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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Tropospheric O_3 over Indonesia during biomass burning events measured with GOME (Global Ozone Monitoring Experiment) and compared with trajectory analysis" *by* A. Ladstätter-Weißenmayer et al.

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General comments :

The manuscript by Ladstätter-Weißenmayer et al. presents tropospheric columns of ozone (O3), nitrogen dioxide (NO2), and formaldehyde (HCHO) derived from measurements of the GOME instrument for September 1997 and September 1998 over the Indonesian region. A strong enhancement of all three trace gases was found for September 1997 (compared to September 1998) consistent with enhanced fire activity in 1997. A combined trajectory-chemical model is used to investigate these obser-

vations. The analysis yields that 3.1 Tg ozone was produced as a result of biomass burning. Enhanced tropospheric ozone over the Indian Ocean is explained by mixing of lightning influenced air masses with high NOx concentrations with air masses affected by biomass burning. Overall, this manuscript does contain some interesting new information.

Criticism:

The main limitations of the present manuscript are that the results obtained in this work are not set into the context of the information available in the published literature and that some assumption in the chemical modelling significantly limit its value as a tool to analyze the GOME observations. The manuscript should only be considered for publication in ACP after significant modifications are incorporated.

For better readability, this annotation is divided into two steps (1.1) to (1.2).

1.1 Annotation:

(1.1) There is a substantial amount of published information available on observations and the interpretation of the enhanced tropospheric ozone column over Indonesia in August - October 1997. This work has not been cited adequately. Especially the works of Chandra et al., 1998, Geophys. Res. Lett., 25(20), 3867-3870 (TOMS observations), Thompson et al, 2001., Science, 291, 2128-2132 (TOMS observation), Chandra et al., 2002, J. Geophys. Res., 107(D14), doi: 10.1029/2001JD000447 (TOMS observations and global model interpretation), and Duncan et al., 2003, J. Geophys. Res., 108(D15), 4458, doi: 10.1029/2002JD003195 (global model simulations) should be referred to in the manuscript and the results of the present study should be discussed in the context of these studies.

Answer:

We thank the author for his fair comments on our study. His suggestions contributed to improvements of this publication and will certainly meliorate further studies on this

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issue.

The suggested references above are now included in the introduction of the paper. All these studies are strongly linked to wildfires over Indonesia and give insight into:

- 3D-modelling aspects and sensitivity studies performed by applying the MOZART model (Hauglustaine et al., 1999) - Increase of total ozone over Indonesia during the October of El Nino years (Kita et al., 2000) - Increase and maximized of tropospheric ozone over Indonesia during the October 1997 based on the TTO (Tropical Tropospheric Ozone) from TOMS (Total Ozone Mapping Spectrometer) (Thompson et al., 2001) - The impact of the Indonesia wildfires of 1997 on the tropospheric chemistry: Application of the GEOS-CHEM model on the Indonesian wildfire episode and analysis of the transport of air masses and radiative forcing resulting from gaseous and particle releases (Duncan et al., 2003) - The increase of tropospheric ozone in the western Pacific and the simultaneous decrease in the eastern Pacific is varying inversely with the upper tropospheric water vapour mixing ratio during the course of the 1997-1998 El Nino. (Chandra et al., 1998) - Impacts of dynamical and chemical effects onto tropical tropospheric ozone columns (Chandra et al., 2002)

All these above studies are to be seen as being complementary with respect to our study, although their individual aim differs from our work. Our work concentrates of the direct impact of the Indonesian wildfires on the tropospheric ozone NO2 and HCHO concentrations in this region. Additionally it accounts for ozone production due to possible mixing of air masses being subject to long range transport.

Nevertheless, references and discussions with respect to the above list of publications is now included into our work.

Kita, K., M. Fujiwara, S. Kawakami, Total ozone increase associated with forest fires over the Indonesian region and its relation to the El Nino-Southern oscillation, Atm. Env., 34, 2681-2690, 2000

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Duncan B. N., R. V. Martin, A. C. Staudt, R. Yevich, and J. A. Logan, Interannual and seasonal variability of biomass burning emissions constrained by satellite observations, J. Geophys. Res., 108 (D2), 4100, doi:10.1029/2002JD002378, 2003.

Duncan B. N., I. Bey, M. Chin, L. J. Mickley, T. D. Fairlie, R. V. Martin, H. Matsueda, Indonesian wildfires of 1997: Impact on tropospheric chemistry, J. Geophys. Res., 108 (D15), 4458, doi:10.1029/2002JD003195, 2003.

Hauglustaine, D. A., G. P. Brasseur, and J. S. Levine, A sensitivity simulation of tropospheric ozone changes due to the 1997 Indonesian fire emissions, Geophys. Res. Lett., 26(21), 3305-3308, 1999

Chandra, S., Ziemke, J. R., Min, W., and Read, W. G., Effects of 1997-1998 El Nino on tropospheric ozone and water, Geophys. Res. Lett., 25(20), 3867-3870, 1998.

Chandra, S., J. R. Ziemke, P. K. Bhartia, and R. V. Martin, Tropical tropospheric ozone: Implications for dynamics and biomass burning, J. of Geophys. Res., 107, 14, doi:10.1029/2001JD000447, 2002

Annotation:

(1.2) One particular focus of the previous studies was the separation of the contributions to the enhanced tropospheric ozone column from dynamical processes induced by the El Nino circulation and the photochemical production of ozone initiated by the biomass burning emissions. Most studies based on TOMS ozone observations and model simulations conclude that both effects contribute about equal to the observed enhanced ozone with some spatial and temporal variation. The present work does not separate the different processes leading to enhanced tropospheric ozone (biomass burning vs changes in the atmospheric circulation), but explains the ozone enhancement solely to photochemistry associated with biomass burning pollution. The authors do not take advantage of the additional observations of HCHO and NO2 from GOME in the present work that might allow a better constrained estimate of the dynamical vs

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chemical contributions than previous studies. This issue should be investigated and discussed before publication in ACP.

Answer:

Our study compares the transport of pollutants during September 1997 with the long-range transport during September 1997. It turns out that the circulation was equivalent in both years when considering the long-range transport of emissions from biomass burning over Africa and Australia. Therefore we concluded that only changes in the local emission can lead to such a dramatic accumulation of ozone, NO2 and formaldehyde which is locally tied to the Indonesian region.

By the means of the 3D-chemistry transport model GEOS-CHEM (Chandra et al., 2002) the impact of the circulation and of the local chemistry onto the ozone loading was differentiated. This is indeed a very interesting and promising approach which is - to our opinion – far out of reach for a transport and box model analysis.

Our study contains a comparison of tropospheric trace gas amounts between September 1997 and September 1998 for the Indonesian region. The scientific aim of our paper was to discuss the differences of tropospheric columns of O3, NO2 and HCHO between these episodes. It was accounted for the photochemical evolution of the emitted trace gases. Any further dynamic analyses which possible influence its chemistry are not examined.

Annotation:

(2.1) There are serious problems with some assumptions in the chemical model simulations. My main concern is that only a very limited set of the known emission products (based on observations over Africa) were used in the model simulations, and that the emissions were modified in such a way that the model results show 'the best agreement between GOME retrieval and model results for ozone, NO2, and HCHO.' [page 3113, line 18], i.e., the model was tuned to match the observations.

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The authors claim that 'hardly any information about VOC released by peat fires is available' [p. 3112, line 21f], and that it is feasible to use observations obtained over Africa in 1992 for the initialization of the model. There is, however, information available for VOC emissions from peat fires, e.g., the study by Christian et al., 2003, J. Geophys. Res., 108(D23) doi: 10.1029/2003JD003704, which presents emission factors for numerous VOCs from the burning of Indonesian peat. This information should be used in the model simulations. This extensive set of emission factors will also allow a more realistic representation of plume photochemistry by adding numerous compounds that are potentially important for the photochemical production of ozone, e.g., acetone. Atmospheric oxidation of these compounds should be included in the model simulations.

Answer:

The basic idea of this part of our model study was to get insight into the photochemically produced amount of ozone, NO2 and HCHO. By varying the model initialisation to match the GOME observations of these trace gases, levels of CO comparable to airborne in situ measurements were finally obtained. CO measurements have (among others) been performed by Matsueda et al. (1999) during September 1997. As a result, our approach can be understood as a very simple inverse modelling approach.

By our model analysis we derived one average chemical model initialisation which was applied to all trajectories. It includes contributions of emissions from flaming and smoldering of woods, peat and grasslands. A distinction of the individual trajectory's origin was not performed.

Our work concentrated on the chemical evolution of air parcels already being outside the flames. In chapter 2.2 it is stated that the initialisation data set represents "air masses which recently left the fire, but are already cooled down to environmental temperatures, so that the complicated fire chemistry does not have to be taken into account by the box model". However, due to nonlinear dependencies within the plume photochemistry, our assumption of an "effective" emission may lead to questionable results.

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Including further volatile organic compounds (VOC) will certainly improves this analysis, although it has to be considered as second step. Numerous emission factors for VOC (depending on the fuel type) are published listed by Christian et al., 2003 (table 6) for African and Indonesian fuels. In order to really account for the initialisation of each air parcel representing burning, the fraction of the different burning regimes on the totally burnt area is to be considered.

Such a detailed analysis would certainly be very interesting. However this would be an approach which is totally different from the one chosen for our study.

Concerning acetone: Acetone is included into the chemistry scheme (as an oxidation product). Nevertheless it is not initialised itself.

Additional information on our approach is now given in chapter 2.2.

Christian, T. J., B. Kleiss, R. J. Yokelson, R. Holzinger, P. J. Crutzen, W. M. Hao, B. H. Saharjo, and D. E. Ward, Comprehensive laboratory measurements of biomass burning emissions: Emissions from Indonesian, African and other fuels, J. Geophys. Res., 108, 23, doi: 10.1029/2003JD003704, 2003.

Matusueda, H., and Hisayuki, Y. I., Aircraft measurements of trace gases between Japan and Singapore in October of 1993, 1996 and 1997, Geophys. Res. Lett., 26, 16, p. 2413-2416, 1999.

Annotation:

(2.2) There might be some scientific value to tune the fire emissions in such a way that the model results are close to the GOME retrieved trace gas columns. The authors should, however, present a more detailed description, how this adjustment was performed. (Just by visual comparisons or by a systematical strategy to minimize the difference between model results and observation?).

Answer:

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The modelled amount of ozone, NO2 and HCHO being photochemically generated due to the Indonesian fires was compared with tropospheric ozone, tropospheric NO2 and HCHO as retrieved from GOME.

For the initialisation of the model, a sensitivity study was performed (which is in detail described in our paper). First, an initialisation data set representing biomass burning emissions released from fires over Africa is applied. All air parcels which were followed by the transport and chemistry analysis are initialised with this data set. Secondly, the VOC and NOx loading of this data set was increased by certain factors. E. g. the VOC loading was increased by factor of 2, 4, 10 and 20.

The comparison of the modelled with the observed trace gas columns of ozone, NO2 and HCHO takes into account its amount (mass) in a defined area, its maximum value and its extension. The amount is computed by integrating the mass of the particular trace gas in an area expanding from 70 to 160° E and from -10 to 10° N. The assumption is that in 1997 this region was mainly impacted by the Indonesian fires; impacts from other sources on this region are regarded as minor.

Secondly the maximum column amount was used as criterion. In addition the extent of enhanced trace gas columns was visually compared.

Results from the model initialisation which led to the best agreement was finally chosen for this study.

Annotation:

(2.3) The statement that 'the differences between the modelled and retrieved tropospheric columns are to be considered as relatively small.' [p. 3116, line 27f] does not hold much scientific value in this case, since the model was tuned to match the observed columns.

Answer:

The initialisation of the chemistry model was adjusted to match the tropospheric

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columns of ozone, NO2 and HCHO retrieved from the GOME instrument. It turns out that the initialisation data set derived from this matching analysis is compliant with air borne measurements performed over Indonesia during this episode (Matsueda et al., 1999) (see answer to annotation 2.1). As a result, this analysis shows that by performing a relatively simple transport and chemistry analysis, reasonable initialisation data can be obtained. Further improvements of the model initialisation (including additional VOC, accounting for emission factors) can be regarded as a next step leading to future studies on this subject. For further details see answer to annotation 2.1.

2.4 Annotation:

(2.4) In my opinion, the model simulations have to be improved substantially for publication in ACP. They should be conducted using the full suite of known VOC emissions from Indonesian peat fires and without any tuning of the emissions. As in previous studies the fire emissions should be based on information of the burned fuel (as a function of space and time) and available emission factors. I am aware that this is a major task and will require some time. I do think that by using the additional information from GOME (i.e., NO2 and HCHO) combined with appropriate model simulations the contribution of biomass burning to the enhanced tropospheric columns of ozone can be better constrained than in previous studies, that relied on ozone measurements only.

Answer:

We are of the opinion that the reviewer's suggestions are fruitful for future approaches. Performing a forward analysis with an optimized initialisation data set and including further VOC into the chemistry model would be interesting from the scientific point of view. However the scope of this study is not supported.

Additionally it has to be stated that a 3D-chemistry transport model is probably the better choice to perform this kind of analysis.

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Conclusive Annotation:

My suggestions at this point are the following: The GOME retrieved tropospheric columns of ozone, NO2, and HCHO for 1997 and 1998 should be evaluated and explored in more detail. It would be interesting to see the temporal and spatial evolution of the tropospheric trace gas columns over the Indonesian region in the second half (June until December) of the years 1997 and 1998.

This data could be compared to and evaluated with the available TOMS data, for the whole region of maybe for the region defined in Thompson et al., 2001. In my opinion such a study, maybe also including some trajectory analysis, could provide substantial new information and insights that would allow to publish such a study without using the chemical model.

I suggest to postpone the publication of the model simulations at this point, to modify the model simulations along the lines suggested above, and to publish the model studies in a separate manuscript after the results from the improved simulations have been obtained and analyzed.

Conclusive answers

Again we want to thank the reviewer for his fruitful suggestions. The advantage of GOME measurement is the continuous data set which expands for near 10 years and is continued with the SCIAMACHY instrument. The analysis of the temporal and spatial trace gas distributions of different trace gases will remain an interesting scientific subject. For future studies many of the reviewer's suggestions will be taken into account.

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 3105, 2005.