Authors' response for ACP-2011-729 "MAX-DOAS tropospheric nitrogen dioxide column measurements compared with the Lotos-Euros air quality model" by T. Vlemmix et al.

The authors greatly acknowledge two anonymous referees for carefully reading the manuscript, for providing positive feedback and providing many useful suggestions for improvements. Also, as a first author, I sincerely apologize to the referees and the editor for the great delay before re-submitting this work.

In this document we will answer to the questions asked by the referees, and indicate how this lead to changes in the revised version of the manuscript. The original text by the reviewers is copied to this document and printed in italics. The authors' response (AR) is written as normal text, and indicated by 'AR'. Pages in the revised manuscript are referred to as 'rev.man.' (for instance, rev.man. p.12).

1.0 Comments by Anonymous Referee #1

Received and published: 20 December 2011

1.1 General comments

The manuscript entitled 'MAX-DOAS tropospheric nitrogen dioxide column measurements compared with the Lotos-Euros air quality model' by Vlemmix et al. describes the comparison of MAX-DOAS measurements of the NO2 vertical column density with results from an air quality model. NO2 is one of the key compounds in urban pollution and its continuous monitoring is of great importance. In contrast to in-situ observations, the remote sensing of NO2 has the advantage of being less sensitive to local variations in the trace gas concentrations, since the average over a larger volume is observed. Therefore MAX-DOAS observations are very suitable for the comparison with model calculations. In this paper, a relatively simple method (compared to full vertical profile retrievals) for deriving the NO2 VCD, including a correction for situations where clouds have an impact on the measurements, is presented and validated against model simulations. Diurnal, weekly and seasonal variation in NO2 are discussed in detail. The scientific questions addressed in this paper fit well within the scope of ACP and I recommend publication after some minor modifications described below.

The paper is well written and the methods are described in a clear and concise way. It is well structured, except that the discussion of the radiative transfer and the definition of the airmass factors (Section 2.2.2) should be presented prior to the discussion of the impact of clouds (Section 2.2.1).

AR 1: The order of sections 2.2.1 and 2.2.2 has been reversed.

Also, a short description of the instrument and the measurement uncertainties is missing.

AR 2: A description of the instrument has been added to sect. 2.1. Uncertainty estimates are provided at two places:

(1) A new figure has been added (Fig. 2 of rev.man.) which is discussed in Sect. 2.2 and which illustrates the detection limit for the measurements of tropospheric NO_2 differential slant columns with the instrument and settings used. Also the typical RMS of fit residual is mentioned in this section.
(2) A table has been added (Tab. 2 of rev.man.) in order to show relative uncertainties in the tropospheric NO_2 column related to uncertainties in the air mass factor caused by: aerosols (AOT and aerosol profile height), cloud bottom height, NO_2 profile height.

The abstract is far too long and requires substantial shortening. It contains many un-necessary technical details which can be omitted and should rather be discussed in the main body of the paper.

AR 3: The abstract has been shortened as requested.

Sections 2.2.1 and 2.2.2 should be swapped: The discussion on the impact of clouds in Section 2.2.1 is not understandable without knowledge on the definition of the box-AMF provided in Section 2.2.2. The definitions and nomenclatures defined in 2.2.2 should be used in the course of the discussion of the sensitivity on clouds in 2.2.1.

See **AR 1**

It is stated several times in the manuscript that aerosols have a relatively small effect on measurements performed at 30◦ elevation angle. However, no evidence is provided for this statement, and it is also not clear to what extent variations in the aerosol load could lead to systematic errors in the retrieved NO2 VCD. Therefore, some sensitivity studies on the impact of aerosols on the retrieved tropospheric VCD should be performed or appropriate references should be provided.

See **AR 2**.

1.2 Specific comments
P28899, L9: It is not clear if 'one kilometer' refers to the distance along the LOS, the vertical, or the horizontal dimension. Please clarify.

AR 4: This line has been rephrased (see rev.man., p. 4, beginning of last paragraph).

Section 2.1: A technical description of the instrument (instrument type, entrance optics, wavelength range, spectrol resolution, spectrometer, detector, field of view, etc.), as well as according references, are missing here. Also, the elevation angle sequence should be specified (although only 30◦ is used), as well as the time required for one sequence. It is not clear what a 'mini MAX-DOAS' instrument is.

See **AR 2**

Section 2.2: Although the retreival has already been described elsewhere, a summary of the quality of the retrieval (typical RMS residual, random and systematic errors, detection limit) obtained during this campaign should be provided.

See **AR 2**

P28901, L17: It is not true that 'the differential cross sections corresponding to the various trace gases are mutually orthogonal'. They are not orthogonal, but linearly independent (which is sufficient for the separation in a fitting procedure).

AR 5: This has been adapted (rev.man., p. 7).

P28902, L6: Describe what a 'Ring' cross section is, and add according reference (e.g., Chance and Spurr, 1997).

AR 6: This has been done (rev.man., p. 7).

P28902, L20: Have the spectra been averaged prior to the analysis or has the average dSCD been calculated after the analysis?

AR 7: this is described now in the paper (rev.man., p. 8, second paragraph).

Section 2.2.1: As already mentioned in the general comments, this section should appear after 2.2.2, since it is difficult to understand the impact of clouds without knowledge on the airmass factor concept and the way simulations of the radiative transfer were performed. The nomenclature specified in Section 2.2.2. should be used in 2.2.1.

See **AR 1.**

P28904, L3: 'Iα 0 is the simulated sky radiance without NO2 and IN α O2 is the simulated sky radiance with N O2 at a certain altitude z.

AR 8: This has been changed (rev.man., p. 12)

P28904, L22: Are you referring to Lidar observations of NO2?

AR 9: Yes (see rev.man., p.13, first paragraph)

P28910, L10: I would say that the agreement between model and measurement presented in Figure 4 is better than 'reasonable' .

AR 10: Thank you. (rev.man., p. 16, first paragraph)

P28912, L13: Here it is argued that cloud-free cases show better agreement because the systematic difference between model and measurement is better for cloud-free than for cloudy cases. On the other hand, the numbers in Table 2 show that the correlation coefficient is better for cloudy covered than for cloud-free cases, from which one could draw the opposite conclusion.

AR 11: This observation has been included in the manuscript (rev.man., p. 18, first paragraph)

P28912, L23: It is not clear whether you have performed these simulations yourself or you have adapted these findings from somewhere else.

AR 12: A part of this section has been removed from the manuscript (p.28912 l.16- p.28913 l.4 of original manuscript), also because of a remark on the elements discussed here by the second referee. The conclusions drawn from the simulations described here (that a slope unequal to one could indicate a different spatial representativity), could not be confirmed by new simulations done with new code written from scratch. It is however important to remark that the fitting results (slope and intercept of linear fit) reported in the paper have been thoroughly checked before resubmitting the manuscript. For example, the results of the slope do not change by more than 0.01 when interchanging x and y in the fitting procedure. To avoid any possible confusion: of course the 'new slope' (after changing x and y) was compared with the reciprocal of the original slope, and also the intercept was only compared after the proper conversion.

P28913, L28: It is stated that summer months were excluded from the comparison because the model would not perform well in this period. I would appreciate if this data would be shown, since this would allow to quantify the differences between model and measurement and to investigate the reasons for the model uncertainties.

AR 13: There is a possible misunderstanding. The reason that summer months were not included in the comparison is not (as understood by the referee) that the model would not perform well in this period. The authors fully agree that it would be interesting to check this by comparing with ground-based observations. In fact, summer months are not included in this study because unfortunately no MAX-DOAS data was available in this period because of instrumental problems (as described in Sect. 2.1 of revised manuscript, second paragraph). The subsequent remark about the results from Huijnen et al. (2010) is included in order to provide the reader with information about the quality of the model in the summer months based on other observations, namely OMI satellite observations.

Figure 11 should be removed, since it shows almost the same map as the right panel of Figure 3, except for the additional arrows which are not really necessary for the discussion in Section 4.4.

AR 14: The authors prefer to not remove this figure. It was originally added after several readers of the draft version were confused about the discussion in Sect. 4.4, where the dependence of the tropospheric NO_2 column on wind direction is discussed for two different sites. It is important that readers of this section realize how differences between figures 11 and 13 (rev.man.) are related to the relative positions of De Bilt, Cabauw and Utrecht. We think that the arrows on the map of Fig. 12 (rev.man.) which refer to the arrows in Fig. 13 (rev.man.) and which are extensively used in the text have added value.

P28918, L15ff: A dependence of NO2 VCDs on wind speed has been observed. What are possible reasons for this dependence?

AR 15: A possible explanation has been added to this paragraph (rev.man., p. 23): "A possible explanation for this effect is that with higher wind speeds the NO2 emitted in a certain time period is distributed
over a larger volume of air than with lower wind speeds. This reduces the observed tropospheric column downwind of the source region."

P28919, L22: It is not clear what is meant with '...despite a strong variability in both data sets...'. Actually, a high variability in the observed parameter should be of advantage when performing a statistical comparison between two independent data sets.

AR 16: This phrase has been removed from the text, and also from the abstract.

P28921, L19: It is not clear what is meant with stronger oscillations in the monthly cycle.

AR 17: This line has been rephrased (rev.man., p. 26, l.3-5).

1.3 Technical corrections

AR 18: all technical corrections listed below have been taken into account in the revised manuscript.

P28896, L11: Replace 'done' with 'performed'.
P28896, L13: Replace 'a viewing elevation angle is used of 30◦ above the horizon' with 'a viewing elevation angle of 30◦ above the horizon is used'.
P28896, L28: 'A correlation of 0.72 was found'.
P28897, L10: Replace 'active nitrogen' with 'reactive nitrogen'.
P28897, L15: 'In addition, NOx ...'.
P28897, L24: 'a comparison of tropospheric NO2 column forecasts over Europe for 2008/2009'.
P28898, L5: Replace 'Whereas' with 'While'.
P28899, L4: Replace 'For concentrations' with 'In terms of concentrations'.
P28900, L7: Delete 'MAX-DOAS'.

P28900, L23: Insert a comma after 'In total'.
P28901, L1: Replace 'at' with 'on'.
P28909, L20: Replace 'equivalent' with 'equal'.
P28911, L2: 'As in this situation the modeled wind comes...'.
P28911, L21: Remove second occurrence of 'this day' in this sentence.
P28912, L4: Replace 'Gaussian' with 'Gaussian distribution'. Mention that σ = 5.5·1015 molec cm−2 refers to all data.
P28914, L15: 'The average diurnal cycle'.
P28914, L16: '... only data from the months September, October (2008), and March and April (2008 and 2009) were used ...'.
P28915, L5: 'The dependence of tropospheric NO2 columns from MAX-DOAS and Lotos-Euros on various meteorological parameters was investigated:'.
P28916, L18: replace 'three hundred thousand' with '300 000'.
P28921, L5: 'For daily averaged tropospheric N O2 columns a correlation of 0.72 is found, and a linear regression shows that ...'.
P28921, L13: '...low weekly cycle in the modeled emissions'.
P28921, L19: Replace 'monthly cycle' with 'seasonal variation'.
Caption of Fig. 3: Replace 'High emissions in the North Sea catch the attention, but note that these have a large uncertainty' with, e.g., 'The high emissions present in the North Sea are subject to a large uncertainty'. Replace 'De' with 'The'. The map on the right side is not topographical.
Caption of Fig. 5: Replace 'pabel' with 'panel'.
Caption of Fig. 6: Replace 'which is argued' with 'as discussed'. Remove 'the' before 'Sect.'.
Caption of Fig. 7: 'In black the number of hourly averages for that month is shown ...'

2.0 Comments by Anonymous Referee #2
Received and published: 21 December 2011

2.1 General comments
This paper compares nitrogen dioxide measurements from a ground based MAX-DOAS instrument to the Lotos-Euros air quality model. Potential sensitivities and biases of MAX-DOAS and the model are investigated and discussed, using data fromDe Bilt and Cabauw, the Netherlands. In particular, the potential impact of partial cloud cover on MAX-DOAS measurements is explored using air mass factor calculations, cloud height measurements, modelled boundary layer heights, and long-term datasets. Through an investigation of comparisons over varying temporal ranges and meteorological conditions, the ability of MAX-DOAS to accurately measure nitrogendioxide concentrations in semi-polluted environments is explored. Overall agreements between the MAX-DOAS measurements and the LOTOS-EUROS model are impressive, with credible explanations given for discrepancies. The methodology overall is robust and the paper well-written. This work merits publication in ACP, with the following recommended amendments:

It would be good to define the "urban" nature of the De Bilt measurement site. Many of the conclusions drawn in this paper depend on the definition of urban or semi-urban measurements, implying the extent to which local emission sources are mixed into the polluted boundary layer. For example, are there any major emission sources near the instrument, or its line of sight? Presumably not, and the lack of variable and major local emission sources may be necessary for such close agreements between large scale models and MAX-DOAS measurements. A mention (or map) of local emissions near measurements (or lack thereof) would be beneficial, and would inform conclusions of the applicability of the agreements found in this analysis.

AR 19: The authors agree with the reviewer that a map with local emissions would be interesting, but unfortunately such a map was not available to us. The best available emission map is shown in Fig. 4 (of the revised manuscript). We would like to point out the following sentence in the paper, where we think the nature of the site is in fact already described as requested here: "This measurement site can certainly not be characterized as rural, with several highways and local roads around it and only four kilometers from the city center of Utrecht (approximately 300 000 inhabitants)" (rev.man. p.5, l.2). Furthermore, it is mentioned in the paper (rev.man., bottom of p.22) that "the relatively large agreement of the wind-direction dependence between the (semi) urban De Bilt and rural Cabauw sites indicates that for tropospheric column observations, the distinction between rural and urban sites is not so important (for a model or satellite versus MAX-DOAS comparison) as in the case of in-situ observations, Blond et al. (2007)". In our view, the use of the word 'indicates' implies that more observations are needed to confirm this hypothesis or to shed new light on this issue. Unfortunately, such observations are not available to us at present.

For analysis of cloud bottom height - please add an explanation of why was the minimum hourly height used rather than the average.

AR 20: An explanation is given on p.14 (rev.man.): "In the case of mixed cloud conditions, the minimum cloud bottom height in the one hour time interval was used. This procedure was followed instead of using the mean cloud height primarily because of two reasons. First, when the cloud conditions vary between cloudy and cloud free, it is not trivial how to define a mean cloud height. Second, because the minimum cloud bottom height H c provides more than the mean cloud bottom height a good estimate of the boundary layer top height H b. Therefore Eq. 5 approximates Eq. 4 better when using the minimum cloud height than when the mean cloud height was used (the mean could be affected by high cirrus and therefore be considerably higher than H b)."

Use of ECMWF boundary layer height is key to a part of the analysis within this paper, and yet no error estimate on this data product is mentioned. Please include one if it is available.

AR 21: We added a reference to a study which addresses the accuracy of boundary layer heights in models (Seidel et al. (2012), rev.man. p.13), and we would like to refer to Table 2 (rev.man.) where the quite low sensitivity of the tropospheric NO2 column to the NO2 and aerosol layer height is reported.

The conclusion of line fitting on page 28912 is an interesting one. If this has a statistical or mathematical foundation, please reference it. This may perhaps be an artefact of the particular mathematical line-fitting routine used in this case.

See **AR 12**

2.2 Minor typographicals

AR 22: all technical corrections listed below have been taken into account in the revised manuscript.

p28898 - line 11 - replace "one-sidedness" with "bias"
line 24 - run at 7 x 7 km resolution for the Netherlands and surrounding area.
line 29 - are determined and used to convert.
C13506
p28901 - line 23 - In this study a 3rd order polynomial was used.
p28902 - please add a suitable reference for the 30 degree viewing elevation angle parameters discussed.
p28904 - line 8 - I would recommed that the word "always" is replaced by "typically" - as there have been a few examples of data retrieved using non-zenith reference spectra.
p28907 - line 8 - remove the word "in" - and often over many days.
p28908 - line 16 - given by the boundary layer height
line 24 - the cases was more than 10% of the NO2 found above
p28909 - line 21 - equivalent to the resolution of the emission inventory.
p28910 - line 4 - use "significant" instead of "striking"
p28912 - line 7 - the impact of processing different subsets.
p28913 - line 19 - Figure 7 shows monthly averages of tropopsheric NO2 concentration.... etc.
line 20 - The observations show..
line 24 - (and other places) - repace "less" with "fewer"
p28915 - line 1 - emission of NO2 due to
p28917 - line 4 - replace "quite some" with "considerable"
line 8 - It is hypothesized that the loss in tropospheric... is compensated by the NO2 added..
p28920 - line 29 - leading to larger..
Figure 3 caption - High emission in the North Sea are a dominant feature, however it should be noted that these have a large.. De/The MAX-DOAS The map on the right does not show elevations, and is therefore not topographical.
Figure 5 caption - left panel (not pabel)

3.0 Other changes compared to the previous version of the manuscript:

The list of co-authors has changed compared to the initial submission of the manuscript (M. Schaap and F.J. Sauter have been). Also there are several changes in the authors' affiliations.