**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 #2

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:

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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 SCIA-MACHY 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 ± 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

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-

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:

This manuscript presents an application of the cloud slicing technique to SCIAMACHY CO retrievals. This method generates valuable vertical information on the distribution of CO which is then used to test calculations of two transport models. The manuscript is overall well presented and it should be of large interest to the readers of ACP and I recommend publication of the manuscript after addressing my comments below. This manuscript deals with a new method for the use of SCIAMACHY CO columns and I believe that it is necessary to provide some information on the quality, uncertainties and characterization of the inferred CO sub-columns. As briefly mentioned in the

manuscript, the method involves multiple uncertain steps (e.g. photon path in clouds, effect of non-cloudy fraction, assignment of cloud heights, effects of interfering gases) and it appears necessary to attempt some level of quantification of the expected uncertainties. Furthermore, as for any remote sensing dataset, it is difficult to judge the value and quality of the datasets without any validation (although this might turn out to be very difficult). As a consequence, I believe that several conclusions drawn on the performance of models are somewhat pre-mature. You would first need to establish the quality and uncertainty of the satellite dataset before you can argue with confidence that observed model-measurement differences are the result of model shortcoming.

Author Reply: We agree with the reviewer and added more information on the uncertainties of the retrieved CO PVCDs (see general point 3 above). We also added some basic validation (see general point 4 above). The uncertainties caused by the contribution from the clear part are largely reduced by using the concept of cloud radiance fractions (see general point 1 above). The uncertainties caused by different penetration depths in both spectral ranges were quantified and found to be negligible for our study (see general point 2 above). We quantified the uncertainties of the CO PVCDs in detail in the new section 2.2. of the revised version of our manuscript.

Minor comments: p. 11661 IR -> infrared (IR)

Corrected

p. 11662 Our retrieval of the total atmospheric CO vertical 5 column density (VCD) and its validation is described in detail in Liu et al. (2011). -> It would be beneficial for the reader when the manuscript would include a brief summary of the clear-sky CO retrieval and its validation

Author Reply: We added this information at the beginning of section 2.

p. 11663 Therefore, the photons that the satellite detects are either scattered by the cloud or reflected at the Earth's surface ->Or scattered by aerosols

corrected

p. 11663: However, different from the study of Liu et al. (2011), here we use only observations for (partly) cloudy conditions (effective cloud fractions > 10%) -> The CO columns as described in Liu et al., 2011 are corrected for effects of clouds. For the cloud slicing method, I assume that you need to turn off such a correction. Is this what you do?

Author Reply: This is correct. We added this information at the beginning of section 2.

p. 11663 : : :the signal from the clouded part usually still dominates the measured spectra, which thus mainly contains information from the atmospheric above the cloud. -> I am not convinced that this is necessarily true for a cloud fraction of 10% only. If we assume a cloud albedo of 1 (probably much too high for 2.3 micron) and a sur- face albedo of 5% then the weight of the clouded part if 0.1x1 = 0.1 compared to 0.05x0.9=0.045. So the non-clouded part can easily contribute 50% to the total radi- ance.

Author Reply: We agree and we applied the concept of cloud radiance fractions in the revised version (see general point 1 above).

p. 11664 In contrast to the systematic dependence of the CO PVCD on cloud height, the CO PVCDs are almost independent of the selected effective cloud fraction thresh- old (see Fig. 2). -> There is a somewhat larger difference between 20 and 40 N for high clouds between 10%CF and 40%CF. Could this be caused by ice clouds that have a relatively low cloud albedo (due to strong ice absorption at 2.3 micron) so that a 10%CF criteria is too low?

Author Reply: In the revised version of the paper we use different thresholds (CRF instead of effective cloud fractions) and we also consider the contribution from the clear part of the ground pixel. In the revised version of Fig. 2 (new Fig. 4) the mentioned differences are not obvious anymore.

p. 11667: In other words: The retrieved (too low) cloud top height fits well to the retrieved (too high) CO PVCD. -> Do you have some indication that the magnate of both effects is similar otherwise there will be no significant compensation.

Author Reply: We compared Cloud heights retrieved from the CH4 absorption at 2330nm with FRESCO cloud heights (see general point 2 above) and found very good agreement. Thus we conclude that both effects largely cancel each other. We added this information to the text.

p. 11667 In this section, we compare CO profiles from SCIAMACHY observations with the results of two atmospheric models. -> Do you also consider averaging kernels in this comparison?

Author Reply: We do not consider averaging kernels for the clear part of the satellite pixel, because the shielding effect of clouds dominates the vertical dependence of the measurement sensitivity. This information is added to section 3.

p. 11670 : : :were also reported in other studies (e.g. Gloudemans et al., 2009, De Laat et al., 2010) -> this is only including SCIAMACHY and MOPITT. What about TES or IASI? -> How do these models compare to aircraft profiles ?

Author Reply: Similar results are also found in comparison studies using TES, IASI and AIRS. We added this information to the text and referred also the following references (see also general point 5 above). Shindell et al., 2006. Klonecki et al., 2012. Worden et al., 2013. Pechony et al., 2013. Junhua et al., 2010. Kopacz et al., 2010.

The following information about comparisons of the EMAC model with aircraft mea- surements were added to section 3.2:

CO results from the MATCH-MPIC model have been extensively compared to ground based and aircraft measurements an good agreement was found (von Kuhlmann et al., 2003b).

p. 11672: In general, very good agreement between SCIAMACHY observations and

model results is found (except for the systematic underestimation of the measurements by the models discussed above), with some distinct differences discussed below. -> I do not believe that you can argue that there is a very good agreement. As you rightly point out in brackets, the values are very different. So do you mean that there is a very good agreement in profile shapes or in the latitudinal and longitudinal distributions??

Author Reply: The sentence is changed to: In general, very good agreement of the spatial patterns, while the absolute values are systematically smaller in the models.

p. 11673: These spatial patterns are not found in the model data, which might be related to the vertical distribution of biomass burning emissions in the model, partly related to mixing processes between the boundary layer and the free troposphere -> The heat generated by fires will cause pyroconvection which is not well captured by models while most models simply assume that biomass burning emissions are injected only in the boundary layer. Which schemes are adopted by the models here?

Author Reply: The convection schemes are described in sections 3.1 and 3.2. However, as stated above, due to the new processing of the model data using CRF, the strong differences in spatial patterns largely disappeared.

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