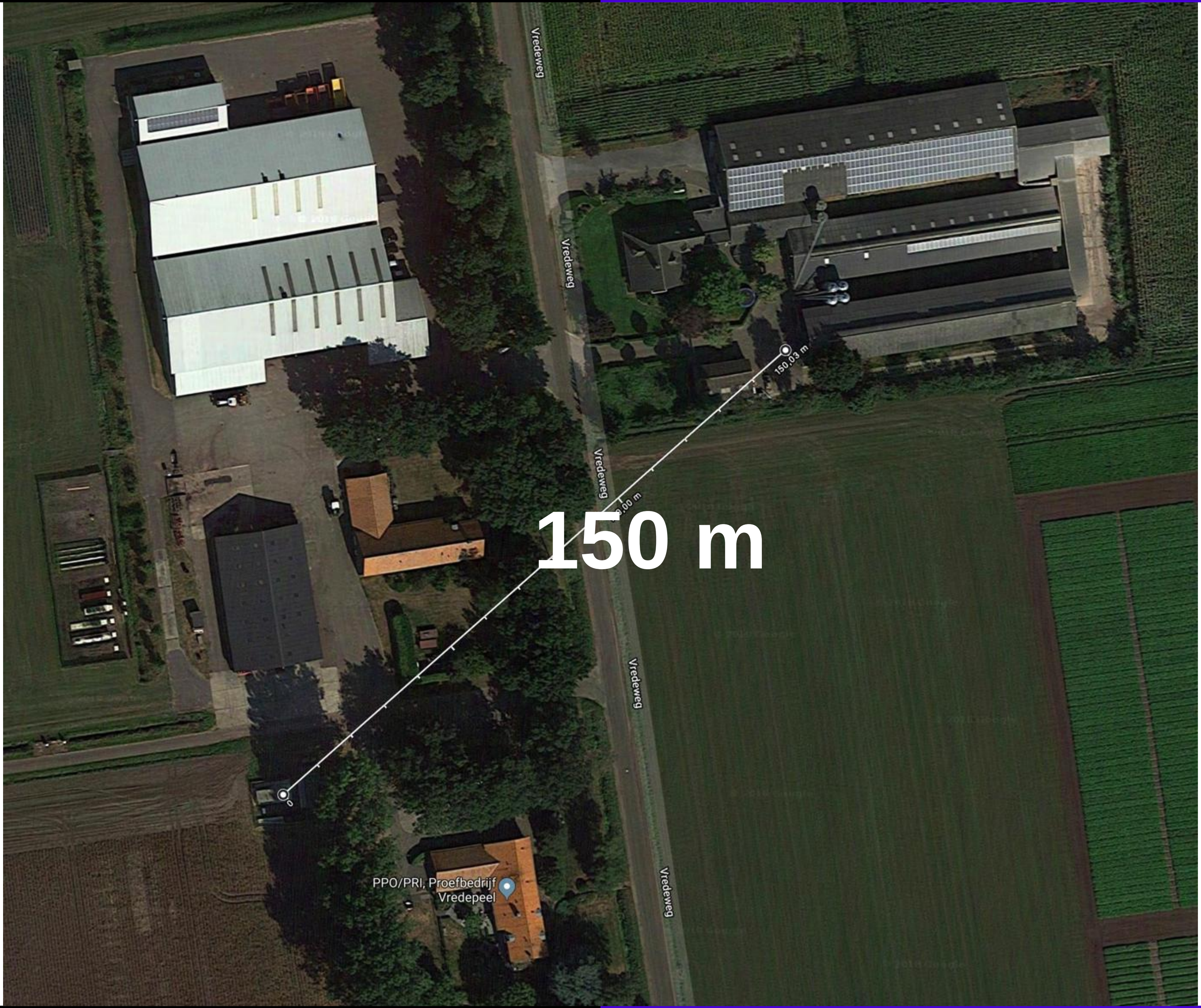
“De mogelijke direkte beïnvloeding door lokale bronnen bij concen-tratiemetingen moet zo gering mogelijk zijn. Uitgangspunt hierbij is een afstand van **300-500 m tussen meetpunt en puntbron of oppervlaktebron** afhankelijk van de grootte van de bron.”

Boermans, G.M.F., Erisman, J.W. 1990. *Meetstrategieontwikkeling voor het representativiteitsonderzoek als onderdeel van het additioneel meetprogramma ammoniak; fenomenologie van NH_3 en meetritsimulaties,* RIVM. Rapport nr. 222105001, p. 13.

“***Locations for representative-ness measurements*** - For the distance to local emission sources, these locations are selected in the same way as the LML stations. This means **at least 300 m from local ammonia sources in emission- and average areas**, ….”

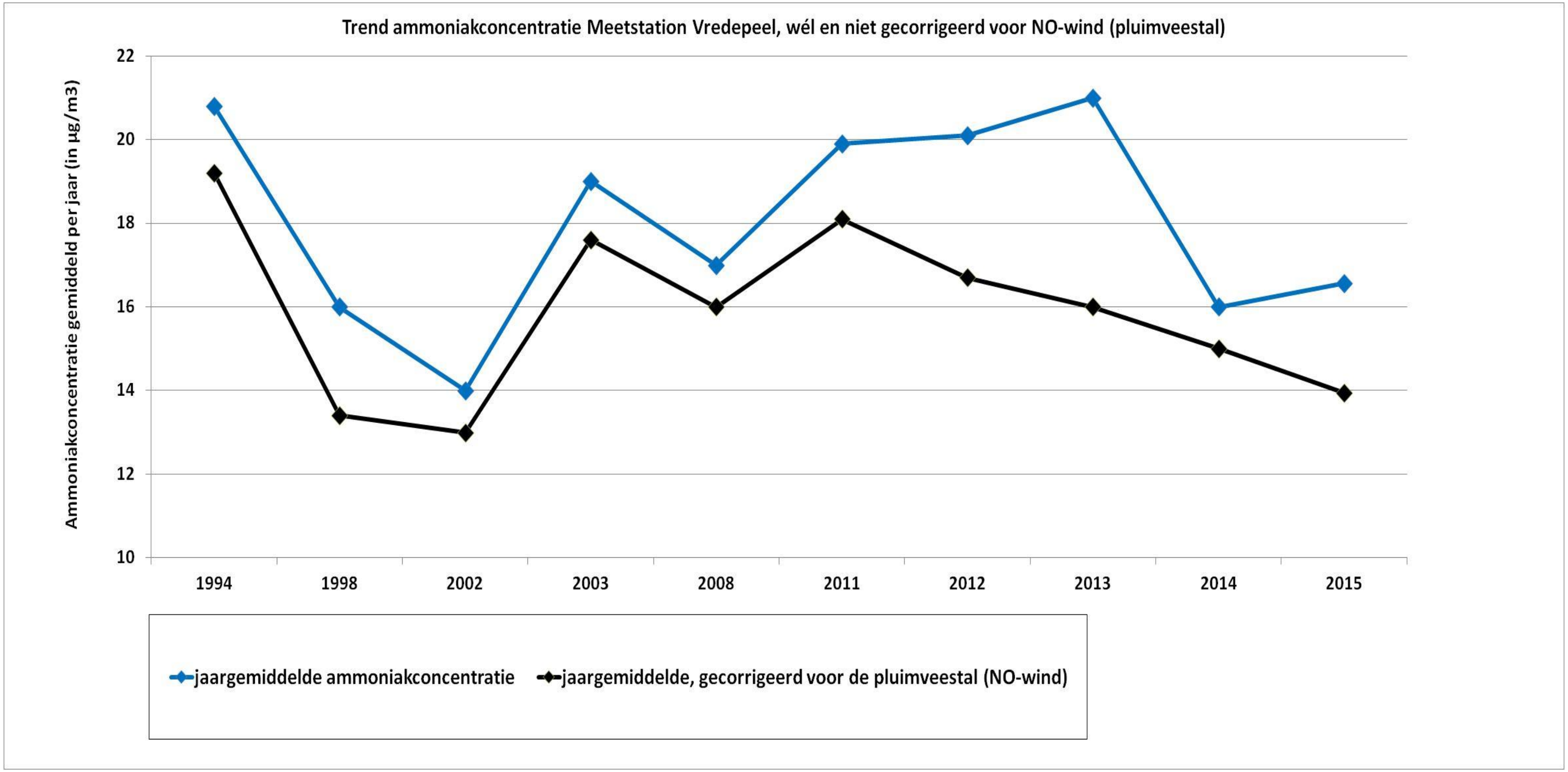
Van Elzakker, B.G., Buijsman, E., Wyers, G.P., Otjes, R.P. 1995. *The measurement of ammonia in the National Air Quality Monitoring Network (LML): (1) instrumentation and network set-up.* In: G.J. Heij and J.W. Erisman (Editors). 1995. *Acid Rain Research: Do we have enough answers?* Elsevier.

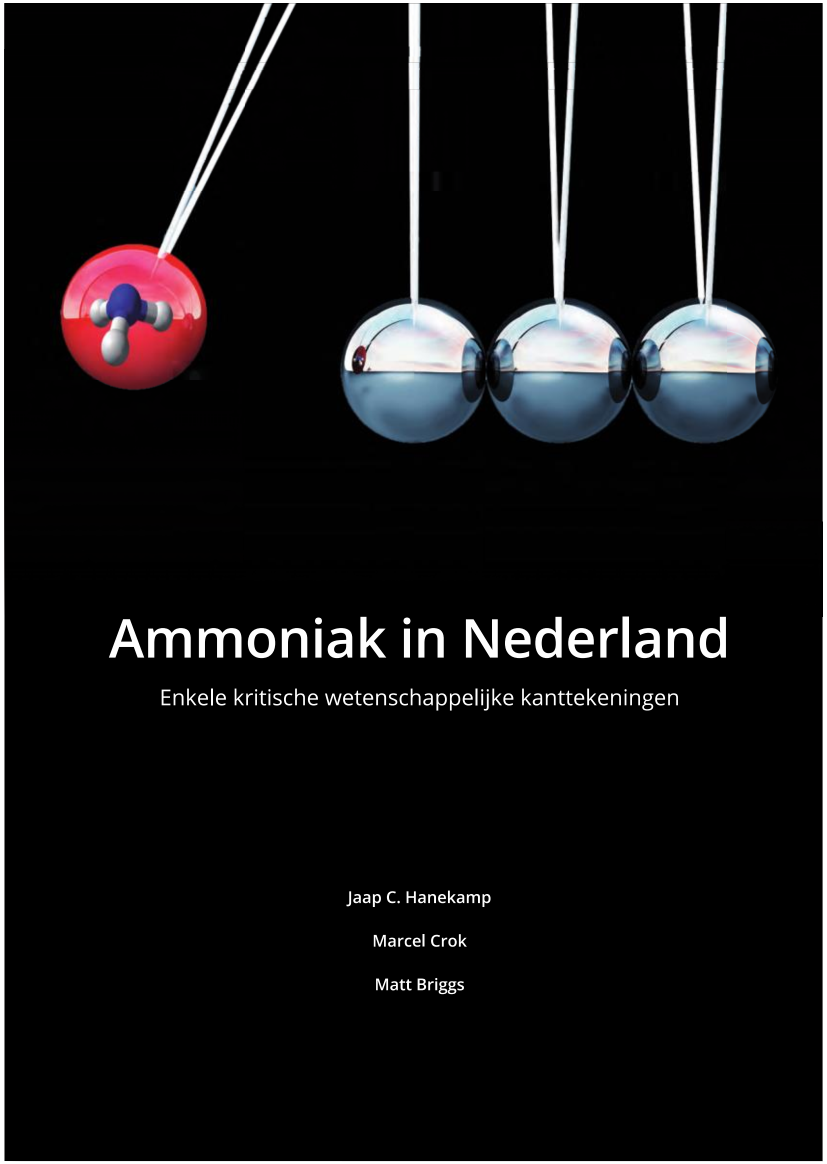
Locatie Vredepeel



1. Het ammoniakmeetstation staat (van begin af aan) veel te dicht op een pluimveestal (**volgens eigen methodologie van RIVM en ECN**)
2. Bij het bemesten wordt het meetstation min of meer ‘meebemest’ (persoonlijke observaties)
3. De meetserie verkregen bij Vredepeel wordt niet gecorrigeerd voor deze directe bronnen

De ammoniakconcentraties worden ‘opgeplust’ tot 25% (in winterperiode tot 30%) als de pluimveestal-emissie wordt meegenomen





Soil Use and Management

Soil Use and Management, June 2017, **33**, 276–287 doi: 10.1111/sum.12354

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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LETTER TO THE EDITOR

Response to van Pul, van Zanten and Wichink Kruit

Pul *et al.* offer a critique of our recent contribution in Soil Use and Management. Regrettably, their critique contains a number of confusing statements and unstated assumptions, some of which we identify here.

Van Pul *et al.* state that the 'statistical correlation between hourly ammonia concentrations between regional measurement stations is weak', which, as we pointed out, is true. They ascribe this weakness to 'large variability in local agricultural practice and in weather conditions', which is also true. They then say: 'If data are aggregated to longer timescales, correlations between stations clearly increase', which happens, but they claim this increased correlation is 'due to the removal of noise at the hourly timescale'.

First, the 'removal of noise' logically assumes that their statistical model is more real than the measured values. By implication, the measured values seem to be viewed by Van Pul *et al.* as a corrupted form of reality in the sense there is a 'true' value to which 'noise' is added, and that their statistical model can discover this 'true' value. This clearly is false. Instead, the sum total of all causes results in certain ammonia concentration at time x and location y and recorded as *n* $\mu g/m^3$. These measured values are the *experienced* values; that is, what is experienced is not some mysterious truth after which noise has somehow been subtracted. The subsequent measured data are the actual values. As we made clear in our study, no assumptions are made by us concerning the data from the Dutch National Air Quality Monitoring Network (LML).

This brings us to our second point: that data aggregation increases correlation. What Van Pul *et al.* ignore, and we pointed out (Hanekamp *et al.*, 2017, p. 284), is that 'smoothing the data' does not entail increased causation. As an experiment, take any two sets of numbers, wholly unrelated to each other, sampled at some finite timescale. Clearly, the correlation between the two series will be quite small. Then, aggregate the series by averaging (at least two) time points. The correlation will, on average, increase (i.e. the absolute value). Aggregate again at even coarser levels and the correlation increases once more and may reach 'significance'. Aggregation is a form of statistical smoothing, and smoothing any two (or more) series increases their correlation, on average, as Briggs (2016, p. 242–243) formally proved. Thus, how much of the correlation in the ammonia series Van Pul *et al.* report is due to the artefacts of smoothing, and how much is capturing larger-scale causes is unknown. Yet, Van Pul *et al.* simply assume, without proof, the latter to be the dominant, or indeed the only source of the increased correlation.

Van Pul *et al.* next state that 'annual concentrations at the various stations over the period 1993–2014 tend to be remarkably similar when normalized for the station's concentration level.' By implication, they were *not* similar *until* after statistical manipulation. There are, of course, models that help characterize uncertainty in heterogeneous time series like this, such as hierarchical modelling, which might suggest larger-scale correlations, but, of course, *never* causes. But even without these, it is obvious from the data themselves that local causes dominate larger, more regional causes. Otherwise, there would no need for statistical manipulation.

We reiterate that the mean is an inadequate value to represent average exposure. Indeed, ecosystems do not experience average exposures at all, but a continuum of differing concentrations in time, of which the high values are short-lived, as is clear from the LML-data. Indubitably, it would be best not to use point measures at all for complex phenomena and use distributions. If the interest were in total deposition, then it would be better to form a real measure of total deposition and not use a proxy that gives undue weight to temporarily high values, which *does not* represent average system behaviour (Galton, 1907).

Van Pul *et al.* claim 'annual concentrations of ammonia … appear to be only piecewise linear' over various times. They then suggest that 'most stations have the largest positive trends somewhere after the year 2000.' and indicate ways of statistically handling these pieces. This we consider to be their greatest mistake. First, the periods Van Pul *et al.* chose were biased and *not* with respect to known causes. Anyone can pick arbitrary periods. Second, statistical models are not needed to say whether a trend was present or absent in any set of data. All one needs is a definition of 'trend' and then just observe the data. Does 'trend' mean more 'ups' than 'downs', or greater averages at the end than the beginning, or so many per cent more 'ups', or higher or lower at the end? Many more definitions of trends can be proffered. Even if the definition is the trend coefficient of (say) a linear regression model, then whether that 'trend' is 'significant' carries no meaning. It is certainly not an indication that some linear cause was in effect, as the data themselves by definition never harbour causal relations (Briggs, 2014, 2016). No: something *causes* each data point, and if the causes are unknown, which clearly is the case here, then, statistical models are *only* worthwhile for making predictions of what has *not* happened yet (Briggs, 2016).

Put differently: What makes the time periods noted by Van Pul *et al.* the 'correct' periods? They simply observed these periods after all. Why not, say, 2001 to 2002, or 2002 to 2003? Was there a trend in any of these series?

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LETTER TO THE EDITOR

Comments on 'A volatile discourse- reviewing aspects of ammonia emissions, models and atmospheric concentrations in The Netherlands' by Hanekamp Briggs and Crok

Recently, J.C. Hanekamp, W.M. Briggs and M. Crok have published 'A volatile discourse- reviewing aspects of ammonia emissions, models and atmospheric concentrations in The Netherlands' in Soil Use and Management (Volume 33, pp. 276–287).

Two important statements in this article are (1) hourly concentrations of ammonia cannot be aggregated to annual averages, and (2) concentrations are local so no representative countrywide average can be determined. Both statements lead them to conclude that no trends can be detected over the period 1993–2014. Furthermore, Hanekamp *et al.* state that given the nature of the concentration distribution throughout the year, the median should be taken as a representative yearly value and not the mean. In their opinion, using the mean leads to an overestimation of the ammonia concentration (and deposition).

We disagree with these three statements as explained below.

The statistical correlation between hourly ammonia concentrations between measurement stations is weak due to large variability in local agricultural practice and in weather conditions. If data are aggregated to longer timescales, correlations between stations clearly increase due to the removal of noise at the hourly timescale. It is important to notice that due to the residence time of ammonia of several hours, not only a local signal is measured in the hourly data, but also a regional signal.

Analysis by Van Zanten *et al.* (2017, Figure 4) clearly shows the course of the annual concentrations at the various stations over the period 1993–2014 to be remarkably similar when normalized for the stations concentration level. This leads to the conclusion that the mean of the normalized time series of the eight stations is representative for the behaviour of ammonia in the Netherlands and can be used to monitor the effectiveness of emission reduction policy.

The exposure of man or ecosystems to air pollutants is best represented by cumulative exposure over long periods, in which case the mean value is the most suitable. So in the field of air quality, it is very common or even obligatory (e.g. in the EU Air Quality Directive, 2008) to use the mean. In European and national trend studies for ammonia and other air pollutants, monthly and annual means are used (e.g. Bleeker *et al.*, 2009; Colette *et al.*, 2016). The causal reason why we use the mean is because we have to account for all emission or deposition events. The median is, by definition, less sensitive to higher and lower values. Thus, the median would discard high emission events (that for instance occur during manure application) and would underestimate deposition on ecosystems. Therefore, the conclusion by Hanekamp *et al.* that using the mean leads to an overestimation of the concentration or deposition is wrong.

Van Zanten *et al.* (2017) clearly showed that the time series of the annual concentrations of ammonia do not vary linearly in time over 1993–2014 but appear to be only piecewise linear. There is a clear decay in the concentrations from 1993 up to around 2004 and an increase from thereon. Therefore, in the statistical analysis on trends in ammonia, these periods should be considered separately when using linear regression. Hanekamp *et al.* largely neglect this fact. That the concentrations do not vary linearly in time is actually also visible in Figure 4 in Hanekamp *et al.*, which shows most stations to have the largest positive trends somewhere after the year 2000. When the full period is considered, the trend at most stations, however, becomes negative or close to zero again, which consequently implies a negative trend in the first period. This would have been clearly visible if Hanekamp *et al.* would have extended their trend analysis using a fixed starting date instead of only using a fixed end date. It is therefore erroneous to state that 'no countrywide trend signal is evinced, and neither is there any clear indication of a consistent trend at any station'. The decay in the concentrations from the first half of the time series and the increase from thereon can be seen as well in Figure 6 of Hanekamp *et al.* in which allegedly monthly data are depicted (actually the data show annual values with the 2015 values being incorrect). Note that the median follows more or less the same pattern as the mean. So using the median instead of the mean would not considerably affect the conclusions on the trends.

Besides this, Hanekamp *et al.* neglect the importance of atmospheric chemistry processes in their study. It is well known that ammonia chemically reacts with sulphur dioxide and nitrogen dioxide in the atmosphere. Due to the large decrease in particularly sulphur emissions in the 1990s, the trend in ammonia concentration is influenced (e.g. Sutton *et al.*, 2003; Wichink Kruit *et al.*, 2017). Moreover, due to the decrease in sulphur dioxide concentrations, dry deposition of ammonia has decreased. Both processes mask trends in ammonia concentration and have quite substantial effects (Wichink Kruit *et al.*, 2017).

Overall, the study of Hanekamp *et al.* comes with a large number of assumptions and conclusions that are questionable

"… **high emission events** (that for instance occur during manure application) [should not be discarded as not to] **under-estimate deposition on eco-systems.**"

LML station Vredepeel (Van Zanten et al. 2017. *Atmospheric Environment* 148: 352-360)

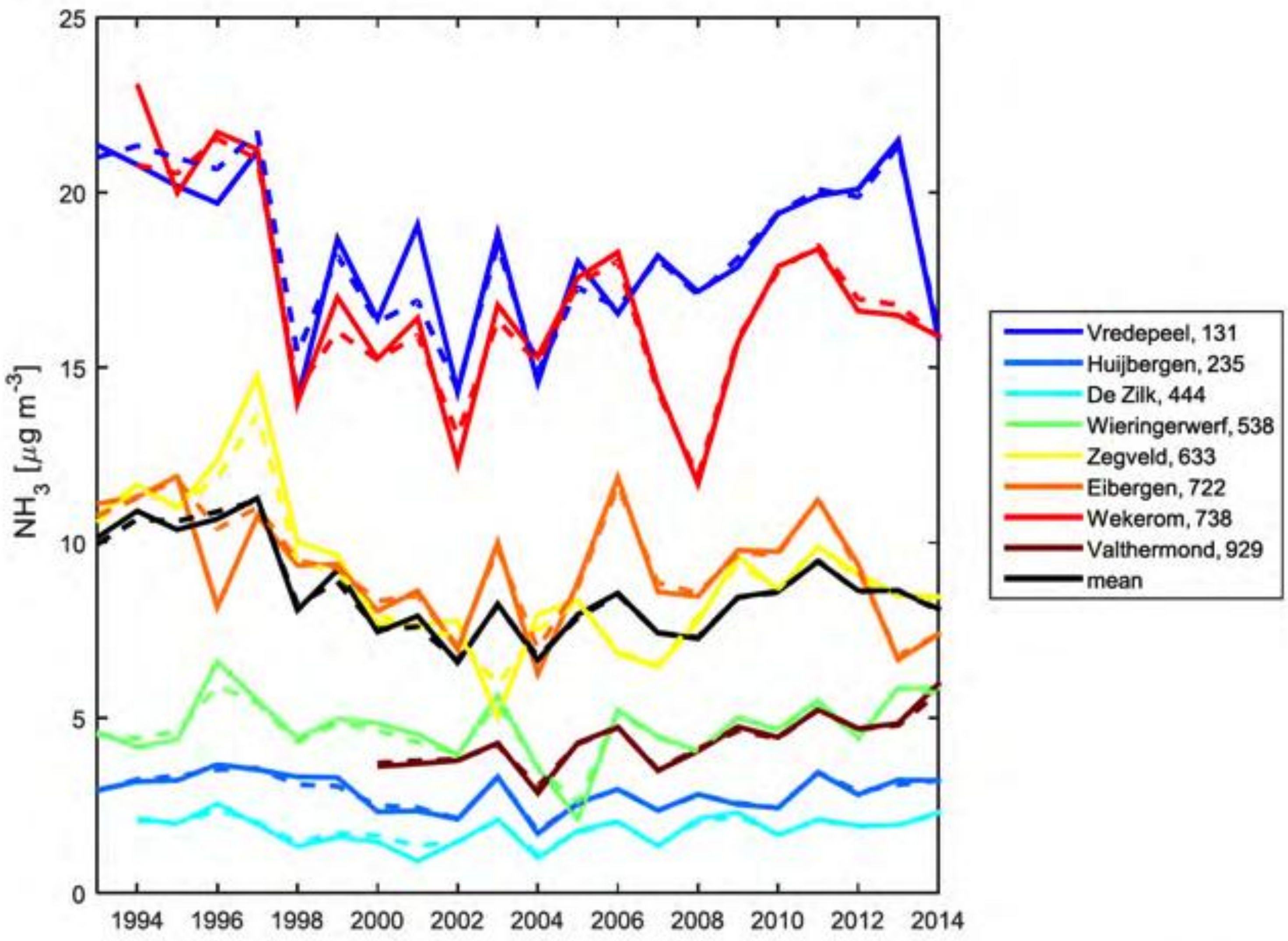

Fig. 3. Time series of the annual mean ammonia concentrations at the eight LML monitoring stations. The continuous lines denote the original dataset while the dashed lines refer to the gap filled dataset.

 Van Zanten et al. 2017. *Atmospheric Environment* 148: 352-360

"A large variability in the mean concentrations from year to year is also visible. This is most clearly for the two monitoring stations (**Vrede-peel** and Wekerom) in high emission areas: several changes of the order of 5 [mg/m^3] occur from year to year. **This is most likely due to changes in local emission sources, but meteorological influences can play a role as well**."

1. Het RIVM neemt dus, in hun reactie op ons SUM-studie, **zonder melding van de foutieve plaatsing van LML station Vredepeel**, een lokale bron mee om “ammoniakdepositie niet te onderschatten”
2. Van Zanten *et al.* (2017) doet hier het zwijgen toe: de pluimveestal wordt niet genoemd
3. De Brabantse “Rapportage van de lucht-kwaliteit gemeten in De Peel van 2008 tot en met 2015”, waarin LML station Vredepeel een centrale rol speelt, **geeft verder inzicht in observaties 1 en 2**

Rapportage van de luchtkwaliteit gemeten in De Peel van 2008 tot en met 2015. Rapport no. 4257342, 8 maart 2016

“Met behulp van windroosanalyses is de bijdrage van PM_{10} en NH_3 bepaald vanuit het LOG (landbouwontwikkelingsgebied) op de meetstations Blaarpeelweg en Vredepeel. Deze rapportage beschrijft de resultaten van de metingen van 2008 tot en met 2015.”

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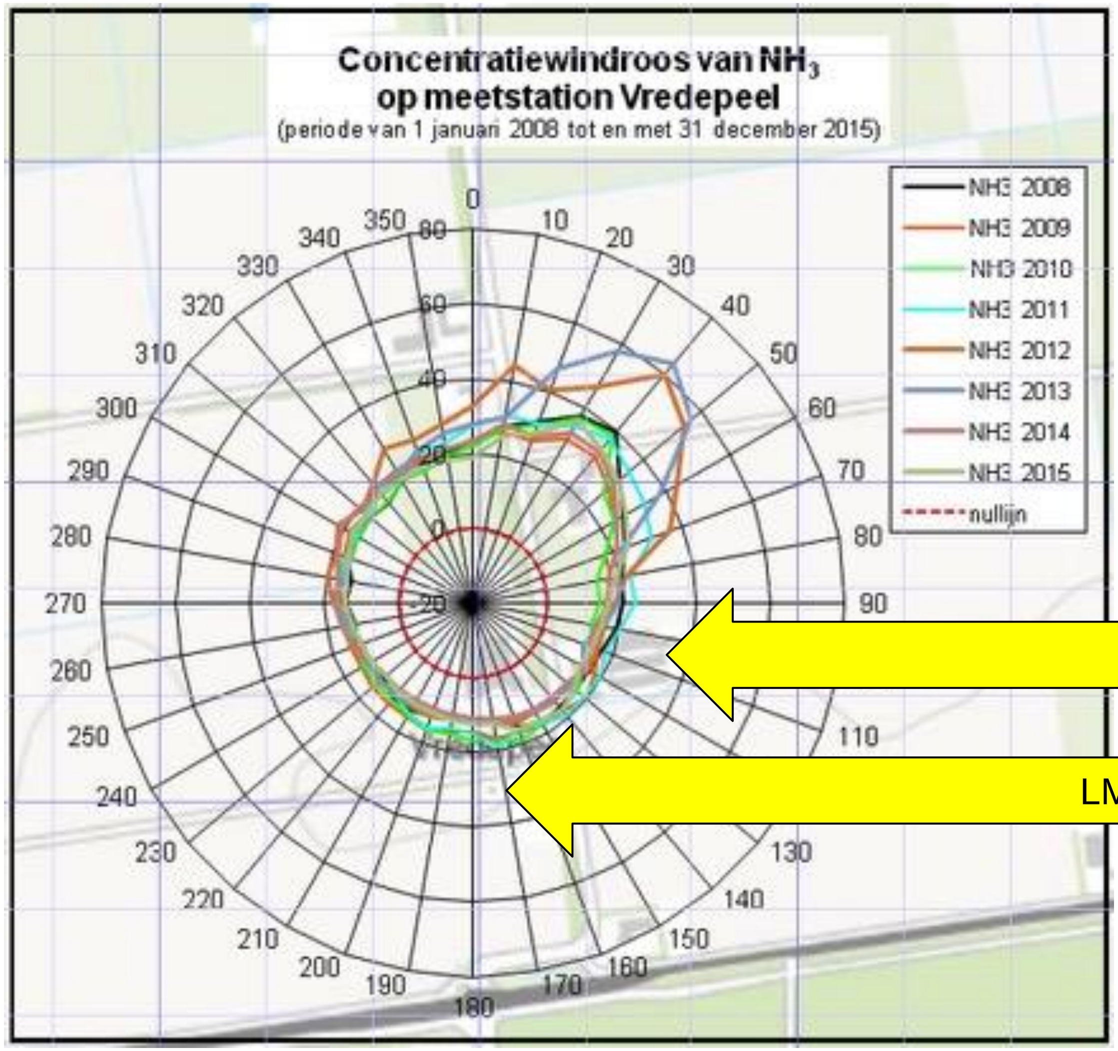
Rapportage van de luchtkwaliteit gemeten in De Peel van 2008 tot en met 2015. Rapport no. 4257342, 8 maart 2016



Provincie Noord-Brabant
Cluster natuur &milieu
Postbus 90151, 5200 MC 's Hertogenbosch, p. 23

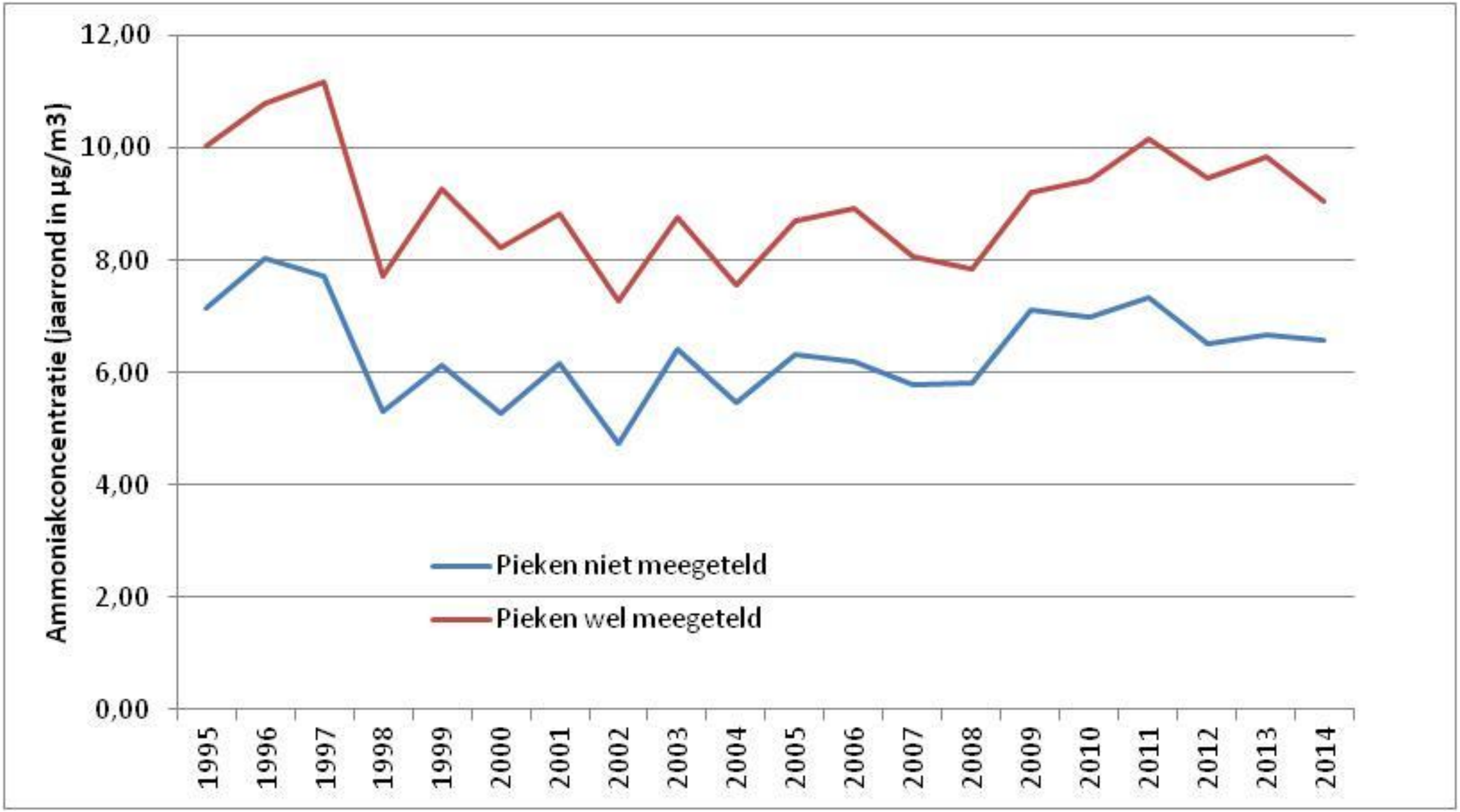
- Publicaties over de ammoniakmetingen op Vredepeel maken geen melding van de naburige pluimveehouderij, **ondanks dat deze veel te dicht op het meetstation staat**
- De “Rapportage van de luchtkwaliteit gemeten in de Peel van 2008 tot en met 2015” van de Provincie Noord-Brabant past de LML locatie Vredepeel zelfs aan op de kaart zodat deze bron buiten beeld blijft
- Dat wil zeggen: de provincie heeft het meet-station niet op de feitelijke locatie ingetekend, maar noordelijker

Rapportage van de luchtkwaliteit gemeten in De Peel van 2008 tot en met 2015. Rapport no. 4257342, 8 maart 2016, p. 6

"**De metingen op het meetstation Blaarpeelweg zijn op 1 april 2012 beëindigd. Dit had te maken met de oprichting van een nertsenbedrijf in 2011 op korte afstand aan de oostkant van het meetpunt.** Aanvankelijk was het plan het meetstation te verplaatsen naar een andere locatie. Echter dit leverde dusdanig veel problemen op dat in overleg (RIVM en NBr) is besloten het station vanaf 1 januari 2013 weer in gebruik te nemen. **De invloed van de nertsenfarm op de ammoniak- en fijn stof concentratie op het meetstation Blaarpeelweg wordt met de windroosanalyse uitgefilterd**."

- We nemen aan dat het RIVM bekend is met de, volgens eigen randvoorwaarden, foutieve plaatsing van LML station Vredepeel. Waarom is de locatie nooit gecorrigeerd?
- De “cartografische correctie” van LML station Vredepeel door de provincie Brabant (p. 23) is oneigenlijk. Het houdt de pluimveestal als ammoniakbron bewust buiten beeld. Uitleg van de provincie is op zijn plaats!
- En: Is het niet zo dat voor de uitschieters in de meetwaarden moet worden gecorrigeerd (zoals gedaan bij Blaarpeelweg)?

Ammoniakconcentraties, mét en zónder pieken



- De foutieve plaatsing van LML station Vredepeel wordt nergens in publicaties ter discussie gesteld, dan wel verdedigd of zelfs maar genoemd
- De ammoniakpieken veroorzaakt door de pluimveestal worden zonder uitleg én in tegenspraak met de eigen randvoorwaarden meegenomen in de meetdatasets
- De “cartografische correctie” van LML station Vredepeel door de provincie Brabant suggereert onterecht dat de ammoniapieken als landbouwachtergrondwaarden moeten worden beschouwd
- Dit alles lijkt de conclusie te rechtvaardigen dat ammoniakconcentraties en -depositie bewust worden opgeplust
- **Er is alle reden om het ammoniakonderzoek in Nederland wetenschappelijk te wantrouwen**