Atmos. Chem. Phys. Discuss., 13, C8846–C8857, 2013
www.atmos-chem-phys-discuss.net/13/C8846/2013/


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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Received and published: 6 November 2013

Second Reply to Joanna Joiner

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

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:

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



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

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



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 comments from Joanna Joiner:

This paper applies the cloud slicing method to CO observations from SCIAMACHY. Cloud slicing has been used previously to derive information about tropospheric ozone. While the use of the cloud slicing approach for CO observations is new and very interesting, the paper leaves open several fundamental questions that are critical to the analysis of cloudy data at CO-affected wavelengths.

Major points: 1. There are significant questions related to the interpretation and use of the effective cloud fraction derived at wavelengths near the oxygen A-band and ap-



plied at CO wavelengths. The authors reference two papers on the FRESCO algorithm. Please see also Stammes et al. (JGR, 2008). Within the context of the mixed Lambertian cloud model used in many trace-gas retrieval algorithms, the effective cloud fraction, f_c, is the fraction of a hypothetical Lambertian cloud with 80% reflectivity that produces the radiance that is observed. Studies have shown that this model also well reproduces the amount of Rayleigh scattering and absorption that is observed. The clear sky fraction (in this model context) is then (1- f_c). However, to mix clear and cloudy air-mass factors or atmospheric column values, one should use the so-called cloud radiance fraction (see e.g., Veefkind et al., IEEE Trans. Geosci. Rem. Sens., 2006, Joiner et al., ACP, 2009) that is typically higher than the effective cloud fraction. This is alluded to on p. 11663 where it is stated that “the signal from the clouded part usually still dominates the measured spectra, which thus mainly contains information from the atmospheric above the cloud” (note the typo there). It appears that the authors are treating the PVCDs (not well defined in the paper, but understood to be the “observed” slant column normalized to a vertical column in the presence of clouds, including partial and thin clouds) as if this were the PVCD above the cloud pressure. This is equivalent to the assumption of a cloud radiance fraction of unity. While cloud radiance fraction over the dark ocean of near unity may be a good assumption, it is not clear that it is the case over land. The cloud radiance fraction needs to be computed, and the implications of the assumption of cloud radiance fraction equal to unity should be evaluated. Since land surface reflectances around 2.3 microns are not near zero, the cloud radiance fraction will not be unity over land. The authors are using data with effective cloud fractions down to 10%, so this needs to be carefully examined. The differences in PVCDs between effective cloud fraction cutoffs of 10 and 40%, shown in Fig. 2, appear to be quite significant. This is why Ziemke et al. in several papers apply the cloud slicing approach with only very high effective/radiative cloud fractions (see e.g., Ziemke et al., 2009). If the goal is to use more data to produce global coverage, a different approach may be needed. For example. Joiner et al., (ACP, 2010) attempt to account for cloud shielding effects in partly cloudy conditions in the analysis

of ozone column measurements. They show that if one assumes a well-mixed troposphere, the concept of an effective scene pressure can be used (i.e., the surface and cloud pressure are weighted appropriately by the cloud radiance fraction at the appropriate wavelength). They applied this approach to ozone data and use ozone sondes for validation, showing excellent results overall for tropospheric ozone.

Author Reply: We are very thankful for this comment! In contrast to our original expectation, it turned out that Joanna Joiner was absolutely right with her concerns. In principle there are at least two ways to deal with the problem of the influence of the clear part of the satellite pixel: a) One might use only SCIAMACHY observations with cloud radiance fractions (CRF) close to unity. Then, the comparison with the model results could have been done as in the original version of our paper. However, especially over land, only a few observations fulfil this requirement leading to poor statistics. Thus this option was not chosen. b) One could continue using SCIAMACHY observations also for rather small effective cloud fractions (in the revised version we use SCIAMACHY observations with CRF >30%). In contrast to the original version of our manuscript, we now extract the model data using the CRF concept as described in the general point 1 above. Using this approach, CO PVCDs extracted from the models are substantially higher, especially for high altitudes. However, the substantial discrepancies between SCIAMACHY observations and model results found in the original version of our paper, especially over biomass burning regions, largely disappeared.

It should be noted that we chose procedure (b) as described above, which is slightly different from the procedure of Joiner et al., 2009. Joiner et al., 2009 first calculate an effective cloud centroid pressure. Then from the derived tropospheric O3 column density an average mixing ratio for the tropospheric column above the effective cloud centroid pressure is derived. Instead, we first extract the CO columns for the clear and cloudy parts, respectively, from the models. Then we average these column densities according to the CRF an (1-CRF). We chose this procedure, because – in contrast to Joiner et al., 2009 – in our case we have explicit information about the CO profile from



the models. The CO PVCD for the effective cloud centroid pressure would not be the same as the average of both CO columns from the clear and cloudy part.

We updated the sections on the profile retrieval and results (also abstract and conclusions) accordingly. We refer to both studies (Veefkind et al., 2006, Joiner et al., 2009), and added the following additional references to the paper:

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.

We also corrected the typo – many thanks!

2. It is not clear that the mixed Lambertian-equivalent reflectivity (MLER) cloud model can be applied at the wavelengths relevant to CO retrievals. The assumptions in this

model begin to break down in the near-IR where cloud absorption becomes an issue. There is a statement on p. 11663 that "within the spectral range of the CO analysis clouds are not as bright as in the visible spectral range", but the implications of this are not investigated. It is not clear that the cloud pressure (or the cloud radiative fraction) derived from the O2 A-band is applicable at CO wavelengths, because photon path lengths (and cloud single scattering albedo) are different in the CO vs O2-A band wavelengths, see e.g., Platnick (JQSRT, 2001). In order to convince a reader that the cloud slicing approach with FRESCO is applicable at CO wavelengths, simulations over a wide range of conditions should be conducted. Note also that we refer to the cloud pressures derived in the VIS and UV as "cloud optical centroid pressure" – see also Vasilkov et al. (JGR, 2008), Ziemke et al. (JGR, 2009), Joiner et al. (AMT, 2010 and AMT, 2012).

Author Reply: We added information on the uncertainty of the retrieved CO PVCDs caused by errors of the CRF in the new section 2.2. We also 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. Excellent agreement was found, and we conclude that the differences in penetration depths are negligible for our study. In particular, we conclude that deviations of the MLER cloud heights from the true cloud top heights are similar in both spectral ranges and should thus mainly cancel each other. This information is added to section 2.1.

We explicitly mention the findings of the Vasilkov et al. 2008 paper in section 2.1.. We also refer to the Platnick, 2001 to illustrate differences in penetration depth of backscattered light for different wavelengths (see also general point 5 above).

3. Validation is a necessary exercise that should be conducted as well as possible in order to establish whether the approach is working properly. There is a wealth of CO profile information available from MOZAIC aircraft measurements at landing and takeoff sites around the world. In ozone work, many comparisons to ozone sonde data have



been made and extrapolated stratospheric column data have also been compared with independent measurements (see e.g., Ziemke et al. ACP, 2009). Similar extrapolation of the results shown here may lead to unrealistic estimates of stratospheric column CO and that would leave the reader to question the validity of the results. Validation should be conducted before model comparisons in order to establish the relevancy of such comparisons.

Author Reply: In contrast to other trace gases (like O3) validation of SCIAMACHY CO profiles is a very complicated task as described in the general point 3 above. Nevertheless, we added some basic validation (new section 2.2) by comparing time series of the CO PVCDs (seasonal averages from 2003 to 2005) to total CO VCDs at several TCCON stations, for which good agreement was found.

4. Justifications as to why further analysis is done using PVCDs instead of derived concentrations (or mixing ratio) do not make sense to me. If the results in terms of concentrations are unphysical, then the results in terms of the PVCDs are also unphysical, they are just less obviously so. For example, if the above-cloud VCD is constant with altitude as it nearly is near the surface in several of the plots shown, that would imply a mixing ratio in those altitudes near zero. This would appear to be unrealistic, particularly over China near the surface, as shown in Fig. 4. In summary, the results shown are physically unrealistic in many respects. Possible explanations as to why the approach may not be working as intended warrant further investigation.

Author Reply: The term 'unphysical' was no good choice and we removed it from the revised version. Both representations for the CO profiles (mixing ratios and PVCDs) could in principle be used. 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.

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