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Atmospheric Chemistry and Physics Discussions

Interactive comment on "CO profiles from SCIAMACHY observations using cloud slicing and comparison with model simulations" *by* C. Liu et al.

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Reply to reviewer #1

Before we give our detailed answers to the reviewer comments we want to thank this reviewer very much for the important comments! Based on these comments (and also the comments from Joanna Joiner and another anonymous reviewer) we largely modified our manuscript. The major changes are described in the next sections. Following this overview, we give our detailed answers to the reviewer comments.

Major changes of the revised version:

1) Application of cloud radiance fractions (CRF): Joanna Joiner and two reviewers strongly recommended to investigate the influence of the clear part of the ground pixel to the retrieved CO columns. They argued that especially over surfaces with high albedo the contribution from the clear part of the ground pixel plays an important role. We thank Joanna Joiner and both reviewers for these important comments! Based on these suggestions we repeated our approach taking into account both the contributions from the clear and cloudy part using the concept of cloud radiance fractions (CRF). In detail we made the following changes: a) Instead of using observations with effective cloud fraction >10% we now use observations with CRF >30%. We again chose a rather low threshold to increase the number of useful SCIAMACHY observations (see also below). We found that CO profiles for CRF >30% and CRF >50% are almost identical, see e.g. (new) Fig. 4. This finding is not surprising since both contributions from the clear and cloudy part of the ground pixel are now taken into account. b) Like in the original version of our manuscript, the model data are sampled for the exact time and location of the satellite measurements. However, in contrast to the original version, we now sample the model taking into account also the contribution from the clear part of the ground pixel: For a given measurement, from the model data the CO PVCD above the cloud is extracted for the cloudy part and the total CO VCD is extracted for the clear part of the pixel. Both column densities are averaged weighted by the CRF and (1-CRF), respectively. Using this approach, the extracted CO PVCDs are substantially higher than in the original version of our manuscript, especially for high cloud altitudes. It should also be noted that due to the contribution from the clear part of the satellite ground pixel, the altitude registration of the retrieved CO profiles does not represent the true altitude. Fortunately, this has no influence on the comparison with the model results, because the models are sampled taking into account the contribution from the clear part. The new cloud selection and the application of CRF are described in detail in sections 2 and 3. It is interesting to note that using this new procedure, the substantial discrepancies between SCIAMACHY observations and model results (as shown in the original version of our paper over biomass burning regions) largely disappeared:

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the spatial patterns are now very similar in the satellite and model data indicating that the transport over this regions is well represented by the models. We discuss these new findings in detail in sections 3.3, 3.4 and in the conclusions.

2) Comparison of cloud properties derived around the oxygen-A-band with those at 2330 nm: One important concern of Joanna Joiner and the other reviewers was, whether cloud information retrieved around 760 nm was representative for the much larger wavelengths of the CO retrieval. As suggested, we used the CH4 absorption analysed from the CO fitting window to determine cloud top heights representative for the interpretation of the CO PVCDs. We considered observations with effective cloud fractions >80% to make sure that the contribution from the clear part of the satellite ground pixel can be neglected. From the comparison of the retrieved CH4 VCD with the CH4 profile from the US standard atmosphere (scaled by the latitudinal dependent average CH4 VCD for 2004, see Bergamaschi et al., 2009), an effective cloud height for about 2330nm is derived. A comparison of these 'CH4 cloud heights' with the FRESCO effective cloud height is shown in the (new) Fig. 5 of the revised version of the manuscript. Excellent agreement (slope: 1.06, r2: 0.96) is found indicating that differences in the penetration depth of photons into the clouds between both spectral ranges are small and can be neglected. From this finding we conclude that cloud information from the FRESCO+ algorithm is well suited for the application to the CO PVCDs retrieved at 2330nm. We added this information in section 2.1.

3) Validation using ground based measurements: We agree that validation of our SCIAMACHY CO profiles is important. However, in contrast to other trace gases (like e.g. O3) validation of SCIAMACHY CO profiles is a very challenging task because of several reasons: a) The uncertainties of individual SCIAMACHY CO observations are large and the global coverage is rather poor. Thus validation on the basis of individual measurements is difficult, and instead rather large numbers of measurements have to be averaged. In the revised version of our manuscript we compare time series of seasonal averages of the CO PVCDs for the lowest cloud level with independent ground

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based observations and found rather good agreement. However, from this validation exercise only little can be concluded on the accuracy of (individual) CO profiles. b) Especially over biomass burning regions no adequate validation data set (CP profiles with good temporal and spatial coverage) is available. c) As pointed out in the original version of our manuscript, the derived CO profiles constitute complex composites of very different atmospheric situations. Thus they can not directly be compared to 'real profile data' in a meaningful way. In order to provide some basic validation, in the revised version we added a detailed comparison of seasonal averages of CO PVCDs (2003 to 2005) to total CO VCDs at several TCCON stations (new section 2.2). In addition to the SCIAMACHY observations, also the model data are included.

4) Quantification of the uncertainties of the CO PVCDs: In the revised version of our manuscript we estimate the uncertainty of the SCIAMCHY CO profiles. From the comparison with the ground based observations (Fig. 7) we conclude that the CO PVCDs for effective cloud heights <0.5 km have a systematic bias of –3 % and a standard deviation of 12%. While the interpretation of the bias is complicated because of the cloud shielding of the lowest part of the atmosphere, the standard deviation can be regarded as representative for the CO PVCDs. Unfortunately, the (additional) uncertainties of the CO PVCDs for higher cloud altitudes can not be quantified from this validation exercise. They are mainly caused by uncertainties of the effective cloud heights and the errors of the CRF. We estimate these uncertainties from the uncertainties of both cloud properties (see Figs. 1 and 5 of the revised version) by assuming an average CO profile and average measurement conditions with a CRF of 60 %. The uncertainties (see new Table 1) increase with height, but are smaller than the general uncertainties of the CO retrieval as derived from the comparison of the SCIAMACHY CO PVCDs with the ground based observations. This information is added at the end of the new section 2.2.

5) New content of the paper The following figures and tables were added to the paper:

Fig. 1 Dependence of the CRF on the effective cloud fraction for different values of the

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surface albedo (see also Fig. 2). The black dotted line indicates a CRF of 30%, which is used as threshold value for the CO measurements from SCIAMACHY in this study.

Fig. 2 Global map of the surface albedo at 2130 nm over the continents from observations of the MODIS instrument (white sky albedo for the first half of March 2004, image from http://modis-atmos.gsfc.nasa.gov/ALBEDO/).

Fig. 5 Comparison of effective cloud height retrieved from the CH4 absorption around 2330 nm with the effective cloud height retrieved from the FRESCO+ algorithm (January and February 2005).

Fig. 6 Comparison of seasonal averages of the CO PVCD from SCIAMACHY and models (coloured lines) with the total CO VCD observed from ground based FTIR stations (black lines). Thick lines represent CO PVCDs above cloud heights of 0.5 km; thin lines those above 3.5 km. (units: molec/cm^2).

Fig. 7 Correlation analysis of seasonal averages of the CO PVCDs for clouds < 1km versus total CO VCDs from ground based FTIR stations. Besides SCIAMACHY measurements also the coincident results from both atmospheric models are shown. In addition to the results of the linear regression, also the ratio of the averages (RA) and the average of all ratios (AR) for all data pairs of the considered data sets are shown.

Fig. 10 Relative differences ( ) for all regions shown in Fig. 9.

Table 1 Typical errors of the CO PVCDs introduced by uncertainties of the cloud height and CRF. Uncertainties of the CRF are calculated for a measurement with CRF of 60% assuming uncertainties of the surface albedo and cloud top albedo to be about $\pm$5 %.

The following figures were modified : Fig. 3 (old Fig. 1) is updated using new CRF threshold. Fig. 4 (old Fig. 2) is updated using new CRF threshold. Fig. 8 (old Fig. 3) is updated using new CRF threshold. Fig. 9 (old Fig. 4) is updated using new CRF threshold and CRF weighting. Fig. 11 (old Fig. 5) is updated using new CRF threshold and CRF weighting. Fig. 12 (old Fig. 6) is updated using new CRF threshold and CRF

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weighting. Fig. 13 (old Fig. 7) is updated using new CRF threshold and CRF weighting.

Supplement: Fig. S1 in the supplement is updated using new CRF threshold and CRF weighting. All latitude-height and longitude-height cross sections are updated using new CRF threshold and CRF weighting.

The following references were added:

Interpretation of cloud top height and cloud slicing: Veefkind, J. P., J. F. de Haan, E. J. Brinksma, M. Kroon, and P. F. Levelt, Total ozone from the Ozone Monitoring Instrument (OMI) using the OMI-DOAS technique, IEEE Trans. Geosci. Remote Sens., 44(5), 1239– 1244, 2006.

Joiner, J., Schoeberl, M. R., Vasilkov, A. P., Oreopoulos, L., Platnick, S., Livesey, N. J., and Levelt, P. F.: Accurate satellite-derived estimates of the tropospheric ozone impact on the global radiation budget, Atmos. Chem. Phys., 9, 4447-4465, doi:10.5194/acp-9-4447-2009, 2009.

Stammes, P., M. Sneep, J. F. de Haan, J. P. Veefkind, P. Wang, and P. F. Levelt (2008), Effective cloud fractions from the Ozone Monitoring Instrument: Theoretical framework and validation, J. Geophys. Res., 113, D16S38, doi:10.1029/2007JD008820.

Vasilkov, A., Joiner, J., Spurr, R., et al.: Evaluation of the OMI cloud pressures derived from rotational Raman scattering by comparisons with other satellite data and radiative transfer simulations, J. Geophys. Res., 113, D15S19, doi:10.1029/2007JD008689, 2008.

Ziemke, J. R., Joiner, J., Chandra, S., Bhartia, P. K., Vasilkov, A., Haffner, D. P., Yang, K., Schoeberl, M. R., Froidevaux, L., and Levelt, P. F.: Ozone mixing ratios inside tropical deep convective clouds from OMI satellite measurements, Atmos. Chem. Phys., 9, 573-583, doi:10.5194/acp-9-573-2009, 2009.

Comparison of CO from different satellite observations with model simulations: Klonecki, A., Pommier, M., Clerbaux, C., Ancellet, G., Cammas, J.-P., Coheur, P.-F., Co-

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zic, A., Diskin, G. S., Hadji-Lazaro, J., Hauglustaine, D. A., Hurtmans, D., Khattatov, B., Lamarque, J.-F., Law, K. S., Nedelec, P., Paris, J.-D., Podolske, J. R., Prunet, P., Schlager, H., Szopa, S., and Turquety, S.: Assimilation of IASI satellite CO fields into a global chemistry transport model for validation against aircraft measurements, Atmos. Chem. Phys., 12, 4493-4512, doi:10.5194/acp-12-4493-2012, 2012.

Worden, J., K. Wecht, C. Frankenberg, M. Alvarado, K. Bowman, E. Kort, S. Kulawik, M. Lee, V. Payne, and H. Worden, CH4 and CO distributions over tropical fires during October 2006 as observed by the Aura TES satellite instrument and modeled by GEOS-Chem, Atmospheric Chemistry and Physics, 13, 3679–3692, 2013, doi:10.5194/acp-13-3679-2013, 2013.

Pechony, Olga, Drew T. Shindell, and Greg Faluvegi, Direct top-down estimates of biomass burning CO emissions using TES and MOPITT versus bottom-up GFED inventory, Journal Of Geophysical Research, 118, 1–13,, doi:10.1002/jgrd.50624, 2013, 2013.

Liu, Junhua, J. A. Logan, D. B. A. Jones, N. J. Livesey, I. Megretskaia, C. Carouge, and P. Nedelec, Analysis of CO in the tropical troposphere using Aura satellite data and the GEOS-Chem model: insights into transport characteristics of the GEOS meteorological products, Atmos. Chem. Phys., 10, doi:10.5194/acp-10-12207-2010, 2010.

Detailed response to the reviewer comments:

In my opinion there are a number of major issues with this manuscript in its current form. The majority of those were also raised by one of the other reviewers (Joiner). Nonetheless I feel the need to repeat those as most of my concerns are not taken away by the response of the authors as published on 5 June 2013.

Main concerns : 1. I do not understand why the authors do not stick to the original SCHIAMACHY product they derive which are subcolumns in time. These represent physical quantities that people can easily understand, which unfortunately does not

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hold for the constructed PVCDs. These seasonally and spatially averaged PVCDs are very hard to interprete as the authors themselves also note when writing for example in the abstract 'the profiles retrieved from the CST have to be interpreted with care'. So why is this then done?

Author Reply: As mentioned in the paper, large numbers of measurements have to be averaged to achieve a consistent data set of SCIMACHY CO profiles. Thus it is not possible to extract meaningful time series of CO PVCDs (with low uncertainties) with a high time resolution. We found that averaging measurements over three months and three years is a good compromise. Reducing these averaging periods (e.g. using seasonal averages for individual years) leads to many data gaps for high cloud heights. Nevertheless, for a validation studies based on several ground based stations (new section 2.2), we used time series of seasonal averages for individual years, because for the lowest clouds there are enough observations to achieve sufficiently low uncertainties for a meaningful comparison with the ground based data. Future studies based on other satellite instruments with better signal to noise ratio and better temporal coverage can probably apply the CST with much better time resolution.

Moreover, when showing PVCDs in Fig. 4 the use of PVCDs actually hides the problems related to this approach as becomes clear in the reply to one of the reviewers (Joiner) under point 4a and which was also noted in the manuscript (p.11665). With this method one can get negative mixing ratios under certain conditions, which obviously is physically impossible. By using the PVCD representation this is not directly obvious and the authors find this 'aesthetically' more preferred (as they write in their response 5 June 2013). From a scientific point of view I have to disagree. As a scientist I want to know where a chosen approach fails. More concerning is the fact that this product may thus give wrong/inaccurate results which may not be so obviously wrong such that it produces unphysical results, but which are nonetheless inaccurate. There is however no way of telling when and where this is the case. I do not know how to get out of this dilemma if one is not able to validate the product.

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Author Reply: The use of the terms 'aesthetically more preferred' and 'unphysical' were no good choices. We don't use the latter term in the revised version of our manuscript. Nevertheless, it seems that were was a fundamental misunderstanding. Both representations of CO profiles (mixing ratios or PVCDs) could in principle be used and are both 'physical'. We chose the representation as CO PVCDs because CO PVCDs are not negative, and they also provide direct information about the absolute CO PVCDs. We changed the text in section 2, which should now be clearer to the reader.

2. Validation of a satellite product is essential, so one knows what the product is worth. The response of the authors to this comment by Joiner is 'However, since the retrieved CO profiles do not constitute 'real' atmospheric profiles, but complex composites of CO measurements made under different meteorological conditions, we have some doubts that MOZAIC data are really appropriate for such comparison'. This is another good reason to stick to your original subcolumn product. This is a physical quantity that does constitute a real atmospheric subcolumn at a certain moment in time and as such can be validated by MOZAIC (or other data such as aircraft data from NOAA).

Author Reply: As mentioned above, increasing the time resolution (e.g. using seasonal averages for individual years) leads to many data gaps, especially for high cloud heights. Thus a meaningful validation can only be applied to averages over several months (see new section 2.2).

3. A back-of-the-envelope calculation shows that under all circumstances over land and with low cloud fraction there will be a VERY significant contribution coming from the non-cloudy part of the observation. In fact for cloud fractions <10% I expect in the majority of the cases that most of the signal is from the non-cloudy part. Take cloud fraction 10%. The lowest surface albedo over land is _5% in this wavelength range. Already in this case the contribution to the total signal of the non-cloudy part is as good as equal (if not larger) to the contribution of the cloudy part. Cloud reflectivity in this wavelength range does not get higher than _50%, and is more often quite lower. Sur-

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face albedos over land vary between _5% and _60% in this spectral range, so in most cases over land –assuming 10% cloud fraction- the contribution from the noncloudy part is in fact the largest contribution to the observed signal and I don't see how the approach taken in this paper would then work. Therefore, I expect you need to take cloud fractions of say >70% for this approach to work well everywhere over land. The reason why the results in Fig. 2 might suggest otherwise could be due to the fact that this is a mixture of cases over land and over ocean. Over the oceans the approach works for any cloud fraction [ref. Gouldemans, 2009] as the ocean surface reflectivity is very low (Âń1%) (apart from sunglint situations). The authors responded -in reply to a similar comment by Joiner- they will calculate the relative fractions of received signal from the clear and clouded part of the observations and will effectively re-do their analyses based on that approach. Also, they will provide cloud radiance fractions for the selected threshold values of effective cloud fractions for a set of representative surface types. I think this is very important.

Author Reply: We are very thankful that both reviewers and Joanna Joiner strongly suggested to take into account also the contribution from the clear part of the ground pixel. As described in the general point 1) above, we completely re-calculated our data sets using the concept of cloud radiance fractions.

4. Why is FRESCO used to obtain information on cloud top height, which implies the use of the oxygen band at a very different wavelength, and more particularly why is not methane used in the same spectral range where CO is measured ? (see also point 2 raised by Joiner.) The fact that in both spectral ranges the photons penetrate the cloud to some degree –as described in section 2.1- is not a very strong argument as the point is that the effective penetration will be different in the different spectral ranges.

Author Reply: FRESCO+ was used because this data set is a well characterised and validated data set. We investigated the effect of the different penetration depths of photons at 2330nm compared to 760nm. For that purpose we extracted CTH from CH4 and compared them to FRESCO CTH as described in the general point 3 above.

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Excellent agreement was found, and we conclude that the differences in penetration depths are negligible for our study. This information is added to section 2.1.

5. Given the considerations above (in particular those under point 3) I do not know what the conclusions in section 3.4 w.r.t. model performances are worth. This needs to be re-assessed after the analyses is re-done as replied by the authors to the comments by Joiner (point 3 above).

Author Reply: As described above, the striking differences in the spatial patterns largely disappeared in the revised data set. In the new version of our paper we thus focus on the quantitative comparison between the models and SCIAMACHY. These differences are discussed in sections 3.4, 3.5 and the conclusions.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 11659, 2013.