Review on the scientific underpinning of calculation of ammonia emission and deposition in the Netherlands

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Summary report

Key messages

- Ammonia (NH_3) is a key air pollutant for which major efforts have been made in the Netherlands since 1993 to reduce its emissions. However, NH_3 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.
- The present review has been stimulated by these issues, leading to the key question of whether the nitrogen deposition calculation on a regional level is a good scientific approach for the real deposition that occurred.
- The review has addressed several associated questions related to the quality of emissions data and calculations, of atmospheric monitoring, and of atmospheric dispersion and deposition modelling approaches. Finally, the Review Panel was asked for its own explanation of the divergence in trend between calculated ammonia emissions and measured ammonia concentrations.
- **On the whole, the methods used in the Netherlands for emission estimation, measurement and modelling of atmospheric ammonia are generally sound. However, there are concerns regarding the analysis of long-term trends, especially given that current trends are now starting to flatten out following implementation of the most effective measures.**
- Specifically, the Review Panel concludes that:
  - **Emissions of ammonia have decreased since 1990 by less than the values of 64-70% currently estimated by Dutch researchers.** The real reduction may be closer to 40-60%. Several factors suggest that emissions for 1990 are overestimated (e.g. winter spreading, uncertainties in excretion and grazing emissions), while present-day emissions may be underestimated due to sub-optimal use of low emission techniques by some farmers (e.g. incomplete slurry injection).
  - **Recent closure of two out of eight intensive ammonia monitoring sites will compromise future assessment of long-term trends.** Innovative new technology currently being implemented at the intensive sites now allows cost savings that should be exploited to re-instate the recently closed (LML) monitoring sites. The current method applied in the extensive ammonia monitoring network (MAN) uses passive samplers that are not state-of-the-art, with more sensitive and precise methods being available. More effort should be made to analyse and interpret the network data from both the intensive and passive sites to understand trends (e.g. diurnal differences, trends in import from neighbouring countries, characterization of the story behind the temporal patterns at each monitoring site).
  - **Dutch dispersion and deposition modelling with the OPS model is not currently adequate for the purpose of assessing long-term trends in ammonia concentrations.** This is mainly because the coupling between ammonia, sulphur dioxide and nitrogen dioxide is treated too simplistically, but also because the effects of meteorological variability and climate change are not fully incorporated. Current efforts to address such model limitations need to be strengthened.
  - **Dutch dispersion and deposition modelling can be considered as well fitted for assessment of regional spatial patterns.** In particular, the Netherlands is to be

congratulated for its recent development of the AERIUS tool to support future local decision making.

  - **As SO_2 and NO_x emissions have decreased, NH_3 now has a longer lifetime in the atmosphere.** This is part of the explanation for why NH_3 concentrations have not decreased as fast as the calculated trends in NH_3 emissions. Together with other dispersion interactions, the Dutch OPS model appears to be able to explain around half of this difference. Future efforts should more strongly emphasize the coupling of ammonia with trends of ammonium in particulate matter ($PM_{2.5}$) and ammonium in precipitation.

- If improvements in next-generation atmospheric modelling are combined with necessary inventory corrections and improvements in monitoring, it is anticipated that the Dutch scientists should be able to largely close the Ammonia Gap within the next 3 to 5 years.
- Such an improvement in the present situation can, however, only be expected with appropriate resources. **The Panel noted that there had been a much weaker level of integration, synthesis and uncertainty analysis than they had expected based on the previous world-leading position of Dutch ammonia scientists**. This appears to reflect the much less stable resource to support ammonia science in the Netherlands over the last decade.
- There are major economic opportunities for the Netherlands in relation to its achievement in reducing ammonia emissions. This comes especially as other countries increasingly recognize the environmental and health impacts of all nitrogen compounds, including ammonia.
- **Emerging international efforts to reduce ammonia emissions therefore put the Netherlands in a strong position, enabling it to capitalise by sharing technologies and expertise.** To achieve this, however, will require sustained investment in innovative farming practices, measurement techniques and the underpinning ammonia science.

Background

1. International application of models and comparisons of measurements show that ammonia (NH_3) concentrations in the Netherlands are among the highest in Europe. This is a consequence of the high density of agricultural activities in the Netherlands, especially livestock agriculture. These emissions combine with emissions of nitrogen oxides (NO_x), mainly from transport and combustion sources, to increase rates of nitrogen deposition. As a consequence, there are significant threats from both high ammonia concentrations and nitrogen deposition in the Netherlands. These threats include loss of ecosystem integrity and biodiversity (especially for Natura 2000 sites), while also adding to the burden of particulate matter (PM) in the atmosphere, which is a significant threat to human health.
2. Ammonia losses can represent a substantial fraction of the available nitrogen added to farming systems. For example, emissions of ammonia during surface spreading of manure can lose more than half the available nitrogen to the atmosphere (based on total ammoniacal nitrogen, TAN). Ammonia losses are therefore also a contributor to reducing nitrogen use efficiency (NUE) in agriculture. While reducing ammonia requires farmers to take extra care, it can therefore improve resource use efficiency, while helping to reduce a major environmental problem.
3. In 1993 the Dutch government instituted a policy to reduce ammonia emissions from agriculture. This focused especially on requiring farmers to use "low-emission" techniques for manure spreading and requiring slurry stores to be covered. Since that time, policies have increased requirements to use low-emission animal housing. As a result of these policies, ammonia emissions are expected to have reduced substantially since 1993.
4. In the late 1990s it was noticed that monitored atmospheric ammonia concentrations in the Netherlands had not decreased in-line with emissions. This difference was called the "Ammonia Gap". Subsequent international reviews showed that there were significant differences between the trends in the different ammonia components of the atmosphere, and that these differences could also be seen for other countries (e.g. UK, Denmark, Hungary and others; see e.g. A. Bleeker *et al*., in "Atmospheric Ammonia", Springer 2009).
5. Emissions of gaseous NH_3 lead to more than one kind of nitrogen component in the atmosphere. Chemically, NH_3 reacts to form ammonium (NH_4^+) ions, producing salts such as ammonium nitrate and ammonium sulphate, which are present as fine particles in the atmosphere (e.g. particulate matter smaller than 2.5 μm diameter, $PM_{2.5}$). Clouds, rain and snow scavenge gaseous NH_3 and particulate NH_4^+ from the atmosphere. This reactive nitrogen in precipitation is called "wet deposition". The other removal process is when the gases and aerosol are directly absorbed by the ground, which is called "dry deposition".
6. The relative share of ammonia emission that is present in the atmosphere as gaseous NH_3, particulate NH_4^+ and wet/dry deposition also depends on other factors. In particular, emissions of sulphur dioxide (SO_2) and nitrogen oxides (NO_x) produce acids in the atmosphere which speed up the rate of particulate ammonium formation. This means that analysis of trends over time needs to distinguish between trends in gaseous NH_3, particulate NH_4^+, concentrations in rain, and wet and dry deposition. It also means that these ammonia-related trends depend on parallel trends in the SO_2 and NO_x emissions. As SO_2 and NO_x emissions have decreased over Europe since 1990, this means that NH_3 now has a longer life-time in the atmosphere.
7. While the first Ammonia Gap concerned a difference between estimated emissions and monitored trends, a second Ammonia Gap was identified in the mid-2000s. At this time, it seemed to Dutch scientists that the decreasing trend in ammonia could be understood, but there

was a significant difference between model estimates and measurements: measured concentrations were larger than estimated by modelling.

8. The Dutch ammonia gap now seems to be entering a third phase. Analysis of the atmospheric NH_3 monitoring data shows that measured concentrations did not decrease much since 2000. This points to the need to understand the trends in emissions and concentrations better. Further component questions emerging include:
   a. Is the absolute magnitude of ammonia emission in the Netherlands correct (as currently estimated for recent years)?
   b. Is the estimated trend in ammonia emissions in the Netherlands correct? For example the NEMA model (Velthof *et al.*, 2012, *Atmospheric Environment* **46**, 248-255) indicates that ammonia emissions decreased by 64-70% from 1990-2013. Is the reduction overestimated? (The two-page paper by Groenestein for this review states a 70% reduction, while a value of 64% may also be derived from the values submitted to EMEP, see Appendix to this report.)
   c. Whatever the exact value, has the Dutch ammonia policy been successful in achieving a substantial reduction in ammonia emission?
   d. Does the explanation of the difference between estimated emission and measured ammonia concentrations vary in different parts of the Netherlands?
9. These questions provide perspective on the main questions identified by the Dutch Government. We therefore also consider these points while addressing the main questions.

Answers to the questions posed by the Dutch Government

Question A.

10. **Is the nitrogen deposition calculated on a regional level a good scientific approach for the real deposition that occurred?**

11. ***Summary: Overall, Yes, however there are specific concerns. Broadly speaking the models are sound. However, there are discrepancies between the modelled results and the measurements which have not yet been resolved.***

12. The modelling approach based on the OPS model, is well fitted for spatial analysis, but has key weaknesses for assessment of long-term trends. We see opportunities for a more integrated synthesis of the obtained results which have not yet been grasped. This suggests a need to engage more strongly the wider Dutch research community in analysis of the data collected.
13. The Panel is aware that limited resources have led to intermittent and reduced research in ammonia over the past decade. Recent initiatives have started to respond to the 2013 Ammonia Review, which are beginning to address the resulting loss of intellectual capital. However, sustained long-term investment will be needed if the Netherlands should retain its world-leading position as an innovator in this area.

Question A.1

14. **Is the calculation of the emission in the Netherlands (on national and regional scale) scientifically sound?**

15. ***Essential Summary: On the whole, Yes. The principle of using Total Ammoniacal Nitrogen (TAN) flow as the foundation of the NEMA model is good, following the EMEP/EEA air pollutant emission inventory guidebook recommendation, but we see uncertainties which arise both from emission factors and activity data (input statistics on livestock, cropping, nitrogen management practices etc.***).
16. ***These uncertainties propagate when considering the trend between 1990 to 2013. Based on the information presented to the Panel, we conclude that the actual reduction in ammonia emission in the Netherlands is less than the 64%-70% reduction reported for the period.***

17. At the request of the Panel, the Dutch emissions inventory team calculated 'implied emissions factors' for ammonia in the Netherlands. This is the average rate of emission per animal found by dividing total emissions by animal numbers (see Appendix Table).
18. The Panel noted several uncertainties in these implied emission factors which would result in uncertainties in the overall emission trend:
    - The annual ammonia emission factor of 64 kg NH_3 for a dairy cow in 1990 appears to be exceptionally high, compared with other NW-European countries, and needs further verification.
    - The table shows substantial reductions in emission since 1990 for livestock categories (e.g. horses, sheep, rabbits) even when such reductions were not expected, since the Dutch ammonia policy would presumably have had little impact on emissions for these categories.

19. The Panel recommends that the Dutch research community should further develop process-based approaches to ammonia emission estimations to enable the key drivers on emission to be incorporated in order to cope better with short-term meteorological and long-term climate and spatial soil differences. The Panel notes that international evidence indicates that a warming climate will increase ammonia emissions. Future modelling should account for these effects.

Housing and excretion

20. The NEMA model may overestimate the protein content in grass grazed by cattle, sheep and goats in the 1990s and thus overestimate the amount of nitrogen in animal manure (see Appendix 1).
21. The Panel was impressed by the high quality and level of detail of farm level data on housing systems and feed input. The information available in the present situation is allowing a level of detail in calculating the emission and spatial distribution which is world leading and is only possible in a few countries.
22. *However, there is an important discrepancy concerning the ratio of [TAN]:[Total N] when comparing the NEMA model with many measurements of manure at the time of spreading.* This discrepancy, which has not yet been explained, leads us to conclude either: a) that the excreted TAN in the model is an overestimate (and hence the ammonia emission from manure spreading, by 5-10%) or b) that there is a missing source of nitrogen loss from the housing system (e.g. as N_2 or NH_3), or c) that there is an unaccounted-for immobilization of TAN to organic N.
23. *There is uncertainty about the activity data when going back to 1990 because less activity data were gathered for the earlier period.* The Panel considered that there is a need for more transparency in the assumptions about feed intake over time and the splitting up over different areas of the Netherlands. The Panel also considered the estimated nitrogen uptake and TAN excretion by livestock to be very high in 1990. More information is needed to justify this.
24. The Panel noted evidence presented that naturally ventilated cattle buildings are more open today than in the early 1990s. It is understood that the extra ventilation will have increased the emission factor for ammonia. However, this effect does not appear to be included in the Dutch inventory and would warrant further testing.

Manure spreading

25. *The Panel concluded that the ammonia emission factor for manure spreading for the period around 1990 was likely to be an overestimate.* This means that the actual reduction in ammonia emission since 1990 is expected to be less than currently shown in the NEMA model:
    a. In the early 1990s a larger fraction of slurry was spread to fields during winter-time, while in more recent years a larger fraction of the slurry is spread in spring and summer-time.
    b. At the request of the Panel, the LML network data were analysed to compare the seasonal pattern in NH_3 concentration in the 1990s with recent years. This confirmed that there has been a change in the seasonal pattern of ammonia emissions, with autumn peaks more prominent in the 1990s, while spring peaks are more prominent for recent years, which can be attributed to policies on manure application timing.
    c. It is known that the ammonia emission factor for manure spreading is smaller in colder, winter conditions. However, the NEMA model does not account for this effect of seasonality in manure spreading.

d. The NEMA inventory estimates for manure spreading are mostly based on measurements using the Integrated Horizontal Flux (IHF) method. Based upon the evidence presented, the Panel is of the opinion that this could overestimate emissions by 0-20%, due to uncertainties with instantaneous turbulent fluctuations and, to a much smaller extent, due to uncertainties in the zero plane replacement. Since manure spreading contributed a larger share of total ammonia emissions in the early 1990s, this will propagate to a small over-estimation of emissions for 1990 relative to 2013.

26. The Panel noted that there is a remaining question about the "oasis effect" on ammonia emissions measurements. According to this effect, a small emission area will have a larger emission factor than a large emission area, as a build-up of concentrations in a larger area tends to reduce emissions. This would imply that experimental studies using small plots lead to larger emission factors. However, insufficient evidence was presented to demonstrate that this effect is of quantitative significance in the Dutch ammonia emissions inventory. Further research and peer review publications on this topic are needed.
27. The Panel recognized that such simplifications of the IHF method apply for all countries in Europe who use this method. Current advantages in technology in ammonia emission measurements should be fully exploited to further develop the state-of-the-art in this area.
28. *The Panel also noted evidence presented that not all farmers implement the required injection approach for manure spreading to the standard intended. Evidence from a survey indicated a lower level of ammonia abatement was achieved compared with that assuming application of best practice according to the manure spreading requirements.* This differential is not currently included in the NEMA model, which therefore is expected to under-estimate ammonia emissions for recent years. Further farm surveys should be made to monitor improvement in implementation of the policy, and to allow quantitative temporal estimates of this effect to be included in NEMA.

Other ammonia sources

29. The Panel noted the high uncertainty in the temporal trend in the emission factor for grazing animals. While the basis from the original measurements appears to be sound, these reflect studies in the late 1980s under high grazing intensities. The Panel considers the extrapolation from the original data too uncertain for implying such a strong reduction in the grazing emission factor. The currently estimated emission factor of 2% is even lower than the emission factor from ammonium nitrate fertiliser. Further data from a wide range of grazing situations (including extensive grazing and other livestock) should be integrated and extended to provide a more robust estimate that does not require such extrapolation.
30. The Panel noted a high uncertainty in ammonia emission for the new crop residue emissions recently included in the inventory. Some crops are not included (e.g. oil seed rape, *Agric. Forest Meteo.* **105**, 327-445) while further experimental underpinning is warranted.
31. We note that the ammonia emissions for fertilizer application are based on few data from NW Europe. As ammonia emissions from urea are larger than other fertilizers, the increase in urea use over recent years appears to be offsetting gains from decreased fertilizer use. However, insufficient evidence was presented on the extent to which the recent increase in urea use is associated with low-emission urea products (e.g. with urease inhibitors). Such product differences need to be incorporated into the NEMA inventory.

32. The Panel was surprised by the high emission from some non-agricultural sources such as human sweat and breath. Although the panel recognized that the Netherlands has a detailed internet-based reporting system, a clear synthesis of total ammonia emissions was not presented.
33. The Panel felt that insufficient attention has been given to combine the trends from all emission sources (agricultural and non-agricultural) which would be necessary to allow easier comparison with atmospheric measurement and modelling data.

Uncertainty Analysis in Emissions

34. A partial sensitivity and uncertainty analysis of the NEMA inventory was presented to the Panel. However, *no uncertainty analysis of the trend in ammonia emissions was presented. Similarly, no overall synthesis in the uncertainty in the trend of total ammonia emissions (agricultural and non-agricultural) appears to have been conducted.*
35. Given the ongoing national debate about the Ammonia Gap, the Panel found it surprising that such a temporal uncertainty analysis in ammonia emissions had not been conducted. Work to achieve this needs to be established as a matter of urgency. Such an analysis should not be restricted to the NEMA model, but should consider the trends in all contributions to Dutch Ammonia emissions.
36. Given the need to ensure an independent view on the Dutch ammonia inventory (beyond that which is possible by this Panel), the *Dutch government should consider whether such a trend analysis should be implemented as part of independent auditing of the emissions inventory*. It is recommended that such an *independent uncertainty analysis* in emissions trends be conducted regularly, in an integrated way with trends in concentrations and deposition.
37. Overall, the conclusion of the Panel is that the Dutch ammonia policy has been successful in achieving a substantial reduction in emissions. This is based on knowledge the evidence of implementation of well-tested mitigation methods. *The Panel suspects that the emission reduction since 1990 may be around 40-60%, rather than the claimed figure estimates of 64-70%. However this indication reflects our expert judgement based on the information presented.* Inclusion of the factors discussed into the recommended uncertainty analysis is required to provide a definitive uncertainty distribution.

Question A.2

38. **Are the set-up and measurement techniques of the ammonia monitoring network scientifically sound for the determination of ammonia concentrations on a regional and national scale?**

39. ***Summary: Yes, overall the setup and techniques are sound. However there are specific concerns about the need to maintain long-term sites and to ensure consistency of measurement performance. This is essential to provide evidence to assess the long-term trend in ammonia and ammonium concentrations.***
40. ***In addition, the LML network is not fully representative of Dutch conditions (several sites are located near the border of the Netherlands and are therefore influenced by foreign sources rather than typical of Dutch conditions; cattle-dominated sites are under-represented, given the importance of cattle in the emission totals).***

41. ***We are impressed by the extensive network of passive sampling around Natura 2000 sites (MAN network). However, the measurement method used is not state-of-the-art, which compromises the usefulness of the data collected.***

42. The strategy of the Dutch ammonia monitoring network combines a small number of continuous measurement (hourly integrated) sites with a large number of low temporal resolution (monthly integrated) measurements. This is a robust approach which allows the meteorological interactions with ammonia to be investigated at a few key sites, while long-term trends can be addressed by using a larger number of sites.

High temporal resolution ammonia measurements (LML network)

43. The Panel congratulates the Netherlands on its globally unique long-term use of the AMOR monitors, which were developed in the Netherlands during the early 1990s based on the AMANDA methodology. The resulting data have been essential for assessing long-term trends in the Netherlands. However *the Panel is very concerned about the closure of two sites in 2014, which compromises the network integrity for future assessment (see further below).*
44. The Panel is encouraged to see the development and current testing of the mini-DOAS system, given its potential to improve the time-response of ammonia monitoring data, while *substantially reducing the operating costs* compared with AMOR. Peer review publications need to be made that compare the performance of the mini-DOAS with AMOR, including at least 1 year of measurements by both methods simultaneously at the LML sites.
45. With the new mini-DOAS systems being implemented, we understand that substantial cost savings in ammonia monitoring can now be made. *The Panel therefore recommends it as absolutely essential that some of the cost savings be used to allow urgent reinstatement of the two LML stations that were closed in 2014.* Re-establishing these two sites will ensure that the investment of the last 20 years in the LML network at these sites is not wasted and that trends at the full 8 site network can be continued into the future.
46. The strength of the LML network is that continuous measurements can be analysed in relation to meteorological conditions and local sources. By comparison, a network of only six sites is a much weaker basis to assess long-term trends. The Panel was therefore extremely surprised that efforts had not been presented by the Dutch scientists to characterize the meteorological interactions with ammonia concentrations, and to explore how these dynamics interact with long-term trends.
47. *The Panel strongly recommends that new efforts be placed to characterise the local context at each of the 8 LML sites to allow a more comprehensive analysis of the reasons behind the trends and other differences.* For example, at request of the Panel, data were presented for each site, with one site showing an increasing trend in ammonia concentrations. Apparently this site was in an area receiving imported manure from other parts of the Netherlands. However, it remains speculation that this was the reason for the increase. A similar issue applies to LML sites near the borders of the Netherlands. *For example, trend analysis should be conducted for different wind sectors to show if the trend in NH_3 concentrations is different for air masses passing over the Netherlands than for imported air masses.* With several of the LML sites near borders of the Netherlands, there is also a case for better cooperation with adjacent countries to establish how differing trends in estimated NH_3 emissions between the Netherlands and adjacent countries contribute to the observed trends in NH_3 and NH_4^+ concentrations and deposition.

Comprehensive analysis needs to be carried out for each LML site to assess such effects, and the Panel was surprised that this had not already been done.

48. In addition, due to the difference in the overall concentration level at individual sites the long-term national trend should potentially be based on relative trends at the sites rather than just an overall trend based on the average over all sites (i.e. giving equal weight to each site).
49. With substantial resources saved by introducing the mini-DOAS system, the Netherlands Government should also consider whether it is possible to increase the LML network further by establishing new continuous monitoring sites. In particular, this would allow better representation of sites characterized by dairy farming and sites more distant from the Dutch border would be warranted. If such an extension is implemented, it is essential that NH_3 monitoring be linked with parallel particulate matter and wet deposition monitoring.

Low temporal resolution ammonia monitoring at many sites (MAN network).

50. The Panel congratulates the Netherlands on its establishment of a comprehensive network for long-term measurement of ammonia concentrations in nature areas. This has allowed a network of >235 sites with measurements in many different Natura 2000 sites. This high degree of coverage is unique in Europe.
51. *However, the methodology used for the MAN is not state-of-the-art.* It uses Gradko type passive diffusion tubes according to a design that was developed in the 1980s (35 mm diffusion path x 10 mm diameter), which has a rather slow sampling rate. By comparison FERM samplers (as used in Sweden, diffusion path 10 mm x 23 mm diameter) and ALPHA samplers (as used in the UK, diffusion path 6 mm x 23 mm diameter) sample 12 to 20 times faster and include methods designed to reduce contamination of samplers. *The result is that the Gradko tubes require a correction (c. 2 $\mu g\ m^{-3}$) which is similar to background air concentrations in the Netherlands.* This means that the samplers, as currently deployed in the Netherlands, cannot be used reliably to assess seasonal patterns, apart from for high concentration sites. (Note for example that this correction is even larger than the 1 $\mu g\ m^{-3}$ UNECE "critical level" for protection of lichens and lower plants from ammonia).
52. As an additional concern, the Panel notes that the Gradko tubes are only replicated at selected sites in the MAN network. *Most sites are operated without replication (i.e. single tubes).* This means that it is impossible to know whether a high value is real or a measurement artefact. While the Panel agrees with the Dutch scientists that a correction factor is needed, it is concerned that the use of this sampling method requires such a large correction factor to be applied in the first place. In situations where the correction factor is of the same magnitude as the corrected signal, the data need to be considered as too uncertain for trend assessment.
53. Questioning of the scientists by the Panel indicated that the choice to use this kind of sampler was based on: a) much lower analysis cost per sampler offered by the Gradko company than offered by contractors in the Netherlands or elsewhere, b) a perception of lower transport cost, c) experience that use of a rain shelter and Velcro attachment for alternative samplers (UK ALPHA samplers tested) led to higher visibility and rates of sampler vandalism at monitoring sites. In response to these points the Panel notes:
    a. While a lower analysis cost is attractive, this may not end up being a cost saving where the data fail to meet international performance standards. This applies both to sampler type and the choice not to replicate samplers.
    b. The actual sampler sizes of the Gradko, ALPHA and FERM samplers are similar. This means that postage cost differences are a function of how well samples are protected during

transport. The reason for protecting samplers is to improve data quality. Costs could be reduced by establishing a cost-effective chemical analysis laboratory in the Netherlands.

c. The use of a rain shelter means samplers are more visible. However, this principle applies equally to all three sampler types. Therefore if the benefit of a rain shelter is less than the benefit of reduced sampler visibility, a site set up should be used without rain shelters. This is a simple matter of logistic optimization (Velcro can also be replaced with clips if needed).

54. *In conclusion, the Panel is concerned that substantial resources are being spent on the MAN network, which could be used to obtain data of much higher quality.* The present data quality, while sufficient for monitoring near sources, is expected to be insufficient to analyse seasonal patterns of NH_3 concentrations or long-term trends at individual sites.

55. With substantial averaging over many sites, these uncertainties are likely to average out. Therefore, so long as the monitoring network sites have not been changed during the period 2005 to the present, the overall average of all the MAN sites is still a sound basis to assess long-term trend in the Netherlands. Two caveats should be added:
    a. It was not presented to the Panel how the MAN network has evolved over time. If the monitoring sites have changed, then this may affect the trend interpretation. It was not clear whether the comparison of measured MAN trend with model estimates was based on an average for the Netherlands, or receptor modelling for each site, then averaged up to the Netherlands for the same years for which monitoring was available at each site.
    b. The MAN network has been set up to sample in nature areas. This means that the monitoring sites are more than the average distance from ammonia sources. The overall level of ammonia concentration measured at the MAN sites can therefore be expected to be lower than for average locations in the Netherlands.

56. At present, the Netherlands does not have an extensive network of low-cost time-integrated ammonia monitoring for agricultural and other parts of the country. Future implementation of such monitoring would allow the temporal trends in different areas to be compared. For example, how do temporal trends compare in cattle dominated versus, pig or poultry dominated areas, as well as in agricultural and urban versus forest and other nature areas?

Other ammonia-related monitoring

57. *Reductions in NO_x and SO_2 emissions across Europe are expected to have reduced the rates with which gaseous ammonia reacts to form particulate ammonium in the atmosphere. For this reason it is essential that trend interpretation considers each of the different ammoniacal forms: gaseous NH_3, particulate NH_4^+ as well as concentrations in rain and wet deposition of NH_4^+.* With this in mind, the Panel was very surprised that so much attention had been given to interpreting gaseous NH_3 monitoring, while comparatively little attention given to the monitoring of NH_4^+ in particles and in precipitation.

58. The Panel noted that trends in *NH_4^+ in precipitation* are monitored in the Netherlands and were pleased to see the data reported. However, more attention could have been given to these data, including information on the number of sites, their representativity and the extent to which there are uncertainties in these measurements. This quality analysis is important as a foundation for comparison with models.

59. Concentrations of *ammonium in particulate matter* were not initially presented, and the Panel had specifically to ask to see these data. *In the 1990s, ammonium concentration in particles was*

found to decrease faster than the gaseous ammonia concentration and even faster than the estimated emission reduction (see chapter by A. Bleeker *et al.*, Atmospheric Ammonia, Springer 2009; based on earlier analyses from 2000 and 2003).

60. The more recent trends in ammonium aerosol in the Netherlands which were presented do not appear to show the earlier trends continuing. However, the sampling method also changed since then. *There appears to be a step change in the data linked to the change in method (around 2005-2010), which calls in to question the reliability of the trend assessment for ammonium particles.* Further efforts are needed to see if this dataset can be reconstructed more reliably, and to assess whether the network structure for ammonium aerosol measurements is sufficient.
61. The Panel notes that, with the increasing recognition of *human health impacts of particulate matter,* a higher focus on monitoring ammonium particles would be justified as part of overall efforts in monitoring composition and trends in particulate matter.
62. The current programme of Dutch monitoring does not yet use *satellite methods*. This point was raised in the stakeholder consultation part of the review. It is recommended that an independent assessment be made on the current status of satellite-based monitoring for ammonia, to see how much it could contribute to the Dutch monitoring programme.
63. One of the weak points of deposition monitoring internationally is the *measurement of dry deposition*. This is because of the difficulty of making these measurements. The Netherlands is one of the few countries which does include a few sites with such ammonia dry deposition monitoring, and should be congratulated for doing so. The treatment of dry deposition is also a key uncertainty in modelling (see below), and these data give the Dutch scientists a unique possibility to evaluate modelled dry deposition and potentially improve the applied dry deposition model.

Question A.3

64. **Is the scientific underpinning of the Dutch dispersion and deposition model sufficient?**

65. ***Summary: No. Sufficiency must be defined in relation to the purpose. At the heart of this review is the question of how to explain the difference in trend between ammonia emissions and atmospheric composition and deposition of ammonia and ammonium. International studies over the last 15 years have shown that one of the keys to explaining these differences is the relationship with SO_2 (and NO_x) emissions and their transformation. It is well documented that these interactions have an impact on the lifetime of ammonia in the atmosphere, allowing concentrations to be higher even when NH_3 emissions reduce.***

66. The main model currently used by the Dutch government for assessment of ammonia dispersion and deposition is the OPS model. This is a statistical model that includes an empirical description of chemical conversion rates. This means that there is no mass consistency between the transformation products of NH_3, SO_2 and NO_x emissions. Analysis of long-term trends using this model cannot therefore adequately consider the coupled chemistry interactions.
67. The Panel was nevertheless encouraged to see that the Dutch scientific community has now started to cooperate to improve the modelling capability by developing the combined model LEO (Lotos Euros - OPS). Further development of such alternatives needs to continue.

68. *The Panel was surprised that model tools like OPS and Lotos-Euros had not already been applied to test* <u>*specific hypotheses*</u> *to investigate the apparent discrepancy between trends in emissions and concentrations.* For example, Horvath et al. ("Atmospheric Ammonia", Springer, 2009) tested the hypothesis that the Hungarian "ammonia gap" could be explained by changing SO_2 emissions across Europe. They did this by comparing a model run using SO_2 emissions at 1990 levels with a run using actual changing SO_2 emissions over following decade, and found that this could explain the ammonia gap. With less SO_2 in the last decade (and still decreasing), the question now arises to what extent changing NO_x emissions and chemistry affect the Dutch ammonia gap. This could have already been tested comprehensively by the Dutch scientific community and presented to the Panel, especially considering the previous ammonia review.
69. At the request of the Panel, further information was presented on the outcomes of running the OPS model with different SO_2 emissions. Only initial results were available, but these suggested that such chemical differences may account for c. 1 µg m^{-3} difference when compared with the average concentration trend at the LML sites. Such analyses need to be extended to a) compare the effect on such trends using other models (i.e. with coupled atmospheric chemistry), b) assess the trend differences under different chemical scenarios for individual LML sites, c) consider the extent to which a longer lifetime of NH_3 is reflected in an increasing trend in NH_3 imported from neighbouring countries.
70. Initial assessment of the atmospheric chemistry interactions using the Lotos-Euros model (M. Schaap et al., 2015: *Transportafstand van reactief stikstof in Europa verandert over de periode 1990-2009*, Figure 2c) are welcome, and provide evidence to support the idea that reducing NO_x and SO_2 emissions has led to a longer atmospheric residence time of NH_3, and hence higher NH_3 concentrations.
71. By contrast to the question of temporal trends*, the OPS model can be considered as well-fitted for local scale ammonia dispersion studies.* This is important given the use of OPS for the new AERIUS model tool. Nevertheless, the Panel noted several areas in operation of the OPS model which were not very transparent. For example:
    a. the approach used to calibrate OPS to measurements was not very transparent,
    b. the temperature correction of national ammonia emissions was not clearly highlighted. For example, information on the temperature correction was buried deep in a technical report (OPS report, page 56).
    c. It was not clear how emission/deposition exchange is handled for grassland (e.g. whether there is double counting between net grazing emissions and dry deposition).
72. Taking account of such effects is important for model performance and needs to be made transparent so users (both the Dutch Government and other stakeholders) can understand the main principles of how the models work.

Question A.4

73. **Is the regional differentiation / local detail in the AERIUS model of ammonia emission, dispersion and deposition scientifically sound?**

74. ***Summary: Yes, as far as the Panel can see, based on the information presented. The model is based on the OPS model which uses the well-established principles of Gaussian dispersion.***

The model allows local information on factors that affect ammonia emissions to be included. However, insufficient information was provided on the technical basis of the AERIUS model, which should be subjected to ongoing review.

75. The Panel congratulates the Netherlands in its work to establish the AERIUS model as a user-friendly tool to assess local relationships between ammonia emission, concentrations and deposition. This tool can be expected to play a key role in supporting the future Dutch nitrogen policy, allowing the link between emissions and impacts to be assessed.

76. However, while technical information was provided to the Panel on the OPS model on which AERIUS is based, the Panel was left unclear about which simplifications had been made. The Panel agrees that such simplifications are necessary in developing such a user based system. However, the paperwork provided to the Panel was only a general description of purpose. Within the time allotted for the review, there was not space to delve further into the technical details of the AERIUS application itself.

77. The following comments can be made:

    a. The high resolution approach of using farm level emission is world leading in terms of the access to detailed farm datasets.
    b. The AERIUS model appears to allow specific information on building type at a farm level to be incorporated. This has the critical benefit that the model can take account of low emissions buildings where these are established.
    c. The AERIUS model does not appear to take on board specific farm information on manure spreading or storage practices at present. The philosophy appeared to be that all farmers should be complying with national regulations. However, the panel noted that there were differences in the way regulations applied (e.g. relaxation of the requirement to inject manure on peat soils, where trailing shoe applicators could be used instead).
    d. At present there does not appear to be any peer-review publication on the AERIUS model approach. Providing such a technical description for an international audience should be considered a high priority.
    e. Despite the incorporation of detailed emission information, an operational system like AERIUS will be associated with larger uncertainty than regional estimates because other local factors are not represented (e.g. local variations in meteorology). This uncertainty should be analysed in detail for transparency, including comparison with observations.

Question B

78. **Does the panel have an explanation for the divergent trends in calculated ammonia emission and measured ammonia concentrations?**

79. ***Summary: The panel is of the opinion that a substantial part of the differences can be explained by: a) less NH_3 emissions reduction 1990-2013 than estimated by the Dutch researchers (e.g. emissions overestimated in 1990; sub-optimal implementation of low emission techniques by some farmers, see question A.1), and b) non-linear responses in the***

atmosphere (question A.3), especially due to decreasing SO_2 and NO_x emissions, linked to an increasing lifetime of NH_3 in the atmosphere, with larger import expected from neighbouring countries.

Smaller decrease in ammonia emissions than modelled

80. The panel considers it most likely that ammonia emissions are currently overestimated for the early period, around 1990. This means that the estimated emission reduction to the present of 64-70% is probably an overestimate of the effectiveness of the Dutch ammonia policy.
81. Contributing factors to an overestimation of ammonia emission around 1990 are thought to include:
    a. The NEMA model may overestimate the protein content in grass grazed by cattle, sheep and goats in the 1990s and thus overestimate the amount of nitrogen in animal manure.
    b. The NEMA model does not take account that *more manure was spread in winter in the early period* than at present. Since emissions are temperature sensitive, this would mean that manure spreading emissions were not as large as currently estimated.
    c. The NEMA model appears to *overestimate the TAN in manure at the time of manure spreading* compared with measurements. This would tend to overestimate ammonia emissions in the early period when unabated surface spreading of slurry was allowed.
    d. It looks like the NEMA model may *overestimate ammonia emissions from some other livestock categories and source*s for 1990 compared with the present (e.g. sheep, rabbits, grazing) and underestimate current emissions for grazing.
    e. It was suggested that *recent years have been warmer, which may have increased emissions*. While this remains a likely explanation, insufficient data were presented to the review panel to demonstrate this effect.
    f. For recent years it has been demonstrated by a survey presented to the Panel that *not all farmers apply the low emission techniques to the full extent expected*. For example, independent assessment compared with farmers' views suggests that when injecting manure, some farmers only achieve an abatement effectiveness equivalent to trailing shoe. This occurs when the injection equipment is not used effectively.
    g. The NEMA model subtracts exported and burned animal manure in its estimate. An incorrect estimation of this may lead to an over/underestimation of the national emission.

Non-linear response of atmospheric concentrations to changing emissions

82. It is well established that atmospheric concentrations do not always change linearly with changing emissions. In the case of ammonia emissions, each of the atmospheric nitrogen components needs to be considered: gaseous NH_3, particulate NH_4^+, precipitation concentrations and wet deposition.
83. The contributing factors to the discrepancy that are related to atmospheric processes include:
    a. Decreasing SO_2 and NO_x emissions mean that the reaction of NH_3 to form particulate NH_4^+ proceeds more slowly. This means that NH_3 has a longer residence time in the atmosphere, keeping concentrations higher. It also means that NH_4^+ particulate matter should have decreased more than the NH_3 concentrations. (While preliminary analysis with the OPS model has shown effect on average NH_3 in the Netherlands, it should be examined more comprehensively using suitable models with coupled atmospheric chemistry, in relation to the NH_4^+ aerosol and wet deposition trends, as well as for individual sites.)

b. The consequence of a longer atmospheric lifetime of NH_3 is that *an increasing fraction of the benefit of Dutch ammonia policy is seen in countries down-wind of the Netherlands, rather than in the Netherlands itself*.
c. A further consequence of a longer atmospheric lifetime of NH_3 is that a larger relative contribution to Dutch NH_3 concentrations will be made by import from neighbouring countries. Such an increase in relative import is expected to have contributed to the lower-than-expected reduction in Dutch NH_3 concentrations. This emphasizes the need for an international approach to NH_3 emission control.
d. These points especially apply to the LML network sites, where several of the sites are close to the border of the Netherlands.
e. The LML network only has data from 8 sites (6 since 2014). This small number means that it is a challenge to use these numbers as representative for the Netherlands. By contrast, the hourly temporal sampling provides critical information which has so far not been analysed sufficiently to reveal patterns in the data.
f. The local 'stories' at each of the LML sites have not been fully developed. For example, on presentation to the Panel, it was mentioned by the Dutch scientists that an increasing trend of NH_3 concentrations for one LML site might be connected to this being an area receiving increasing amounts of manure imported from elsewhere in the country. Such arguments need to be demonstrated by quantitative local analysis.
g. It was suggested by the Dutch scientists that one reason for the lack of trend in NH_3 concentrations was because turbulent differences mean that the effect of building sources is seen more strongly on NH_3 concentrations than the effect of manure spreading sources. Although evidence was not presented to demonstrate this effect, it may be noted that:
    i. Such a differential-weighting is expected because most ammonia emission from manure spreading occurs during the day (due to time of spreading, higher turbulence and warmer surfaces), while emissions from animal houses (especially those with forced ventilation) tend to continue at night when atmospheric mixing is less. The result is that emissions from buildings (especially with forced ventilation) will tend to contribute relatively more to average NH_3 concentrations in source areas.
    ii. This would affect the trend because estimated NH_3 emissions from buildings have not decreased so much since 1993 as from manure spreading.
    iii. The Panel noted that this remained only an untested hypothesis, and therefore requested the Dutch scientists to separate monitoring data from day versus night. This comparison showed that (since 1993) the average ammonia concentration from the 8 LML sites decreased more strongly during the night than during the day.
    iv. This outcome was unexpected and therefore did not support the hypothesis. However, it suggests that there are other interactions which need to be further investigated. Sensitivity studies with models should be used to support the analysis.
h. Most of the estimated reduction in ammonia emissions occurred in the period between 1993 and 2000. Between 2005 and 2013 a much smaller amount of further reduction in NH_3 emissions has been achieved (around 15% further reduction). The further reduction over the last decade is therefore a rather small signal for the MAN network to be able to detect (noting that the MAN network was only established in 2005). Considering the small size of this effect, the following should be investigated:

i. Statistical analysis should be made for the MAN network in combination with modelling to examine what is the smallest amount of change in NH_3 concentration which would be statistically detectable over time.
ii. The large number of monitoring sites in the MAN network should be exploited to see if there are differences in the temporal trend, for example between sites dominated by different source types (cattle, pig, poultry etc.) and between different locations, e.g. near the national border.

General Messages

84. The Panel was encouraged to see that the Dutch government had responded positively to the last international review of 2013. For example, the increased attention to assess meteorological dependence on ammonia emissions and to process modelling was welcomed by the Panel. However*, the last review was only 2 years ago, so the outcomes from that work are therefore not yet complete.* It is anticipated that the outcomes of the present review may take a further 3 years to be implemented, assuming that appropriate resources are made available.
85. The Panel noted that *some stakeholders found it confusing as to why the emissions inventory estimates and trends changed with time*. The Panel recognized that continuing improvements in methodology are natural and to be expected, and noted that explanations of these differences were available. This includes the Informative Inventory Review (IIR), which is required as part of the national inventory procedures, and the Pollutant Transfer and Release Register (PRTR, prtr.nl). However, the Panel considered that further effort needs to be placed to translate technical reporting into non-technical summaries that can be more accessible and useful, especially for non-specialist stakeholders. For example, the IIR focuses mainly on changes since each previous year, with details in principle available via references cited. What stakeholders were missing was an overview across the whole period (last 10 and last 25 years) to explain the rationale and the key changes that have occurred in the calculation methodology and how this relates to the observed trends in NH_3 and NH_4^+. *The Panel encourages the Dutch scientists to publish non-technical summaries for wider readership, which would also provide pointers to where the detailed information could be found.*
86. The Panel commends the Dutch scientific community for achieving a high number of peer review papers on ammonia in the international literature. However, this requirement should be maintained with resources made available to allow continued publication (both for time and publication charges for open access publication).
87. The Panel noted concerns expressed by Dutch scientists where they had been requested to provide stakeholders with very detailed original datasets going back many years. The Dutch scientists noted that this could take a very long time to prepare data into a form useful for stakeholders, especially if this should be combined with all the associated metadata and information required on the methodology used to make the calculations. This especially applied to old experimental datasets, where data were stored in old formats or may no longer be easily available. *The guidance of the Panel is that data should be freely available to stakeholders. However, this does not mean that data are always available without charge. It is the responsibility of those requesting data to pay for the additional work incurred.*
88. As with other countries, research funding has been less stable over the last decade (2005-present) than it was in the previous twenty years (1985-2005). One consequence of this decrease

has been *a stagnation of ammonia research in the Netherlands, where many of the estimates rely on now rather old publications*. This point was already identified in the 2013 international ammonia review. *In addition, the present panel here identifies another feature associated with the decreased and intermittent funding: a reduction of the intellectual capital in ammonia research.* This is indicated to the Panel by: a) much less synthesis of the available datasets than the Panel would have expected, based on the Netherlands' previous leading expertise in this area, and b) involvement of only a few research teams in the Netherlands ammonia programme compared with a wider research base previously.

89. The recent investment in response to the 2013 review is beginning to address these concerns. However, *sustained long-term investment will be needed if the Netherlands are to capitalize on its past leadership of ammonia research.* This is a point that does not only concern research. The Netherlands is still a world leader in ammonia mitigation practices. This is a key opportunity for the Netherlands as other countries develop plans to reduce NH_3 emissions. *Continued investment in novel farming practices, measurement techniques and underpinning ammonia science will all be necessary if the Netherlands are to benefit economically as an international innovator in this area.*
90. As part of this future landscape, *human management of the nitrogen cycle is rising increasingly up the agenda.* The wider importance of nitrogen has been clearly demonstrated by 'The European Nitrogen Assessment' (Cambridge University Press, 2011) and the 'Our Nutrient World' report (2013) prepared for the United Nations Environment Program (UNEP). As UNEP now starts to develop the International Nitrogen Management System (INMS), as a science support mechanism, and to consider the policy options, it is clear that the Netherlands have a lot to offer.
91. 'Our Nutrient World' estimated that improving nitrogen use efficiency by 20% would have net benefits world 170 billion USD per year globally. *Reducing ammonia emissions translates directly into improved nutrient use efficiency. The ammonia mitigation story therefore contributes significantly to developing the Nitrogen Circular Economy. It is an emerging business opportunity where the Netherlands could be leading the charge.*

Appendix 1:
Supplementary Technical Information

Table: Implied emissions factors calculated by the NEMA team at the request of the review Panel.

Emission factors kg NH_3/head/year

	1990	1995	2000	2005	2008	2009	2010	2011	2012	2013
Dairy cow	64.3	33.4	25.9	27.8	26.4	25.4	26.6	26.7	24.8	24.9
Young dairy cattle	24.3	15.2	12.8	11.2	10.5	10	10.6	10.3	10	9.9
Fattening calves	7.2	4.1	4.6	4.2	3.9	4.2	4.3	4.6	4.8	4.5
Young cattle for meat production	24.5	14.2	11.9	11.2	11.1	11	11.2	11.2	10.4	10
Suckler cow	30.5	21	14.9	13.3	12.2	11.6	11.8	12	10.9	10.9
Sheep	3.9	3.8	2.6	1.5	1.4	0.9	0.9	0.8	0.8	0.8
Goats	8.2	8.9	7.5	6.1	5.2	5.1	5.5	5.7	5.3	5.5
Horses and ponies	13.1	13.2	11.9	11.5	10	9.8	10.1	10.3	10.1	10
Fattening pigs	10.2	6.7	5.3	4.8	4.5	4.4	3.9	3.5	3.4	2.8
Breeding pigs	19	10.9	9.5	8.4	7	7.4	7.3	6.2	6	6
Laying hens	0.5	0.36	0.28	0.19	0.19	0.18	0.16	0.18	0.17	0.16
Broilers including ducks and turkeys	0.29	0.18	0.21	0.23	0.17	0.16	0.11	0.11	0.08	0.08
Rabbits and fur bearing animals	2.8	1.3	0.93	0.87	0.51	0.4	0.38	0.42	0.48	0.5

Main drivers for the level and the trend in the national ammonia inventory (NEMA)

1. From 1990 to 2013 has the estimated ammonia emission, based on the latest inventory submission reported to European Monitoring and Evaluation Programme (EMEP) (www.ceip.at), been reduced from 373 Gg NH_3 to 134 Gg NH_3 . This is equivalent to a reduction of 64 %. (This is less than the figure of 70% mentioned in the written report of Groenestein provided to this review). The agricultural sector is the main emitter contributing 94 % to the estimated total emission in 1990 and 86 % in 2013. The reported ammonia emission from the agricultural sector has reduced by 67 % between 1990 and 2013.
2. The emission estimates are annually submitted to EMEP and the EU and published among other countries on the homepage of CEIP in the NFR-format tables (Nomenclature for Reporting) with further written information in the IIR (Informative Inventory Report) www.ceip.at. On this homepage all previous submissions can be found. The IIR is however difficult to understand for the non-scientific community.
3. Due to new knowledge, most national inventories are recalculated every year. The most notable recalculation for the Dutch inventory was the introduction of the National Emission Model for Ammonia (NEMA) in 2011. Its first use was for the inventory year 2009 (reporting year 2011). Implementation of the NEMA model resulted in an increase in the overall Dutch emission for 1990 from 253 Gg NH_3 to 355 Gg NH_3, which is equivalent to an increase of 40 %. Implementation of the NEMA model only slightly increased the reported national total for 2007 from 137 Gg NH_3 to 140 Gg NH_3 (an increase of 2 %).
4. Introduction of the NEMA model thus affected the estimated national trend in NH_3 emissions for the Netherlands between 1990 and 2007, implying a larger reduction than had previously been estimated.

5. From 2007 to 2013 the NEMA model (based on the latest submission) shows a further reduction from 160 to 134 Gg NH_3 /year (a reduction of 16%).
6. The main change in the methodology that has been introduced with the NEMA is a shift from a total nitrogen flow concept (N) to a Total Ammonical Nitrogen flow concept (TAN). TAN is the fraction of N in the feed or manure which is not bound in organic matter and can therefore easily volatilize. The share of TAN in manure of total N is generally 50-60 % for cattle slurry and 75-80 % in pig slurry. Use of the TAN-flow concept is in accordance with the recommendations of EMEP.

The NEMA model

7. By far the largest source of NH_3 emission from agriculture is due to handling of animal manure. Emission occur during housing, during manure storage and following manure application. Ammonia is a breakdown product of protein in the feed. Excretion as urea or other nitrogen compounds (e.g. uric acid for poultry), is followed by microbial breakdown which liberates ammonia. Excessive protein in feed leads to much higher N excretion by animals.
8. In the Netherlands in the beginning of the 1990s there was a high focus on producing enough feed in terms of energy and less focus on the protein level in the feed. High levels of feed energy were produced by using high application rates of mineral fertilizer to crops and grass, resulting in excessive protein contents in the feed, especially in grazed grass. On the other hand, there was less attention on the nitrogen in animal manure, how the manure was handled and the consequent losses of leached nitrogen and NH_3 to the air. In order to minimize storage needs, manure was applied the year around, including outside the growing season, when less effective use of it could be made by crops.
9. The approach of the NEMA model allows many of these factors to be included, including how they may have changed with time:
    a. The input of nitrogen in the NEMA model is based on the total feed production of roughage and sales data on concentrates, poultry and pig feed in the Netherlands.
    b. The nitrogen excretion of NEMA is based on animal models which take into account the digestibility of the different feed compounds.
    c. The ammonia emission from livestock in NEMA is based on the TAN content in the excreted nitrogen in the manure.
10. The Panel was provided with references to different documents describing the methodologies used in the NEMA model and wider NH_3 inventory, but not the currently used driving factors in the inventory. This made it difficult to understand the emission changes in the inventory through time.
11. In general, the Panel considered that the model set-up is scientifically sound and well documented in parts. However, there is a need for a more easily accessible description on the annual emission estimates, and how these have been estimated, together with a description of the major driving forces that lead to estimated changes in emissions over time.

Ammonia emission level and trend 1990 to 2013 with the NEMA model

Main Drivers for dairy and other cattle

12. The main drivers causing changes in estimated NH_3 emissions appear to be:

- Changes in feed N intake, primarily due to changes in the protein content in meadow grass,
- Changes in the manure application methods from surface spreading of the manure to injection,
- Changes in ammonia emission factor for grazed grass due to a lower protein content in grazed grass.

Changes in cattle feeding

13. One of the major changes in cattle feeding that has causing a reduced ammonia emission is the estimated reduction in the nitrogen content of grazed meadow grass. In the inventory for 1990 it is assumed that the grazed grass contained a nitrogen content of 42.9 gram N/kg dm (26.5 % crude protein in dry matter). In 2012 the protein content has been reduced to 18.3 % crude protein (reduction of 32 %). This reduction was explained as due to the reduced nitrogen fertilization level of Dutch pastures.
14. For silage grass and hay, the crude protein content was reported to have decreased from 19 % to 16.6 % (a reduction of 13%). The protein content in grazed grass used in the inventory is based on laboratory data for “fresh grass” as these are the only available data.
15. It is well known that the protein content in grazed grass differs over the grazing season and depends among other things on the fertilization level and the way the grazing is managed. The protein content of the grazed grass used in the inventory in the 1990s is high, but not unrealistic compared with data and expert judgments from the Danish feeding scientists who were consulted.
16. In 1990 grazed grass is estimated to have accounted for >40% of the annual nitrogen intake by cattle, which is estimated to have decreased to 5% of the nitrogen intake in the South-Eastern part of the Netherland and to 19% in the North-Western part (estimates made by the Panel based on information presented). The decreased share is also due to a lower number of grazing days and more housing of the dairy cows. If there is an overestimation of the N content in grazed grass in 1990s, then this would lead to significantly lower ammonia emission per dairy cow in the 1990s.

Manure application

17. Based on information presented to the Panel, the comparison can be made between:
    - 1990, when broad spreading of manure was used all year round, with a default emission factor of 74% of TAN in the manure,
    - 2013, when 70 % of manure is estimated to have been deep injected on arable land with a default emission factor of 2 % of TAN in the manure.
18. The most important driver in the reduction of ammonia emission from manure spreading between 1990 and 2013 is the changes in manure application techniques. In 1990 to 1992 it was assumed that all manure was applied as surface spreading, not incorporated within a few days and that 74 % of the TAN for liquid manure was evaporated as NH_3. This emission factor (EF) is based on Dutch studies but used widely in Europe.
19. The ban on broad spreading from 1992 leads to very large decrease in the estimated NH_3 emission from 1990 to 1994 and further on up to now because the emission estimate in the inventory assumes a change from an almost worst-case scenario to the best-case scenario. However:

- The assumption for 1990 to 1993 may have overestimated emissions as some manure could have been incorporated in the soil, while new data presented to the Panel shows a lower emission factor when the manure is applied in the cold and wet season in winter.
- The assumption for 2013 may have underestimated emissions as evidence from a farm survey presented to the Panel indicates that not all farmers use low emission techniques correctly, leading to a lower than anticipated effectiveness in the emission reduction.

Housing, air cleaning

20. During the last years the demand for installation of air scrubbers in pig houses has increased. According to information presented to the Panel, in 2013 54.7 % of the housing systems for sows were equipped with air scrubbers and 42.8 % of the houses for fattening pigs. Air scrubbers clean the ventilation outlet preferential with sulfuric acid, converting NH_3 to NH_4^+ solution which can then be reused on the farm by adding to farm slurry storage tanks. It was reported that the general efficacy of air scrubbers is around 90 %.
21. The Panel was also informed that there is little demand for air cleaning technologies for poultry houses because these houses have high emissions of dust which easily block the air cleaners. This indicates an opportunity for further technical development.
22. During the meeting the Panel was informed that some of the installed air cleaning technologies do not function properly. In 2009 it was assumed that this represented 40% of the installed air scrubbers, with this reducing to 8% in 2013 as a result of installing an online monitoring system. The NEMA team informed the Panel that the inventory takes account of these estimates.

Distinguishing nitrogen excretion rates for dairy cows between two regions

23. It was reported to the Panel how the NEMA model is set up to distinguish nitrogen excretion rates between two regions of the Netherlands, South-East and North-West, in order to take account of differences in feeding. Maize and grass silage feeding in the South-East and more grazing in the North-West.
24. When NEMA, which is a national model, estimates the NH_3 emission it takes into account all consumed feed regardless of where it is consumed. The use of sub-regions therefore does not affect the mass consistency of the national estimates.
25. The use of two regions in the model allows it to distinguish N excretion (N_{ex}) rates for cattle between contrasting systems. In 2012 the reported N_{ex} in the South-East was 112.8 kg N / dairy cow / yr and in the North-West 135.9 kg N / dairy cow / yr for an average milk production of 8006 liter/ dairy cow /yr (Dierlijke mest en mineralen 2012). The opinion of the Panel is that a N_{ex} of 112.8 kg N / yr is low compared to the nutritional need. The Panel expects that a more appropriate value could be 125-130 kg N / dairy cow / yr.
26. If the distinction of the feed consumed by dairy cows between the two regions is incorrect this may lead to misjudgment of the N load in different regions of the Netherlands. It is recommended that further checks be made to justify the allocation of the consumed feed.
27. Although the two regions are distinguished in the NEMA model, the Panel was informed that these regionally different N_{ex} values are not used to provide spatial input to the NH_3 emissions inventory used in the OPS model (only affecting the national total). The consistency of methodology between models therefore needs to be reviewed.

Appendix 2:

Minutes of the Review Meeting

Panel members: Mark Sutton (UK, chair), Tom Misselbrook (UK), Steen Gyldenkaerne (DK), Ulrike Dragosits (UK), Camilla Geels (DK), Wim Bussink (NL, secretary)

The terms of reference are given in Appendix 3 and the review programme in Appendix 4. These minutes do not include views of the panel (either in agreement or disagreement). The minutes simply summarize key messages presented by those giving evidence to the Panel.

Tuesday June 23

9.00-9.30: Introduction by Peter Munters (Ministry of EZ)

The Dutch Ammonia emission and legislation program was presented briefly. From July 1 onwards a new integrated approach (PAS) becomes effective. It is based on 3 Pillars (reduction in N deposition, ecological restoration measurements, scope of development). In PAS Aerius is used for local deposition modelling in order to quantify the scope for development. For the integrated approach it is essential to know that measures work and reduce deposition. Therefore the scientific basis of the entire knowledge chain must be credible.

9.30-14.30 (Emission modelling and emission factors)

The NEMA model (national emission model agriculture): (Gerard Velthof, Alterra, chair of NEMA)
The general setup of the NEMA model was described. The model calculates the national annual emission from agriculture. In the model all emission factors for housing, slurry spreading and grazing are expressed as a fraction of TAN (total ammoniacal nitrogen). A working group updates the model regularly and reports the results to the Ministries of Economic Affairs and of Environment & Infrastructure

Nitrogen excretion calculations (Cor van Bruggen, CBS)
Calculations of N excretion by animals has been made since the early 1990s. This calculation is based on “intake (amount of feed an N content) minus fixation (retention)”. Calculation takes place for all kind of animal categories based on animal numbers of April 1 of a specific year. Technical details are explained for cattle, pigs and poultry. The overview over the last 20 years shows a decrease in TAN excretion, especially for dairy cattle.

Emission from storage and Housing (Karin Groenestein, Livestock research)
It was shown how the emission calculation from storage and housing takes place. Census data (yearly) are used to identify housing types, livestock numbers and what part of the manure is stored (outside the stable). N-mineralization of manure while stored is taken into account. She showed that there are several hundred emission factors to take the difference in housing and floor types into account. The measurement protocol to determine an emission factor was explained. Examples of a changed management and an updated in emission factors were illustrated.

Application techniques (Jan Huijsmans, Plant Research International)
The application techniques that are allowed nowadays on grassland and arable land were shown. An overview of the emission results from the last two decades showed that there is a tendency of increased emissions with low emission techniques since the early nineties (i.e. the methods did not have such low emission factors as originally expected). This has led to adjustment of the emission factors. Comparison of Dutch data with UK data and modelling data showed similar results. Ongoing research focusses on better modelling to take weather data better into account.

Other sources like grazing, crops and fertilizers (Gerard Velthof, Alterra)
The emission results from grazing were presented and how these results were recalculated to take the strong decrease in N excretion into account. Also the background of deriving emission factors for fertilizers as well as for crop residues and ripening crops was explained.

Trends in agricultural emission (Gerard Velthof, Alterra)
An overview of the emission from 1990 onwards shows a sharp decrease the first 10 years. The last 10 years there is little decrease. He reported a basic sensitivity analysis. Currently the uncertainty is estimated at 20%. He noted that in future a Monte Carlo analysis will be done.

After each presentation there was a discussion. The review panel had a need for clarifications on several issues as animal excretion data two decades ago, grazing, the animal diet, the manure application pattern during the year and decades, the emission factors used etc. Some of these questions could be addressed during a later part of the meeting. This was to a large extent presented by Velthof and Huijsmans on Thursday morning.

Tuesday 15.00 -19.00 hrs and Wednesday: 14.00-16.00 hrs
Dispersion, deposition, monitoring; measurement and calculations

Measurement of ammonia in the Netherlands; the monitoring network (Daan Swart, RIVM)
An overview of the monitoring situation and locations for ammonia was presented. The measurement techniques used like AMOR, DOAS, and passive diffusion tubes were explained. Also the quality programme to maintain the equipment and to ensure proper measurement quality was explained.

The OPS model (Fred Sauter, RIVM)
The functionality of the OPS (long term) model is briefly explained. It is used to calculate ammonia deposition for the last two decades. It takes into account different wind directions and meteo conditions (stable, unstable, neutral). The model is regularly updated in order to improve the matching between measured and modelled. It seems that there is a slight underestimation. The new model LEO may solve this.

Uncertainty between emissions and depositions, (Margreet van Zanten, RIVM)
An overview is given about the uncertainty analysis in the wet and dry deposition measurements. It appeared that there is a mismatch between MAN measurement and deposition measured with OPS especially in coastal areas. This could be corrected by taking an emission factor from sea in to account. The Depac model to emissions from crops into account was briefly explained

Emission and dispersion (Wim van der Maas,RIVM)

The Pollutant Release and Transfer Register was demonstrated.
There is a new update of the national ammonia emission calculations, resulting in a 14% higher emission, because of increased emission factors for pig housing, manure application and traffic and the inclusion of the emission of ripening crops. Now NL is above the NEC ceiling of 128 Gg. On the website (http://www.emissieregistratie.nl/ERPUBLIEK/content/explanation.en.aspx) it is mentioned what has changed compared to the predecessor report.

Aerius, (Mark Wilmot)
An explanation of Aerius and the underlying functionalities was given. Not only agricultural emission sources are taken into account but also other emission sources. Aerius is not restricted to only ammonia, but takes also other N sources into account. Aerius is used to get a permit to expand a farm, factory. It is user friendly. The farmer can fill in his own data and see what this means for his situation.

After each presentation there was a discussion. The review panel had a need for clarifications on several issues, especially about the aerial chemistry. Several questions were asked by the Panel which were addressed by researchers from RIVM on Thursday morning

Wednesday morning 9.00-13.00 hrs

Discussions with other Dutch Scientist who are involved in ammonia related issues.

The review Panel was joined by: Arjan Hensen (ECN), Egbert Lantinga (WUR), Jan Willem Erisman (LBI, Prof integrated N studies), Geesje Rotgers (V-focus), Wim de Vries (Alterra, Prof N impact modelling), Jan Duyzer (TNO), Hans van Grinsven (PBL, evaluation of policies).

1) Arjan Henssen (ECN)
The measurement of ammonia after spreading of manure (grazing, fertilizer application) was presented, with special reference to the micrometeorological integrated horizontal flux method, which had received some criticisms:

- The turbulence effect is mostly neglected this can give an overestimation of 0-20% (it was neglected in the past because there were no fast sensors to measure it). Recommendation: more modelling and new measurements to overcome this.
- Displacement height. If this is not taken into account this may results at max in a few % overestimation.

2) Egbert Lantinga (WUR)
Results of recent experiments were shown were only concentrations measurements (no flux measurements) have taken place. According to this presenter the emission (only based on concentrations) is overestimated by at least a factor 4, partly due to an oasis effect where self-sheltering by build-up of high ammonia concentrations reduces emissions in large fields.

3) Geesje Rotgers (journalist V-focus)
She presented emission data from different years and compiled them in a single graph. The basic questions were: Why is there “every year” a recalculation including the preceeding years? Which of

the lines drawn in the graph is correct? Rotgers requested more transparency in the calculation and presentation of ammonia emissions estimates.

4) Jan Willem Erisman (LBI)

According to Erisman the conclusions of the review 2 yrs ago are still valid. The emissions are more or less correct. However he recommends a more process-based approach, and not simply using TAN. He suggests to look more critically to the emission reduction techniques, in understanding how they work and look more dynamically. Aerius is a great model but he asked is it useful for the regional or local situation?

5) Wim de Vries (WUR)

The problem is the enormous number of emission factors for different housing types etc.. How well do we now this? The ammonia emission from fertilizers is not well underpinned. It is based on the data of the literature study of Bouwman. There are almost no measurements regarding fertilizers. Local to regional scale: How to cope with these scales. An example: For the NFW project the average difference between OPS and MAM model is about 5%. On the local scale the difference can be much larger. A positive from recent studies is that the difference between emission and deposition is decreasing (the ammonia gap). There are 5 reasons mentioned for that, but what is missing is whether the techniques are applied properly by farmers, like air scrubbers, which are often not used properly.

6) Jan Duyzer (TNO)

There is development in the comments (how they are handled) about Aerius. Comments about the Ops model:

- Very little comparison with other models.
- The scale is important. It was developed for national scale assessments and does its job well (<30% uncertainty). Local scale uncertainty is however larger, about 70%. There are some weak points regarding scenario analyses. OPS is a linear model. You can only interpolate and not extrapolate the results (process based would be better). The aerial chemistry of sulphate, ammonia, other N components needs improvement.
- Ammonia compensation point. Dangerous to use it in extrapolation.

7) Hans van Grinsven (PBL)

PBL is using the models in ammonia related policy analysis and policy evaluation for effectivity and efficiency. Trends in deposition and emission are often more important than absolute numbers.
To the opinion of van Grinsven the ammonia trend discrepancy (strong decrease in emission, slow decrease in deposition) is caused by the first 10 yrs due to lack of accurate monitoring data.
The last 10 years there is not much change in emission and deposition. Major nature improvement will come from restoration and management.

After the morning session people provided the Panel with additional information sent by email.

Wednesday afternoon (16.00-19.00 hrs) and Thursday morning (8.00-9.00 hrs)

The review panel started with a first draft of their review and pin-pointed the main issues to be discussed on Thursday morning.

Thursday (9.00-13.00 hrs)

<u>Velthof (WUR)</u>
Manure composition data of the last 20 data for cattle were shown. There is however still a 10% gap between TAN calculated before spreading and the TAN measured in manure samples. The TAN flow model is used in the whole chain and not only for the slurry. There could be a slight overestimation in emission.

For Pigs the gap between TAN calculated and measured is smaller than for cattle, somewhere about 5-6%.

The quality of the data is better nowadays than in the past. The animal excretion calculations are done using the composition of the feed and concentrates. The background of the grazing emission factor was explained.

The differences observed by Rotgers in emission could be explained and are also published and described on the internet, but are not presented in such a way to be easily understandable to a non-scientific audience.

<u>Huijsmans (WUR)</u>
Huijsmans gave additional information about the spreading pattern and when it was allowed to apply slurry during the last decades. Information was given for grassland and arable land (per soil type). Information was presented about the meteorological conditions.

There is some discrepancy between what the farmers say with which technique they apply slurry and what they are doing (based on survey data). The latter is <u>not taken into account</u> in the NEMA model. Currently (2015) a new census is done graphically, using pictures (the technique is not asked). Farmers complain about the amount of data they have to provide.

Conclusion: Due to misunderstanding of the farmers there may be overestimation in the estimation of ammonia mitigation, while farmers apply more manure than they are allowed to apply (for the whole it is about 5% more according to statistics). There is also some fraud. The emission of 1990 may be overestimated.

<u>Roy Wichink Kruit (RIVM)</u>
RIVM provided additional information about the sensitivity study OPS model. It became clear that the aerial chemistry needed improvement to get more accurate deposition values. Therefore the goal is the combination of Lotus Euros and OPS. This is combination is termed LEO. First results seems promising.

A graph of the 8 LML stations was shown. Between sites there was a lot of difference. The panel noted that many of the deposition measurement sites were along the border of the Netherlands. The panel noted that it is important to have several and representative deposition measurement sites.

A detailed discussion took place.

<u>Margreet van Zanten (RIVM)</u>
The deposition data as measured on the 8 LML's sites was presented for the individual sites. Every site has it its own story and is helpful in understanding what is happening. A detailed discussion took place about the effect of deposition modelling versus the location of the deposition measurement sites. Some omissions and possible improvements were observed by the panel

Thursday (14.00-15.00 hrs)

A first summary of the findings of review panel was presented to Mr. Roel Feringa (Ministry of EZ, director nature and biodiversity).

Most of the questions were answered positively by the Review Panel. The positive issues were presented as well as the issues which could be improved. First recommendations were given.
The session was interactive.

Roel Feringa thanked the commissioned for their thorough work in a short time.

It was agreed to deliver the final review in a few weeks.

The meeting closed.

Appendix 3: Terms of Reference for the Review on scientific underpinning of Dutch policy for ammonia deposition

The Dutch Minister of Agriculture has promised to review the scientific basis for the calculation of ammonia deposition during a discussion on ammonia deposition in Dutch Parliament. The reason of the discussion in Parliament was a report of the RIVM, in which the RIVM presented that the trend in the calculated ammonia emission and the measured ammonia concentration diverge. The calculated ammonia emissions has a slightly downward trend, while the measured ammonia concentrations remain stable.

Questions
The review concerns the entire knowledge chain of data collection and emission calculations to deposition calculations. The review should answer the following questions:

- Deposition is calculated on a regional level scientific good approximation of the actual deposition occurred?
  with the sub questions:
  - Is the calculation of the emission in the Netherlands (national and regional) scientifically sound?
  - Are the set-up and measurement techniques of the Dutch ammonia monitoring network scientifically sound for the determination of ammonia concentrations on a regional and national scale?
  - Is the scientific underpinning of the Dutch dispersion and deposition model sufficient?
  - Is the regional differentiation in the Aerius model made scientifically sound?

In addition, the committee will be asked whether they have an explanation for the divergent trends in calculated and measured ammonia concentration.

Type of review
The ministry decided to perform the review in the form of a review committee. The review committee will visit the Netherlands in three days to form an opinion on the submitted questions.

Prior to this visit, they receive limited information containing the questions and brief descriptions (2-pagers) from the Dutch emission calculations, measuring network and distribution models. No reports are sent with it, but internet links can be included in the brief descriptions.

Program of the three days in the Netherlands:

- First day and a half:
  - Explanation Ministry
  - Divergent trends (Emission)
  - (DLO) Calculation of emissions
  - (RIVM) Monitoring networks
  - (RIVM) Dispersion and deposition model
  - Aerius (RIVM?) Regional dispersion and deposition model
  - Personal review other Dutch scientific experts on ammonia
- Half day deliberation committee
- Morning third day any second interviews to work out differences between interviews;
- Afternoon third day feedback findings to Ministry

- The committee provides three weeks of the meeting in the Netherlands in the final report

Appendix 4: Review programme

Date: Tuesday 23 to Thursday 25 June 2015
Location: Grand Hotel Karel V, Geertebolwerk 1, 3511 XA Utrecht

Tuesday 23 June

08:30- 09:00	Instruction and appointments
09:00- 09:30	Introduction by ministry
09:30- 10:30	Emission (modelling part)

Coffee

10:45- 12:15	Emission (emission factors)

Lunch

13:30 – 14:30	Monitoring network
14:30 - 15:30	Dispersion and deposition

Pause

15:45 – 16:45	Aerius
16:45 – 17:45	divergent trends in calculated ammonia emission and measured ammonia concentration.

19:00 Dinner

Wednesday 24 June

09:00- 12:30	Discussion with Dutch scientists

Lunch

13:30 – 17:00	Deliberation by the committee

Thursday 25 June

09:00 – 12:00	A second discussion with scientists to unravel possible differences between views presented during the first day;

Lunch

13:00 – 15:00	Feedback of the main findings to Ministry