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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Transport and build-up of tropospheric trace gases during the MINOS campaign: Comparision of GOME, in situ aircraft measurements and MATCH-MPIC-data" *by* A. Ladstätter-Weißenmayer et al.

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Replies to the comments of reviewer #2:

First of all we would like to thank the reviewer for his valuable comments which in our opinion helped to improve the paper significantly. We have implemented most of the suggested changes and answered to his questions in detail below.

General Comments:

The paper concentrates on two major aspects:

a) to compare (and validate) GOME, and in-situ measurements as well as model results

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of tropospheric NO2 and HCHO.

b) to investigate the origin, transport and importance of tropospheric pollution in the summer Mediterranean region.

Both aspects are of great interest and significance for the scientific community and the results are convincing in its own. However, the motivation, the presentation, and moreover the logical structure of the presented work has several important shortcomings.

In some parts of the manuscript it takes the reader (at least me) a lot of time to assess what the authors might have wanted to tell (and why) I hope that the authors are willing to spend considerable efforts to improve these aspects, because their interesting results should be made available to the scientific community as soon as possible.

In view of the critical remarks of the reviewer, we have tried to improve both the logical structure of the paper and to clarify the main points discussed. We hope that the paper is now more accessible and can be accepted for publication in ACP.

1. Page 2, lines 1-2: 'trace gases NO2 Maybe in an earlier version of the manuscript the authors had combined the statements for NO2 and HCHO. This was possibly a good choice; in the current version this paragraph is a little bit confusing.

The statements for both trace gases have been combined as requested.

2. Page 2, line 30: tropospheric H2O might be included in the list.

The trace gas H2O has been included. (The main scientific objective of GOME is to measure the global distribution of O3 and several other trace gases which play an important role in the ozone chemistry of the earth's stratosphere and troposphere, e.g. NO2, BrO, OClO, H2O, SO2 and HCHO).

3. Page 2, line 43: Does this mean that a given GOME pixel (320x40km2) is treated as cloud free if the cloud fraction of all three GOME pixels lying side by side is <10%. Why did the authors chose this procedure? Why is then e.g. a GOME pixel without

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clouds rejected if there are clouds only in the neighbouring pixels?

This is a misunderstanding. Of course cloud selection is applied to individual pixels and does not depend on the cloud cover of neighbouring pixels. We have changed the corresponding paragraph to clarify the procedure.

4. Page 2, line 43: How do the authors define cloud fraction?

The cloud fraction used in this paper is the operational ICFA product. We have added this information to the text.

5. Page 2, line 43: Do the authors think that a cloud fraction of 10 % can be neglected? Since clouds are usually much brighter than the clear part of the GOME pixel even 10% cloud cover can contribute to up to 70% of the total signal received. Do the authors correct for this potential cloud influence?

We agree with the reviewer that even small sub-pixel cloud contamination can have a significant impact on the retrieval of tropospheric species, in particular in the UV wavelength range. This has been discussed in several publications (Velders et al., Richter et al., Randall et al.), and some groups try to account for it by applying global correction factors based on statistical arguments (Leue et al.) or pixel specific factors based on cloud information and assumptions on the vertical distribution of NO2 (Randall et al.). In this study, we have not corrected for cloud effects as we wanted to avoid the introduction of additional assumption in the data analysis. As a result, the values in cloudy pixels could be too low.

6. Page 3, line 9: The simultaneous use of 'ppm' and '%' is a little bit confusing here. It might be good to replace the error by an absolute value (0.03 ppm?)

This sentence was replaced as follows: Before every flight a calibration of this instrument was performed using a diluted mixture of 2.97 ppmV NO ($<$ 1 %) in N2 (Messer Griesheim) with purified air.

7. Page 4. line 3: for NO2 and HCHO different days of GOME measurements are

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analysed and presented (Fig. 1). Why?

This figure has been modified. The problem was, that not on all days when in-situ measurements were carried out for NO2 there were also HCHO data. As a result, 7 comparisons could be carried out for HCHO but only 6 comparisons for NO2. This is now explained in the text as follows:

Due to the limited temporal and spatial overlap of GOME and aircraft data (e. g. on the 22nd August 2001 when HCHO data are available but no NO2 measurements were performed), only the data of 6 days for NO2 (one flight on the 1st, 12th, 16th, 17th and two flights per day on the 14th and 19th of August 2001) and 7 days (one flight on the 1st, 12th, 16th, 17th, 19th and two flights per day on the 14th and 22nd of August 2001) for HCHO during the MINOS campaign could be analysed and compared (see fig. 5 and 6).

8. Page 4, line 11: What do the authors mean with 'contribution to the information'? If they mean something like 'sensitivity' I don't agree with this statement (although it is true for many cases). However, the sensitivity depends strongly on the solar zenith range, and low zenith angle the sensitivity (often expressed as AMF) for tropospheric species can be significantly higher than for the stratospheric species.

We agree that this sentence was confusing and therefore have rephrased it. In clean regions, the NO2 signal measured from space is dominated by the stratospheric NO2 column. Also, in most cases, the sensitivity of the measurements for a stratospheric absorber is larger than that for an absorber in the boundary layer (see Fig. 7 and 8 and their discussion). To our knowledge, only very special conditions (ice, high sun, enhanced aerosol for multiple scattering, very thin cloud) can lead to a situation where the airmass factor for tropospheric species is larger than for stratospheric species. For the measurements discussed in this paper, this is certainly not the case.

9. Page 4, lines 15-27: I found this section very confusing! What is at least missing is a clear description (or mentioning) of each step performed in the determination of

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the tropospheric columns. As an alternative mentioning adequate references might be sufficient.

We have tried to clarify this section and to give the reader a clearer picture of what has been done in the analysis of the satellite data.

10. Page 5, line 6: I don't fully understand the statement beginning with 'and additionally'. What do the authors mean with 100% efficiency? Of course, it is clear that especially for low ground albedo the AMF for boundary layer profiles becomes low; however, if an appropriate AMF is applied the true VCD is derived (with 100% efficiency').

The reviewer is right, this statement was misleading. It has been rephrased accordingly.

11. Page 6, section 4. Results and discussion: This kind of information would have been helpful already in the introduction. Please also make clear what is meant with: 'The second aim was to interpret these data with the results of the global detected GOME data'.

The part 'introduction' was modified in view to the scientific aim of this paper. The second aim was to interpret these data combined with the amounts of trace gases retrieved from GOME data with respect to the transport and formation of tropospheric constituents during this campaign.

12. Page 8, line 38: What do the authors mean by 'sensitivity regarding to the density of the radiance?

See next comment.

13. Page 8, section starting with line 38: What is the motivation to introduce weighting functions here? So far the authors have used AMFs as a measure of sensitivity of GOME to specific trace gas profiles. Essentially, AMFs for box profiles at different layer heights seem to be similar to the weighting functions concept introduced here. I suggest three possible options: a) To totally remove this section because is does

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not significantly contribute to the findings and conclusions of the paper. B) To replace this section by a formulation consistent with the AMF concept. C) To at least explain and motivate why this section was introduced, explain all quantities in a sufficient and consistent way and give appropriate references for the concept.

This section was introduced to give the reader an impression on the height dependence of the sensitivity of GOME measurements of NO2 and HCHO. This is an important information when comparing in-situ measurements with satellite data, in particular for absorbers that have a maximum in concentration close to the surface. We agree with the reviewer that the height dependence can be discussed in terms of weighting functions, block airmass factors or averaging kernels (see the recent ACP paper by Eskes et al.). Here, we decided to use weighting functions as they are much more common in literature than the block airmass factors, which mainly are used by the DOAS community. As suggested by the reviewer, we have rewritten this section to make it clearer and more consistent with the rest of the paper. Also we have converted the figures from mixing ratios to concentration units and moved the whole paragraph to the section discussing GOME data analysis.

14. Page 10, line 3: What about trajectories starting at the ground?

No trajectories starting below 900 hPa have been considered. The reason is that turbulence on spatial and temporal scales smaller than the resolution of the meteorological data dominate the transport within the planetary boundary layer.

15. Page 10, lines 3 to 7: Why are the levels given in two different measured (meter and hPa)?

The levels are now only in units of hPa, as follows: All back trajectories start over Crete at different heights (at 900, 800, 700, 600, 500, 400, 300 and 200 hPa) and travel backwards in time for 5 days.

16. Page 10, line 8: What is 'traj.x'? Give at least a reference. With whom did the

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authors have their [private communication]?

The following reference describe in detail explain ‘traj.x‘.

Meyer-Arnek, J., "Numerical validation, manual and userguide for Traj.x", iup-Bremen, 2002.

17. Page 10, line 32: How is the ‘Total Totals‘ index calculated? Give at least an adequate reference.

The Total Total index is now explained in the text:

The Total Totals Index (TTI) is commonly used in meteorology. This index is derived from the temperature lapse rate between 850 and 500 hPa (T(850) and T(500)) and from the moisture content at 850 hPa (Tdew(850)).

$TTI = T_{dew}(850) - T(500) + (T(850) - T(500))$

18. Page 11, line2: What is the authors view is the difference between ‘comparison‘ (NO2) and ‘validation‘ (HCHO)? As far as I followed the paper both trace gases have been investigated in a similar way.

The reviewer is right. The sentence was changed as follows: The case studies presented here show comparisons for the trace gases NO2 and HCHO between satellite based GOME measurements and in-situ data.

19. Page 11, line 10: What do the authors mean with ‘based on GOME data‘?

This sentence was modified as follows: By calculating weighting functions it was demonstrated that the GOME results contain information down to the BL.

20. Figure 1a): Have these excess NO2 VCDs been calculated using AMFs for ‘standard‘ profiles? And for which ones? What is the reason for the white gaps?

In figure 1a) the used AMF is based on the measured mean NO2 profile calculated for this MINOS campaign. The white gaps reflect pixels with cloud cover higher than 10%

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or a measurement error above a threshold value.

21. Figures 1a) 1b): For HCHO different days are presented compared to NO2. Why? Also the white gaps are more frequent and at different locations. Why?

This is now explained in the text:

Due to the limited temporal and spatial overlap of GOME and aircraft data (e. g. on the 22nd August 2001 no NO2 data were available in contrast to HCHO), only the data of 6 days for NO2 (one flight on the 1st, 12th, 16th, 17th and two flights per day on the 14th and 19th of August 2001) and 7 days (one flight on the 1st, 12th, 16th, 17th, 19th and two flights per day on the 14th and 22nd of August 2001) for HCHO during the MINOS campaign could be analysed and compared (see fig. 5 and 6). White gaps are more frequent and at different locations for HCHO than for NO2. This is a result of the error threshold applied in the data analysis that excludes more HCHO than NO2 values as the latter usually gives better fit results.

22. Figure 3: It seems that the average NO2 profile' is significantly higher than the true average of the individual profiles.

The average NO2 profile has been corrected in the current version.

23. Figures 5, 6: Figures are a little bit confusing and hard to read. Please increase the font size and remove unnecessary information.

The figures 5 and 6 were modified by increasing the font size and removing unnecessary information.

24. Figure 7, 8: I think these Figures are misleading. They display the change in the column density as function of change in mixing ratio. It is clear that the change in concentration for a given change in mixing ratio depends strongly on altitude. Accordingly, the sensitivity of GOME decrease with altitude above about 4 km (for a given change in mixing ratio). In reality the sensitivity (for a given change in concentration) is almost constant above 4 km. Also the x-axis do not start at 0 which makes the curves even

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more misleading.

We agree with the reviewer that giving the sensitivity in terms of concentrations is more appropriate for a DOAS measurement (that basically is counting molecules) than the use of mixing ratios. However, as we learned during the preparation of the paper, the use of mixing ratios is more convenient for many people involved in in-situ measurements as there measurements usually are given as mixing ratios. Still, we have changed the figures as requested and have also included the origin in the plots to make it even clearer.

25. Fig. 9, 10: What was the reason to select the period from 01-03 August? What about the other days?

The reason to select the period from 01-03 August was to show in a case study the transport of polluted air masses over the Mediterranean region. That means the analyses of satellite based GOME data in combination with calculated trajectories makes it possible to get information of the origin of these polluted air masses.

Interactive comment on Atmos. Chem. Phys. Discuss., 3, 3051, 2003.