

author comments on the manuscript amt-2017-423, referee 1

We would like to thank the referee for the comments to further improve our manuscript. In this document we provide our reply to the comments. The original comments made by the referee are numbered and typeset in italic and bold face font.

1. *Generally, the paper discusses an important subject as we lose high percent of data due to cloud screening. However, the paper needs to be more clear as it is ambiguous at many places and also it might be good if the English is edited by professionals.*

We assume to satisfy the request of the reviewer with our changes to the manuscript.

2. *The following are examples of the parts that need to be clarified: 1-The authors need to explain why they chose the following oversampling size? The SCIAMACHY data are averaged over an area with a radius $r = 90$ km and subsequently oversampled with a longitude/latitude sampling distance of $= 0.5$ degree*

adjusted

To make it more clear we have added the following paragraph at p5,l10:

“ To find an appropriate averaging radius r , a trade-off has to be made between spatial resolution and noise of the averaged CO field. Obviously, this choice depends on the particular application due the number of available CO data points and the brightness of the observed scene. The choice of the sampling distance δ is less critical as far as it is $< r$ to achieve an oversampling of the data field. Therefore, r and δ changes for the applications discussed in the following and are provided accordingly in the discussion. ”

Hence, in Sec. 3 we chose $r = 90$ km, and “ $\delta = 0.5$ ” to analyse pollution by wild fires and in Sec. 4 $r = 40$ km, and “ $\delta = 0.05$ ” to analyse pollution by cities.

3. *2- The following parts need to be rephrased to be more clear Page 2, line 13 Please rephrase to be more clear “Obviously, this assumption does not hold for optically thick water clouds shielding the atmosphere below. Therefore, Buchwitz et al. (2007); Gloudemans et al. (2009); de Laat et al. (2012) used the retrieved CH₄ total column and model vertical profiles of CH₄ and CO to compensate for the shielding effect by clouds on the estimated total column of CO”*

adjusted

We have changed the sentence at p2,l13:

to

“A different approach is followed by Buchwitz et al. (2007); Gloudemans et al. (2009); de Laat et al. (2012) who use vertical profiles of CH₄ and CO taken from model simulations to compensate for the reduced sensitivity when retrieving trace gas columns from cloud contaminated measurements. ”

4. *Page 2, line, 29 Please rephrase to be more clear “This aspect is particularly interesting in the light of upcoming future missions with improved radiometric and spatial sampling performance”*

adjusted

the sentence at p2,l29 is rephrased

to

“ On the other hand, cloudy and clear sky observations with different vertical CO sensitivities provide information on the vertical distribution of CO (Liu et al., 2014). Here, it is necessary to observe similar CO vertical distributions with different cloudiness, which will be met more frequently by upcoming CO missions with enhanced spatial sampling and resolution in combination with improved data quality of the individual CO soundings. ”

5. *Page 2, line 30 I think the following reference needs to be updated because it is 2012, and TROOMI launched in 2017. “October 13, 2017 the Tropospheric Monitoring Instrument (TROPOMI) was success- fully launched on the Sentinel-5 Precursor Mission (S-5P) (Veefkind et al., 2012). ”?*

adjusted The citation is still valid but we rephrase the sentence to make it clear. We changed the sentence at p2,l30 to

“ October 13th, 2017 the Tropospheric Monitoring Instrument (TROPOMI) was successfully launched on the Sentinel-5 Precursor Mission (S-5P). The mission objectives and requirements are provided by Veefkind et al., 2012. ”

6. *Page 3, line 8 Please rephrase to be more clear light path enhancement*

adjusted

The sentence at p3,l8 “The TROPOMI CO data product comprises the estimate of the CO column, its noise estimate, effective cloud parameters and the CO column averaging kernel, which reflects the effect of cloud shielding and light path enhancement by clouds on the retrieved CO column. ”

is rephrased to

“The TROPOMI CO data product comprises the estimate of the CO column, its noise estimate, effective cloud parameters and the CO column averaging kernel, which provides the sensitivity of the retrieved column with respect to changes of the true vertical CO profile. For example for cloudy atmospheres, the averaging kernel reflects the shielding of the atmosphere below the cloud with a reduced CO sensitivity, equivalent to small values of the averaging kernel. ”

7. *Page 9, line 29 Please rephrase to be more clear The following have repeated statements, please delete one of them. “a higher spatial resolution of up to 77 km² , and improved spatial sampling”*

adjusted

The sentence at p9,l29 “ The 2.3 μm spectral range of TROPOMI is covered by SCIAMACHY with the same spectral resolution and spectral coverage but TROPOMI is characterized by a significantly improved SNR of the measurements, a higher spatial resolution of up to $7 \times 7 \text{ km}^2$, and improved spatial sampling with daily global coverage. ”

is changed to “ The 2.3 μm spectral range is covered both by TROPOMI and SCIAMACHY with the same spectral resolution, whereby TROPOMI shows an improved radiometric performance with a high spatial resolution of up to $7 \times 7 \text{ km}^2$ and with daily global coverage. ”

8. *Page 8, line, 28 Please rephrase to be more clear For Tehran and Los Angeles the temporal and spatial sampling of low cloud observations improved the spatial coincidence of the CO enhancement and the MODIS urban area product compared to the clear sky CO product.*

adjusted

We have changed the sentence at p8,l28 to

“Compared to clear sky observations, the temporal and spatial sampling of low cloud observations improves the spatial match of the CO enhancements with the corresponding MODIS urban areas of Tehran and Los Angeles. ”

9. *Page 9, line, 3 Please rephrase to be more clear “The analysis of the CO pollution from megacities from SCIAMACHY soundings with medium-high clouds indicated a significant reduction in the CO enhancement for the three cities”*

adjusted

We have rephrased the sentence at p9,l3 to “ However, when using medium-high clouds for the detection of CO pollution we recognized a significant reduction in the CO enhancement above the three cities. ”

10. *Page 9, line 14 open op new, replace op with up*

adjusted

11. *The following figures have very small font so please increase their size: Figures 2,4,6,7,8*

adjusted

Detection of carbon monoxide pollution from ~~megacities~~-cities and wildfires on regional and urban scale: The benefit of CO column retrievals from SCIAMACHY 2.3 μm measurements under cloudy conditions

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Abstract. In the perspective of the upcoming TROPOMI Sentinel-5 Precursor carbon monoxide data product, we discuss the benefit of CO total column retrievals from cloud contaminated SCIAMACHY 2.3 μm shortwave infrared spectra to detect atmospheric CO enhancements on regional and urban scales due to emissions from ~~megacities~~-cities and wildfires. The study uses the operational Sentinel-5 Precursor algorithm SICOR, which infers the vertically integrated CO column together with effective cloud parameters. We investigate the capability to detect localized CO enhancements distinguishing between clear-sky observations and observations with low (<1.5 km) and medium-high clouds ~~-. Exemplary(1.5km-5km).~~As an example, we analyze CO enhancements over the ~~megacities~~-cities Paris, Los Angeles, and Tehran as well as the wildfire events in Mexico/Guatemala 2005 and Alaska/Canada 2004. The CO average of the SCIAMACHY full mission data set of clear-sky observations can detect weak CO enhancements of less than 10 ppb due to air pollution in these cities. For low cloud conditions, the CO data product performs similarly well. For medium-high clouds, the observations show a reduced CO signal both over Tehran and Los Angeles, while for Paris no significant CO enhancement can be detected. This indicates that information about the vertical distribution of CO can be obtained from the SCIAMACHY measurements. Moreover, for the Mexico/Guatemala fires, the low-cloud CO data captures a strong outflow of CO over the Gulf of Mexico and the Pacific Ocean and so provides complementary information to clear-sky retrievals, which can only be obtained over land. For both burning events, enhanced CO values are even detectable with medium-high cloud retrievals, confirming a distinct vertical extension of the pollution. The larger number of additional measurements and hence the better spatial coverage, improves significantly the detection of wild fire pollution using both the clear-sky and cloudy CO retrievals. Due to the improved instrument performance of the TROPOMI instrument with respect to its precursor SCIAMACHY, the upcoming Sentinel-5 Precursor CO data product will allow ~~to detect CO emission and its vertical extension of many more cities and wildfires and so opens new research opportunities~~improved detection of CO emissions and their vertical extension over cities and fires, making possible new research applications.

1 Introduction

Carbon Monoxide (CO) is an atmospheric trace gas emitted mainly by incomplete combustion processes. Its oxidation with the hydroxyl radical (OH) represents its major sink (Spivakovsky et al., 2000). ~~With~~ Because of its moderate long lifetime ~~of several months~~ and its low background concentration (Holloway et al., 2000), it is an important tracer for atmospheric transport of pollution (Logan et al., 1981). From space, CO is measured by different satellite instruments with global coverage, e.g. MOPITT (Measurements of Pollution in the Troposphere; Deeter (2003)), AIRS (Atmospheric Infrared Sounder; McMillan (2005)), TES (Tropospheric Emission Spectrometer; Rinsland et al. (2006)), IASI (Infrared Atmospheric Sounding Interferometer; Turquety et al. (2004)), and SCIAMACHY (Scanning Imaging Absorption Spectrometer for Atmospheric Chartography; ~~Gloude~~ Gloude ~~mans et al. (2009); Frankenberg et al. (2005); Buchwitz et al. (2007); Gimeno Garcia et al. (2014)~~ Gloude mans et al. (2009), Frank and Gimeno Garcia et al. (2011)).

~~For the interpretation of satellite observations, the~~ The presence of clouds ~~in the observed scene represents a major challenge~~ may represent a challenge for remote sensing of CO from space. Here, light scattering and hence the shielding of the atmosphere below the cloud affects the vertical sensitivity of the measurement. This hampers the retrieval of the vertically integrated total column of CO from cloudy observations and different approaches have been proposed to cope with this problem. ~~Deeter (2003); Buchwitz et al. (2004); de Laat et al. (2006); Borsdorff et al. (2016)~~ Deeter (2003), Buchwitz et al. (2004), de Laat et al. (2006) and Borsdorff et al. (2016) suggest to consider only observations under ~~clear sky~~ clear-sky conditions or weakly cloud contaminated, assuming that the sensitivity to CO in the lower troposphere is sufficient to estimate the total CO column. ~~Obviously, this assumption does not hold for optically thick water clouds shielding the atmosphere below. Therefore, Buchwitz et al. (2007); Gloude~~ the retrieved CH_4 total column and model A different approach is followed by Buchwitz et al. (2007), Gloude mans et al. (2009), and de Laat et al. (2012) who used vertical profiles of CH_4 and CO taken from model simulations to compensate for the ~~shielding effect by clouds on the estimated total column of CO~~ reduced sensitivity when retrieving trace gas columns from cloud contaminated measurements. Alternatively, ~~Rinsland et al. (2006); Borsdorff et al. (2017); Vidot et al. (2012)~~ Rinsland et al. (2006), Borsdorff et al. (2017), and Vidot et al. (2012) discussed the retrieval of the CO column jointly with effective cloud parameters resulting in a retrieved CO column with its vertical sensitivity, where the latter reflects the effect of clouds on the light path and so includes the shielding effect of clouds. This approach is not limited to particular conditions of cloud coverage and so generalizes the above-mentioned techniques providing a higher data yield.

The usefulness of CO total column from satellite observations have been demonstrated by several studies. For example, after temporal averaging of several years of IASI and SCIAMACHY CO measurements, (Pommier et al., 2013; Buchwitz et al., 2007; Clerbaux et al., 2008) detected the relatively weak CO enhancement of urban pollution in ~~megacities~~ cities. Also, the pronounced enhancement of CO due to wildfires have been reported (e.g., Gloude mans et al., 2006; Buchwitz et al., 2007). Depending on the study, only clear-sky observations or both clear-sky and cloudy observations are used. Due to the different vertical sensitivity of the observations, the use of the data have to be considered with care. For moderately high clouds, observations might not be suited to detect enhanced CO concentrations in the lower atmosphere because of the shielding of the atmosphere below the cloud. On the other hand, ~~using cloudy observations jointly with clear sky may provide new~~

applications to constrain the vertical extension of the pollution (Liu et al., 2014). This aspect is particularly interesting in the light of upcoming future missions with improved radiometric and spatial sampling performance.

~~October 13~~ cloudy and clear-sky observations with different vertical CO sensitivities provide information on the vertical distribution of CO (Liu et al., 2014). Here, it is necessary to observe similar CO vertical distributions with different cloudiness, which will be met more frequently by upcoming CO missions with enhanced spatial sampling and resolution in combination with improved data quality of the individual CO soundings. ~~October 13th~~, 2017 the Tropospheric Monitoring Instrument (TROPOMI) was successfully launched on the Sentinel-5 Precursor Mission (S-5P) (Veefkind et al., 2012). The mission objectives and requirements are provided by (Veefkind et al., 2012).

It measures Earth reflected radiances in the ultraviolet, visible, near infrared and shortwave infrared spectral range with a spatial resolution of about $7 \times 7 \text{ km}^2$ at sub-satellite point and daily global coverage. Here, shortwave infrared (SWIR) observations in the $2.3 \text{ }\mu\text{m}$ spectral region provide information on the CO total column amount. In recent years, the ~~Shortwave Infrared-SWIR~~ CO Retrieval algorithm SICOR has been developed for the operational processing of TROPOMI data (Vidot et al., 2012; Landgraf et al., 2016b, a). TROPOMI's high signal-to-noise ratio (SNR) performance in the SWIR will provide clear-sky-clear-sky CO total column densities with vertical sensitivity throughout the atmosphere (e.g., Buchwitz et al., 2004; Gloudemans et al., 2008; Borsdorff et al., 2014) and with a precision $< 10 \%$ for single clear-sky-clear-sky soundings (Landgraf et al., 2016b). However even with the high spatial resolution and sampling of TROPOMI, a major part of the measurements is cloud contaminated (Krijger et al., 2005, 2011). To optimally explore the SWIR measurements, SICOR retrieves CO also for cloudy conditions over land and ocean inferring effective cloud parameters (cloud optical thickness τ_{cld} , cloud centre height z_{cld}) together with trace gas columns (Vidot et al., 2012; Landgraf et al., 2016b). The TROPOMI CO data product comprises the estimate of the CO column, its noise estimate, effective cloud parameters and the CO column averaging kernel, which reflects the effect of cloud shielding and light path enhancement by clouds on the retrieved CO column provides the sensitivity of the retrieved column with respect to changes of the true vertical CO profile. For example for cloudy atmospheres, the averaging kernel reflects the shielding of the atmosphere below the cloud with a reduced CO sensitivity, equivalent to small values of the averaging kernel. Furthermore, the effective cloud parameters provide useful information to e.g. classify measurements by the type of cloud contamination.

~~To evaluate the benefit and maturity of this approach for the detection of localized CO enhancements by TROPOMI we~~ We have applied the SICOR algorithm to the full SCIAMACHY mission data set of $2.3 \text{ }\mu\text{m}$ spectra dataset. Here, SCIAMACHY covers the TROPOMI SWIR band with the same spectral resolution but with inferior radiometric performance, spatial resolution and global coverage (Bovensmann et al., 1999). The SCIAMACHY CO dataset was validated with TCCON/NDACC measurements for clear-sky observations over land (Borsdorff et al., 2016) and with TCCON and MOSAIC/IAGOS airborne measurements (Borsdorff et al., 2017) for cloud contaminated measurements over land and oceans. In general, those studies found a good agreement with the validation datasets considering the high noise error of the SCIAMACHY CO dataset. For most sites the bias is $< 10 \text{ ppb}$ but can increase significantly at CO hot spots due to representation errors of the validation. In this study, we discuss the benefit of CO retrievals from cloud contaminated SCIAMACHY $2.3 \text{ }\mu\text{m}$ measurements with their intrinsic vertical sensitivity for the detection of CO pollution from megacities-cities and wildfires. ~~Here, we distinguish CO~~

~~retrievals from measurements~~ We separate CO retrievals under clear-sky, low (<1.5 km), and medium-high (1.5-5 km) cloud conditions. Exemplary As an example, we discuss ~~the CO pollution by~~ CO pollution over Tehran, Paris, and Los Angeles as well as the wildfires in Mexico/Guatemala 2005 (Herrera, 2016) and Alaska/Canada ~~2014-2004~~.

The paper is structured as follows: In section 2, we present the SCIAMACHY CO dataset. Section 3 analyzes the benefit of the SCIAMACHY CO retrievals under cloudy conditions to detect wildfires, and Section ~~??~~ 4 focuses on the CO emission from megacities. ~~Finally, Section 6 will give a summary and conclusions~~ cities. Section 5 draws some conclusions on the upcoming TROPOMI CO dataset. The summary and conclusion is given in section 6 and finally, section 7 states the availability of the data.

2 SCIAMACHY CO dataset ~~and retrievals~~

- 10 The SCIAMACHY instrument was operational on ESA's ENVISAT satellite from January 2003 until April 2012. We utilize the SWIR measurements of SCIAMACHY in nadir observation geometry with a spatial resolution of about 120×30 km², a swath of 960, and a global coverage within 3 days (Bovensmann et al., 1999). In this study, we analyze the SICOR CO total column densities, retrieved from individual SCIAMACHY $2.3 \mu\text{m}$ spectra for the entire period of the mission from January 2003 until April 2012. The CO data product ~~is available on the public ftp site~~. It consists of the estimates of the total
- 15 column concentrations of CO, H₂O and HDO (c_{CO} , $c_{\text{H}_2\text{O}}$, c_{HDO}), the corresponding retrieval noise (ϵ_{CO} , $\epsilon_{\text{H}_2\text{O}}$, ϵ_{HDO}), averaging kernels, effective cloud parameters (cloud optical thickness τ_{cld} and cloud height z_{cld}) and the SWIR Lambertian surface albedo. ~~Moreover, different auxiliary parameters are provided like the number of iterations of the inversion~~ Auxiliary parameters like signal-to-noise ratio SNR_{max} of the measurement and number of retrieval iterations (N_{iter}) ~~and the maximum signal to noise ratio of the measurement SNR_{max}~~
- 20 ~~are also provided~~. SICOR uses the profile scaling approach, discussed in detail by ~~Vidot et al. (2012); Borsdorff et al. (2014); Landgraf et al. (2014)~~, and Landgraf et al. (2016b). Here, ~~the effective cloud parameters cloud optical depth and height~~ are estimated using prior knowledge about ~~CH₄ using the~~ CH₄, ECMWF surface pressure ~~and the corresponding~~, and the observed CH₄ absorption in the $2.3 \mu\text{m}$ spectral fit window (Landgraf et al., 2016a). The CH₄ data was taken from a TM5 model run (Williams et al., 2013, 2014) spanning the entire mission period of SCIAMACHY with global 3×2 degree² horizontal
- 25 ~~resolution and 3 hourly sampling time~~. The algorithm can also be used for SCIAMACHY CO data processing because of the similarity of the TROPOMI and SCIAMACHY observations. The specific SCIAMACHY settings, e.g. the selection of the retrieval window, are discussed by Borsdorff et al. (2017). ~~The spectral range for the retrieval from 2311-2338 nm was chosen to compensate for the detector pixel loss in the later years of the mission but also to include a strong CH₄ absorption line, which are beneficial for the retrieval of the effective cloud parameters~~. Due to an ice layer on the SWIR detectors and the
- 30 radiometric degradation of the instrument, the processing of SCIAMACHY CO data requires a radiometric re-calibration of the SWIR spectra as described by Borsdorff et al. (2016).

In this study, we ~~apply a data screening of the individual SCIAMACHY CO retrievals~~ filter the SCIAMACHY CO data based on the number of iterations N_{iter} and the estimate of the retrieval noise, which we compared with σ the difference of the 50th and 68th percentile of the retrieval results for the different data ensembles. The data filter reads

1. $N_{\text{iter}} < 15$
2. $\epsilon_{CO} < 4.5 \sigma_{CO}$
3. $\epsilon_{H_2O} < 4.5 \sigma_{H_2O}$
4. $\epsilon_{HDO} < 4.5 \sigma_{HDO}$.

For the analysis of air pollution from ~~megacities~~ cities, we add an additional filter considering the median CO column μ_{CO} of the CO data set of the different cities,

5. $\mu_{CO} - 4.5 \sigma_{CO} \leq c_{CO} < \mu_{CO} + 4.5 \sigma_{CO}$.

This filter removes outliers of our data sets, which we attribute to erroneous retrievals possibly caused by the instrument degradation rather than an atmospheric signal in case of the selected ~~megacities~~ cities. It enables us to detect the relatively weak CO enhancement above cities after averaging the data over the entire mission period. ~~To distinguish the effect of clouds on the retrieval, we consider three different categories of cloudy observations as indicated~~ In this study we consider clear-sky and cloudy-sky retrievals with cloud heights smaller than 5 km latter distinguished in three categories specified in Tab. 1.

An important element of the CO data product is the column averaging kernel A , which provides the sensitivity of the retrieved CO column to changes in the true vertical profile ρ_{true} of CO (Rodgers, 2000), namely

$$c_{\text{ret}} = A\rho_{\text{true}} + \epsilon_{CO}, \quad (1)$$

where ϵ_{CO} represents the error of the retrieved CO column caused by measurement errors. Eq. (1) can be interpreted as a weighted altitude integration accounting for the vertical sensitivity of the retrieval to estimate the retrieved CO column density. Figure 1 shows the total column averaging kernels for four different cloud conditions over Paris. Here, scenes contaminated by optically thin low clouds provide a good vertical sensitivity of the total column of CO and so the values of A are close to 1 for all altitudes. However, for scenes with optically thick clouds, the retrieval loses CO sensitivity below the cloud with averaging kernel values well below 1. Because the CO column is estimated by a scaling of a reference profile, CO variations above a cloud also induce an adjustment of the CO concentration below the cloud, where the measurement is not sensitive to. This explains the column averaging kernel values > 1 at this altitude range (Borsdorff et al., 2016). This limited retrieval sensitivity to the atmospheric composition below a cloud induces the so-called null-space error to the retrieved total column (e.g., Borsdorff et al., 2014). For the profile scaling approach the magnitude of the null-space error depends on the one hand on the loss of vertical sensitivity and on the other hand on the discrepancy between the true vertical profile and the reference profile to be scaled by the inversion. Hence, depending on this discrepancy the retrieved column can over or underestimate the true vertical column.

For individual CO retrievals from SCIAMACHY observations, the retrieval noise ϵ_{CO} can be high and can even exceed 100 % of the retrieved column depending on the SNR of the measurement (Gloudemans et al., 2008). Hence, for most applications individual SCIAMACHY CO retrievals need to be averaged to reduce the noise (de Laat et al., 2007; Gloudemans et al., 2006). In this study, we use an oversampling technique similar to the one used by Fioletov et al. (2011). This means that we first

5 define an equidistant latitude/longitude grid with a sampling distance δ for a considered scene. For each grid cell, an averaged CO value is calculated using SCIAMACHY CO retrieval weighted with its noise error ϵ_{CO} within a circular domain of a radius r around the cell centre. Here $\delta < r$, which corresponds to an oversampling of the averaged SCIAMACHY CO field. To find an appropriate averaging radius r , a trade-off has to be made between spatial resolution and noise of the averaged CO field. Obviously, this choice depends on the particular application due the number of available CO data points and the brightness of

10 the observed scene. The choice of the sampling distance δ is less critical as far as it is $< r$ to achieve an oversampling of the data field. Therefore, r and δ changes for the applications discussed in the following and are provided accordingly in the discussion. In the following, we chose r such that a high spatial resolution is achieved but also the retrieval noise is sufficiently reduced by averaging. Hence, the choice of the parameter depends on the application, the number of individual retrievals available, and the reflectivity of the ground scene. δ is less critical however we chose it smaller than r to achieve an oversampling of the data.

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3 CO pollution from wildfires

After carefully evaluating the SCIAMACHY CO data set, we have selected two examples of wildfire events for further discussion: agricultural fires in Mexico/Guatemala 2005 and forest fires in Alaska/Canada 2004. Buchwitz et al. (2007) discussed the fires in Alaska/Canada 2004 with SCIAMACHY CO retrievals and Pfister et al. (2005) quantified their CO emissions using

20 MOPITT CO data. We will revisit those fires from the perspective of CO retrievals under cloudy conditions.

Figure 2 shows time series of individual SCIAMACHY CO retrievals over Mexico for clear-sky, low cloud, and medium-high cloud conditions as well as the daily GFED4 Burned Area product of MODIS (Randerson et al., 2017). Depending on the signal-to-noise ratio of the measurements the retrieval noise of individual CO retrieval can exceed 100 % of the retrieved column and by that can result in negative CO columns. It is important not to reject negative values when averaging data to

25 avoid artificial biases (de Laat et al., 2007; Gloudemans et al., 2006).

The two burning events indicated by the GFED4 Burned Area product in 2003 and 2005 are clearly reflected in the time series of low cloud and medium-high cloud retrievals but shifted by about 45 days for both events. ~~We ascribe this temporal shift to the atmospheric response time to built up the high atmospheric CO concentration. It is interesting to note that the retrieval shows both a slowly varying increase and decrease of the burning activities over the month and enhanced peak events~~

30 ~~, both also evident in the GFED4 Burned Area data.~~ The reason for the shift is unclear and will be studied in future also looking at other satellite observations. As expected, CO retrieval values increase during the fire season (March- May) each year, coinciding with an increase in burned area. Here, the peak events are evident in both the low cloud and medium-high cloud data records. The time series of the ~~clear-sky~~ clear-sky data is very noisy and has significant gaps because of the dark

ocean surface in the SWIR which does not permit a CO retrieval. Both hamper the detection of fire events. Nonetheless, it seems that the two fire events are also visible in the ~~clear-sky-clear-sky~~ data.

From the clear-sky, low-cloud, and medium-high cloud time series we calculated daily mean values and investigated the correlation of the data sets. For the correlation between the low-cloud and clear-sky data product, the Pearson coefficient is 0.6 with a mean bias of 1.7 ppb, and a standard deviation of the differences of 32.7 ppb. The large standard deviation reflects the noise of the clear-sky data. For the Mexico region, the land surface reflectivity and so the corresponding SNR of the measurement is low causing the high retrieval noise for clear-sky cases. However, the good correlation coefficient and the low bias shows that within the noise limitation the ~~clear-sky-cloudy~~ retrievals are in good agreement with the ~~cloudy-clear-sky~~ retrievals.

The situation differs when inspecting the CO time series for SCIAMACHY observations with low and medium-high cloud coverage. Here the data are much less noisy. In the SWIR, clouds are highly reflective, as demonstrated by Borsdorff et al. (2017) using SCIAMACHY SWIR observations, and so the improved SNR of the SCIAMACHY measurements causes a reduced noise in the CO data product. When correlating the low-cloud and the high-cloud retrievals, we find a Pearson correlation coefficient of 0.8, a bias of 4.6 ppb, and a standard deviation of the differences of 14 ppb. This supports our finding that both low-cloud and medium-high cloud retrievals can capture the burning events equally well, something one may expect since CO pollution from wild fires constitutes a strong source that can reach the free troposphere (see e.g. Yurganov et al. (2005)). In case the CO plume was confined to the near-surface atmosphere, it would be more difficult if not impossible to sense it with cloudy observations. When the CO plume confined near the surface it would be more difficult to sense with cloudy retrievals.

Figure 3 shows the spatial distribution of the SCIAMACHY CO total column over Mexico for the period 15th March - 15th May 2005 and the corresponding GFED4 Burned Area product. The SCIAMACHY data are averaged over an area with a radius $r = 90$ km and subsequently oversampled with a longitude/latitude sampling distance of $\delta = 0.5$ degree (≤ 55 km). We used the same latitude/longitude grid for the MODIS data where we sum up the burned areas for the individual grid cells. Here, the clear-sky SCIAMACHY CO data clearly show the burning hot spot around the state Yucatan Peninsula in Mexico, also indicated by the MODIS Burned Area product. Some fires shown by the MODIS data are not reflected by the SCIAMACHY CO data, which may be explained by the fact that the CO emission of these fires is not sufficient to become detectable with SCIAMACHY observations. For low cloud conditions, the retrieval provides additional information showing the transport of air with high CO concentration into the Gulf of Mexico and over the Pacific Ocean, in agreement with the smoke detection of the MODIS Aqua instrument (<https://earthobservatory.nasa.gov/NaturalHazards/view.php?id=14748>). Also, the earlier burning event in Mexico 2003 in Fig. 2 followed a similar transport pattern of enhanced CO over the oceans (~~not shown as shown in Fig. 4~~). The CO observations with medium-high cloud observations still reflect the CO enhancements but the measurement density is too low to fully capture the event. Analogously, Figure 5 shows the forest fires in Alaska/Canada from 1st July to 1st August 2004 using the same oversampled approach as in Figure 3. Because of different meteorology, clear-sky observations are less frequently for the Alaska fires than for fires in Mexico and hence ~~clear-sky-clear-sky~~ and low cloud observations do not fully capture the Alaska 2004 fire event. For medium-high clouds, the corresponding CO product shows much better coverage and so can detect ~~the wildfires in agreement with~~ enhanced CO concentration transported away from the fires as indicated by

the MODIS burned area product. In particular, this finding agrees with the study by Pfister et al. (2005) who reported enhanced CO concentration even high up in the atmosphere at about 400 hPa due to the Alaska fires.

The benefit of cloudy observations to detect the transport of enhanced CO concentration from wildfires becomes also clear when comparing the number of individual SCIAMACHY CO sounding in Fig. 6. For the considered area of the Mexico fires, the 2402 individual clear sky soundings are more than doubled (6126 soundings) when we consider low cloud observations, partly due to additional soundings over ocean which cannot be exploited for clear-sky conditions. Additionally 3225 soundings are found with medium-high clouds. For the Alaska fires, the relative distribution changes due to the different meteorological situation but confirms a significant gain in number of observations when including cloudy measurements. Here, we obtain 1473 ~~clear-sky~~ clear-sky soundings, 2454 low cloud and 4819 medium-high cloud soundings. Due to this, the means to observe enhanced CO values by pollution transport from wildfire with SCIAMACHY is clearly improved by using cloudy observations in addition to ~~clear-sky~~ clear-sky observations.

4 CO pollution from ~~megacities~~ cities

In this section, we selected the three ~~megacities~~ cities Paris, Tehran, and Los Angeles to discuss the relevance of cloudy observations for the detection of urban pollution. ~~Accumulating~~ We accumulated all SCIAMACHY observations from 2003 to April 2012 around these cities ~~and,~~ distinguishing between clear-sky, low-cloud and medium-high cloud retrievals, ~~we apply~~ . Then we applied the oversampling approach with a longitude/latitude grid of $\delta = 0.05$ degree (≤ 5.5 km) and an averaging radius $r = 40$ km ~~as shown in the~~ (Figs. 7, 8, and 9 ~~together with the MODIS urban area contours of Schneider et al. (2009).~~). Subsequently, we calculated the CO enhancement for the three cities with respect to the background signal estimating the difference of the median CO concentration inside and outside the urban area contours (Schneider et al., 2009). Obviously, the separation of urban and background CO concentrations cannot fully succeed due to the SCIAMACHY pixel size, the averaging approach and atmospheric transport, however the values presented in Fig. 11 give a first indication of CO enhanced due to urban population.

For all three cities, we find that the CO enhancements of clear-sky observations coincide with the MODIS urban area contours. Furthermore, in case of Paris we ~~can~~ detect enhanced CO levels ~~over~~ near the neighboring city Rouen caused by local emissions or transport from the remote pollution of Paris (see Fig. 7). The strongest CO enhancement under clear-sky condition occurs for Tehran with 8.1 ppb, closely followed by Los Angeles with 6.3, and the weakest enhancement we observe for Paris with 4.3 ppb. This difference can be explained by the different source strength but is also influenced by the measurement statistics. The detection of urban CO concentration under low cloud conditions perform comparably well with an enhancement of 8.8 ppb for Tehran, 8.3 ppb for Los Angeles and 3.4 ppb for Paris, where for Tehran and Los Angeles the spatial distribution of the enhancements agree even better with the urban area contours. For observations with medium-high clouds, we see a less distinct CO enhancement over the three cities with 7.0 ppb for Tehran, 3.6 ppb for Los ~~Angles~~ Angeles and only 1.8 ppb for Paris. Medium-high clouds shield the atmosphere below and so the retrieval is less sensitive to the city pollution estimating the CO column from the measurement sensitivity above the cloud as already indicated by the column averaging kernels in Fig. 1.

Consequently, measurements contaminated by medium-high clouds in combination with clear-sky and low cloud retrievals can reveal information about the strength and vertical extension of the CO pollution.

Furthermore, including the cloudy retrievals improves the measurement statistics as indicated in Fig. 10. Including cloud contaminated soundings means about double the amount of data is available for Tehran (a factor of 2.1) and Paris (a factor of 2.6) and Los Angeles (a factor of 1.8). The relative amount of cloud contaminated measurements differs significantly per city. ~~For Tehran, we obtain 47 % (2674) clear-sky and 44 % (2501) and 9 % (537) low and medium-high cloud observations, respectively. A similar distribution holds for Los Angeles with 55 % (2557) and 35 % (1630) and 10 % (482) soundings for low and medium cloud conditions, whereas for Paris the situation differs with 38 % (1338) clear-sky soundings and 22 % (766) and 40 % (1388) low and medium cloud soundings.~~ Los Angeles show a similar distribution with a high number of clear-sky and 35 % (1630) and 10 % (482) soundings for low and medium cloud conditions, whereas for Paris the situation differs with 38 % (1338) clear-sky soundings and 22 % (766) and 40 % (1388) low and medium cloud soundings. Overall, we conclude that for the SCIAMACHY mission, cloud contaminated measurements provide valuable and complementary information to clear-sky measurements.

5 Implications for TROPOMI

The TROPOMI instrument was successfully launched on the ESA's Sentinel-5 Precursor mission on October 13, 2017. The 2.3 μm spectral range ~~of TROPOMI is covered by~~ is covered both by TROPOMI and SCIAMACHY with the same spectral resolution ~~and spectral coverage but TROPOMI is characterized by a significantly improved SNR of the measurements, a higher,~~ whereby TROPOMI shows an improved radiometric performance with a high spatial resolution of up to $7 \times 7 \text{ km}^2$; ~~and improved spatial sampling and~~ with daily global coverage. The spectral analogy of TROPOMI and SCIAMACHY allowed us to apply the operational TROPOMI CO retrieval algorithm SICOR on the SCIAMACHY spectra to test its performance for cloud contaminated measurements in preparation of TROPOMI data exploitation.

The spatial sampling of continuous TROPOMI nadir SWIR measurements with a swath of 2600 km provides 300 times more soundings compared to the limb-nadir observations of SCIAMACHY with a ground pixel size of 120x30 km and a swath of 960 km. Due to the higher SNR of TROPOMI SWIR measurements, CO total column will be provided with a precision < 10 % (Landgraf et al., 2016a, b) compared to the SCIAMACHY CO column precision of 100 % and larger (Gloudemans et al., 2008). Also the radiometric accuracy is significantly improved leading to a overall bias estimate of the TROPOMI CO columns < 10 % for ~~clear-sky~~ clear-sky and cloudy observations (Landgraf et al., 2016a, b). Therefore, TROPOMI SWIR measurements will capture burning events, with atmospheric CO signatures significantly weaker than investigated in this study, and urban pollution on a day-to-day basis with high spatial resolution without precedent. Here, using cloudy data complementary to clear-sky observations will give us a new opportunity to study the vertical and horizontal distribution of atmospheric CO pollution. First results of the TROPOMI CO dataset are reported in Borsdorff et al. (2018).

6 Summary and Conclusions

In this study, we discussed the benefit of using CO total column ~~retrieval~~retrievals from cloud contaminated SCIAMACHY 2.3 μm ~~shortwave-infrared-SWIR~~ spectra to study pollution from ~~megacities~~cities and wild fires ~~complimentary~~complementary to clear-sky soundings. For this purpose, we applied the SICOR algorithm to SCIAMACHY observations. SICOR ~~is~~was developed for the operational processing of the ~~shortwave-infrared-SWIR~~ measurements of the TROPOMI instrument on ESA's Sentinel-5 Precursor. ~~It~~SICOR provides the possibility to retrieve effective cloud parameters together with trace gas columns. To investigate the capability to detect localized CO enhancements at urban areas and wild fires, we distinguished between retrievals under clear-sky, low cloud and medium-high cloud atmospheric conditions. As an example, we analyzed CO enhancements over the ~~megacities~~cities Paris, Los Angeles, and Tehran as well as the wildfire events in Mexico/Guatemala 2005 and Alaska/Canada 2004.

After data averaging over the entire mission period, we found that SCIAMACHY mean clear-sky observations can detect weak CO enhancements of less than 10 ppb over the three considered ~~megacities~~cities and coincide with the MODIS urban area contours. For Paris, ~~it was even possible to detect pollution over~~we detected enhanced CO values next to the neighboring city of Rouen, that can be caused by the city itself or transport of remote pollution of Paris. Furthermore, clear-sky retrievals turned out to be suitable to locate the source of biomass burning in Mexico/Guatemala in agreement with most of the burned area reported by the daily GFED4 ~~Burned-Area-data~~ product. Here, the sensitivity of SWIR measurements to CO throughout the atmosphere including the planetary boundary layer makes clear-sky retrievals a preferable choice for the detection of such sources. However, only a fraction of all measurements fall into this category. For example, due to the meteorological situation during the Alaska/Canada 2004 burning event, insufficient ~~clear-sky~~clear-sky measurements were available to fully capture the wild fires. Moreover, the noise of the retrievals strongly depends on the surface reflectivity. We found clear-sky retrievals for the wild fires in Mexico/Guatemala 2005 inferior to cloudy retrievals regarding the noise performance ~~and~~(clouds are high reflectivity in the SWIR) and the temporal and spatial sampling.

Considering pollutions from ~~megacities~~cities, the CO retrieval performs equally well for ~~clear-sky~~clear-sky and low cloud measurements. ~~For Tehran and Los Angeles~~This is probably because both are sensitive to CO in the planetary boundary layer. Compared to clear-sky observations, the temporal and spatial sampling of low cloud observations ~~improved the spatial coincidence improves the spatial match~~ of the CO ~~enhancement and the MODIS urban area product compared to the clear-sky CO product~~enhancements with the corresponding MODIS urban areas of Tehran and Los Angeles. The low cloud retrievals of the 2005 wild fires in Mexico/Guatemala ~~provides~~provide complementary information compared to clear-sky retrievals indicating the CO outflow over the Gulf of Mexico and the Pacific Ocean, which is confirmed by smoke observations of the MODIS/Aqua instrument. Here, the high reflectivity of clouds allows the retrieval of CO over oceans, which was not possible with clear-sky measurements due to the dark ocean surface in the ~~shortwave-infrared-SWIR~~ spectral range.

~~The analysis of the CO pollution from megacities from SCIAMACHY soundings with~~However, when using medium-high ~~clouds indicated~~clouds for the detection of CO pollution we recognized a significant reduction in the CO enhancement ~~for~~above the three cities. Here, clouds shield the CO pollution and consequently, the retrieval underestimates the total column

of CO. This effect differs for the three cities. While the pollution from Tehran and Los ~~Angles~~-Angeles are still present in the data product for medium-high clouds, it nearly vanishes for Paris, pointing to a CO enhancement localized in the lowest altitude range. Comparing low and medium-high cloud conditions for the Mexico fires, the CO enhancement is detected equally well, which indicates that the CO emission by this strong burning reaches the free troposphere. These examples show that CO
5 retrievals for different cloud conditions are valuable to gain information about the vertical extent of the atmospheric CO pollution.

Overall, the study of SCIAMACHY CO retrievals from cloud contaminated 2.3 μm measurements showed the additive value of the data product compared to ~~clear-sky~~-clear-sky retrievals to study CO pollution on regional and urban scales. Particularly in perspective of the upcoming Sentinel-5 Precursor mission with the TROPOMI instrument as its single payload,
10 the corresponding CO data product will open ~~op~~-up new research opportunities due to the groundbreaking capabilities of the TROPOMI instrument.

7 Data availability

The full-mission SCIAMACHY CO data set used in this study, including clear-sky and cloudy-sky observations is available for download at ftp://ftp.sron.nl/pub/pub/DataProducts/SCIAMACHY_CO/. The underlying data of the figures presented in this
15 publication can be found at <ftp://ftp.sron.nl/open-access-data/>.

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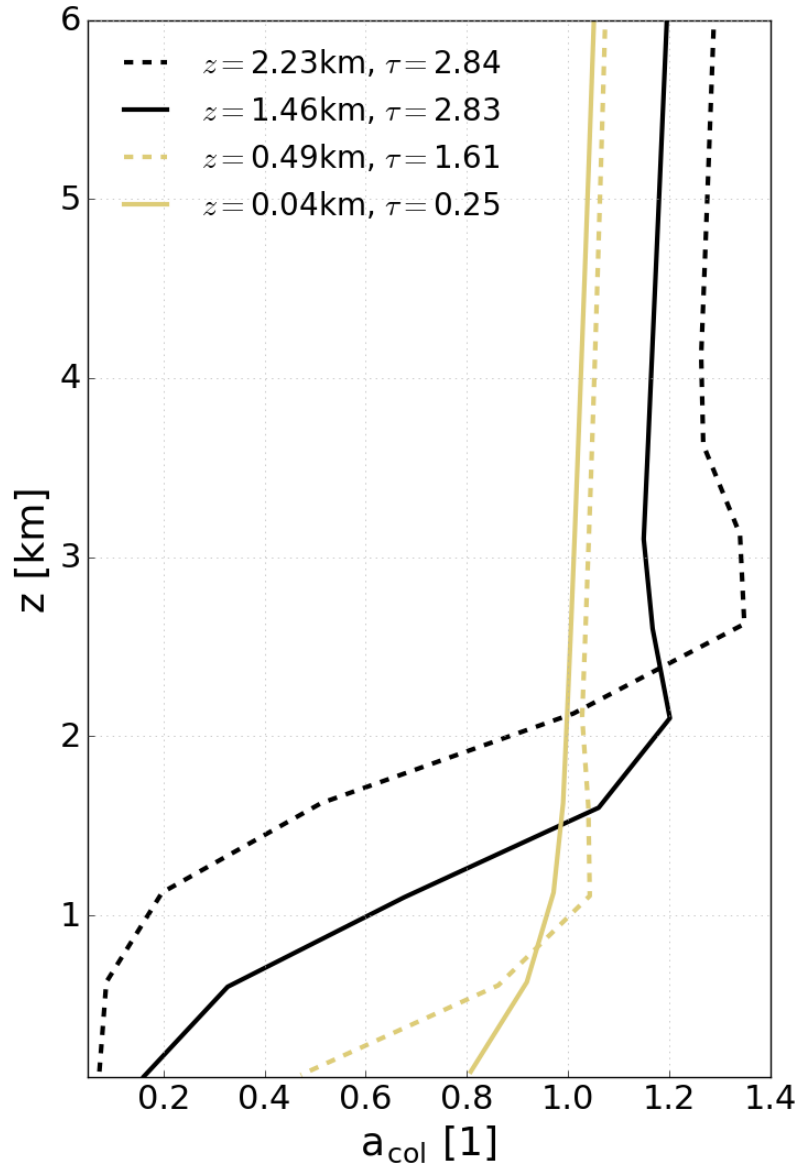


Figure 1. SCIAMACHY CO total column averaging kernels for different cloud centre heights (z_{cld}) and cloud optical thicknesses (τ_{cld}). [Here, the solid yellow line is representative for clear-sky conditions.](#) The figure shows typical cases of the vertical retrieval sensitivity of the SCIAMACHY CO retrievals over Paris.

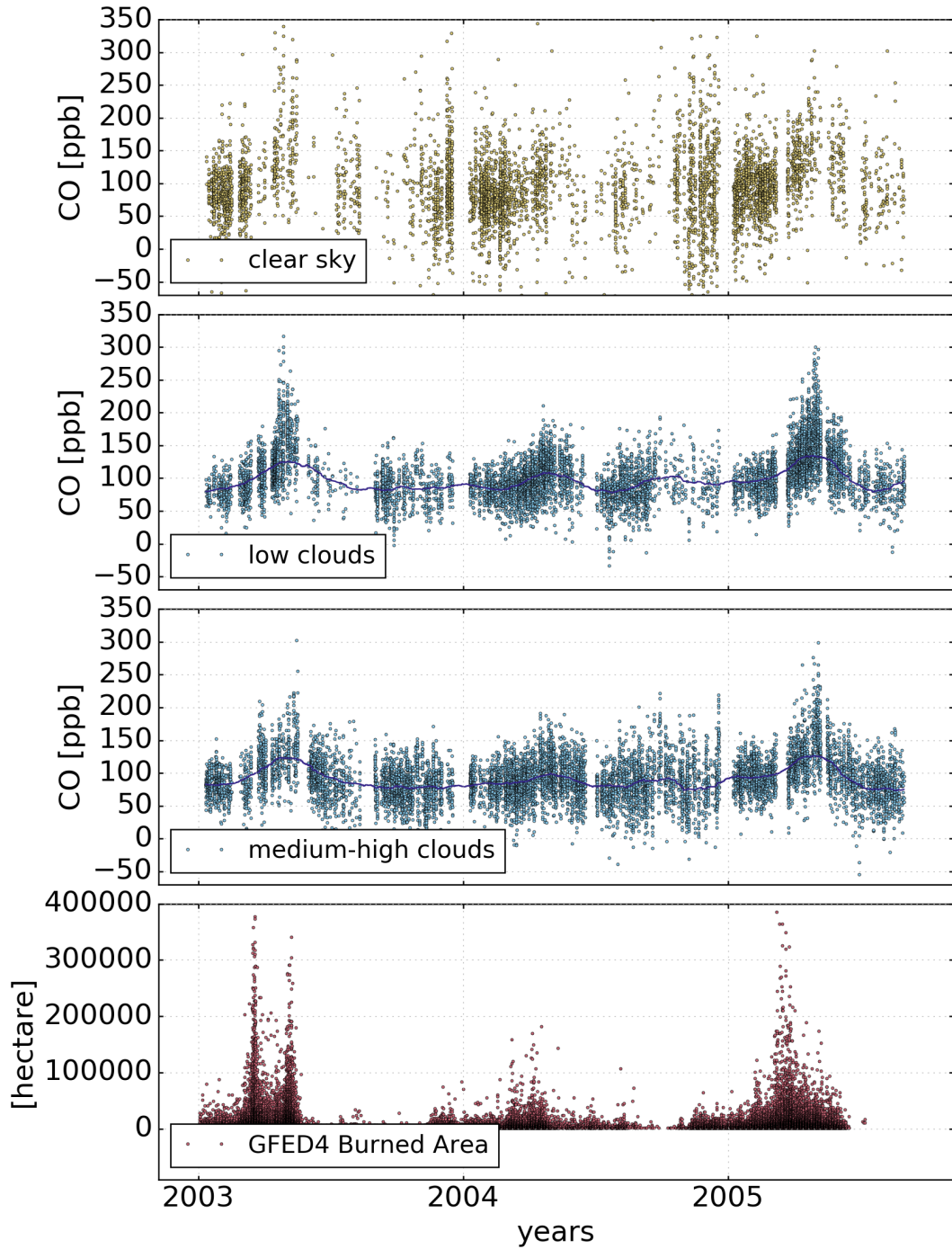


Figure 2. Individual SCIAMACHY CO retrievals under clear-sky, low cloud, and medium-high cloud atmospheric conditions as well as daily GFED4 Burned Area over Mexico/Guatemala in the latitude/longitude box [(22.5°N,100.0°W), (10.0°N,80.0°W)]. The blue line is a running median with half width of 30 days.

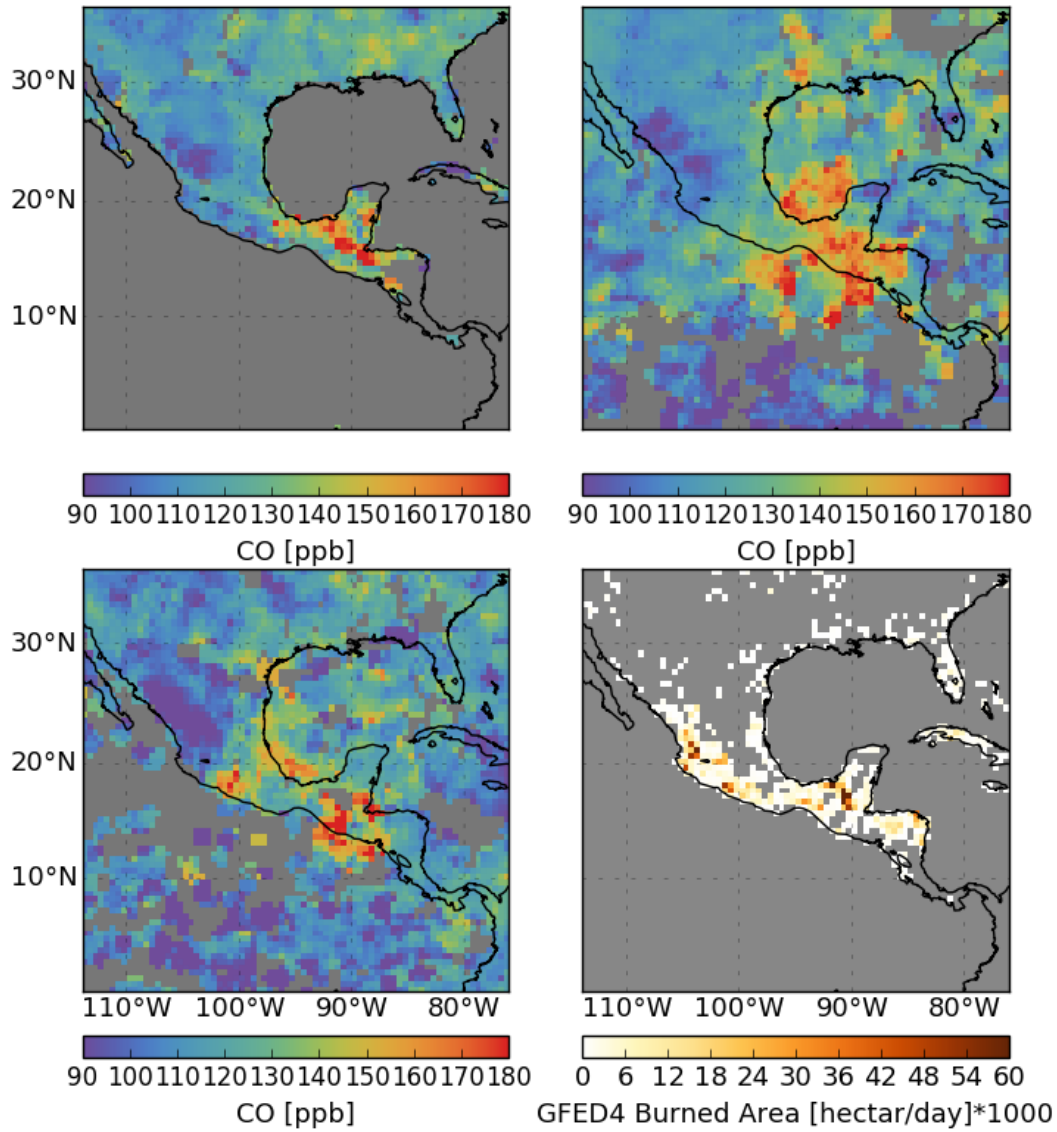


Figure 3. SCIAMACHY CO retrievals under clear-sky (top left), low cloud (top right), medium-high cloud (bottom left) atmospheric condition as well as daily GFED4 Burned Area (bottom right) averaged from 15th March to 15th May 2005 over Mexico and Central America in the latitude/longitude box (36.0°N, 113.9°W), (0.0°N, 76.1°W). The resolution of the plot is 0.5 degree in latitude and longitude and the data is oversampled using a radius of 90km.

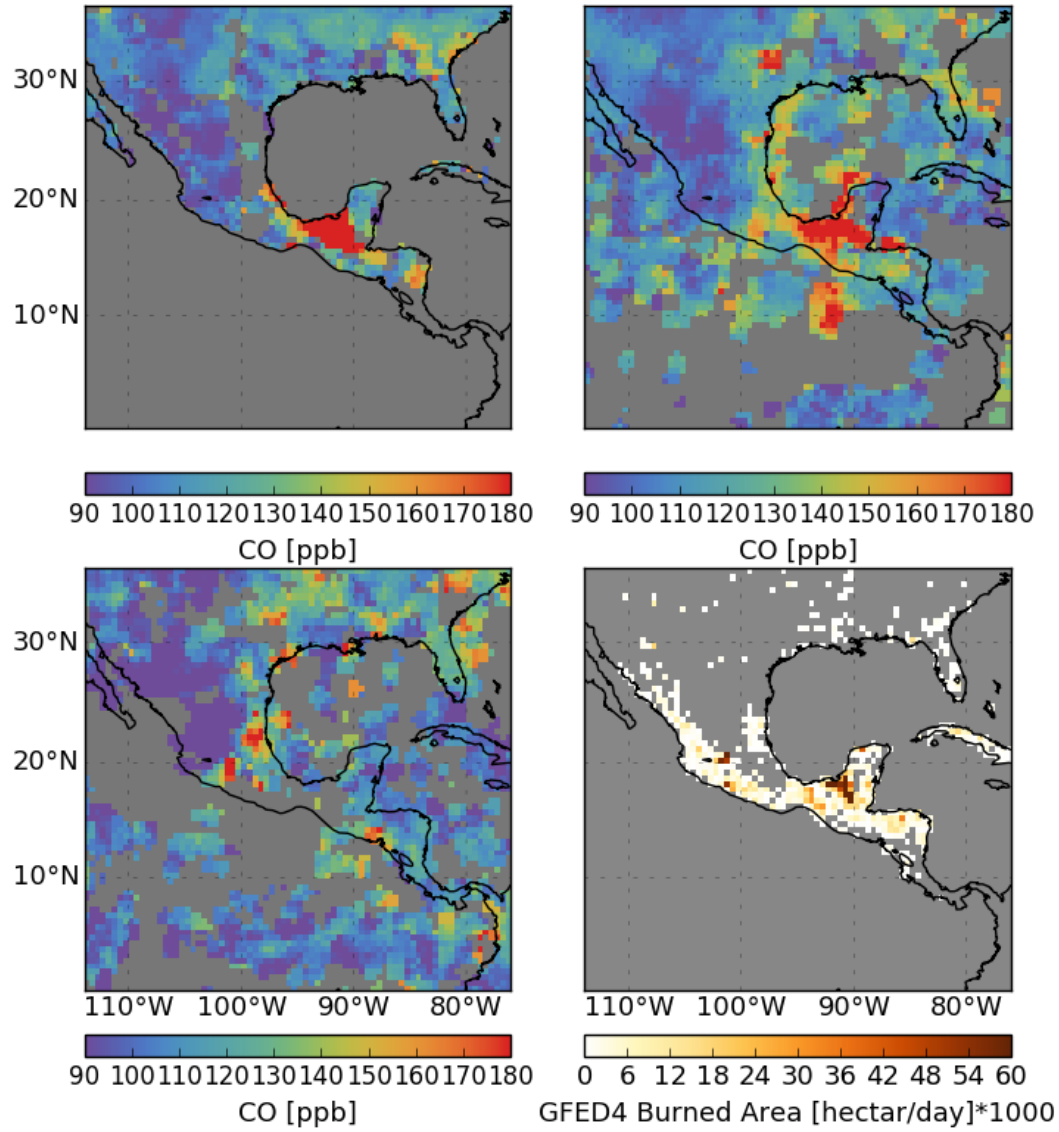


Figure 4. Same as Fig. 3 but ~~for Alaska/Canada. The~~ data is averaged from 1st ~~July-April~~ to 1st August 2004 in the latitude/longitude ~~box (75.0°N,175.0°W), (52.0°N,90.0°W).~~ 15th May 2003.

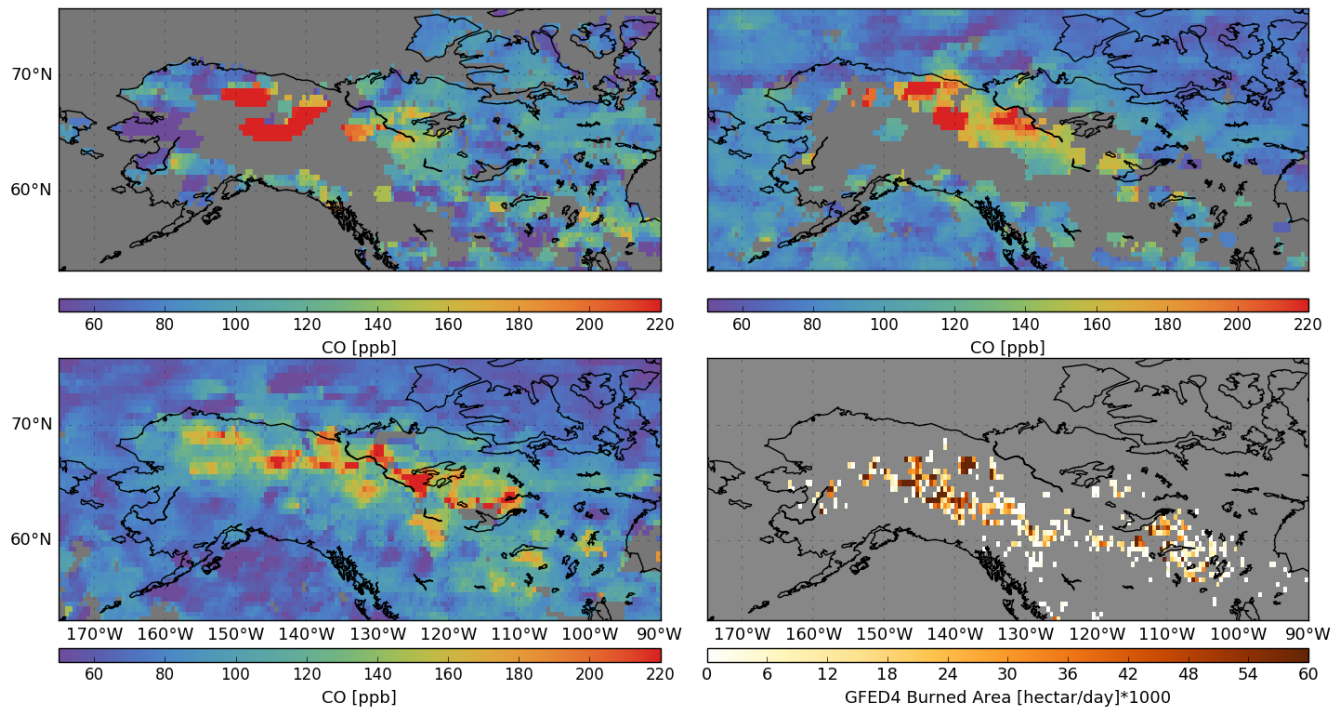


Figure 5. Same as Fig. 3 but for Alaska/Canada. The data is averaged from 1st July to 1st August 2004.

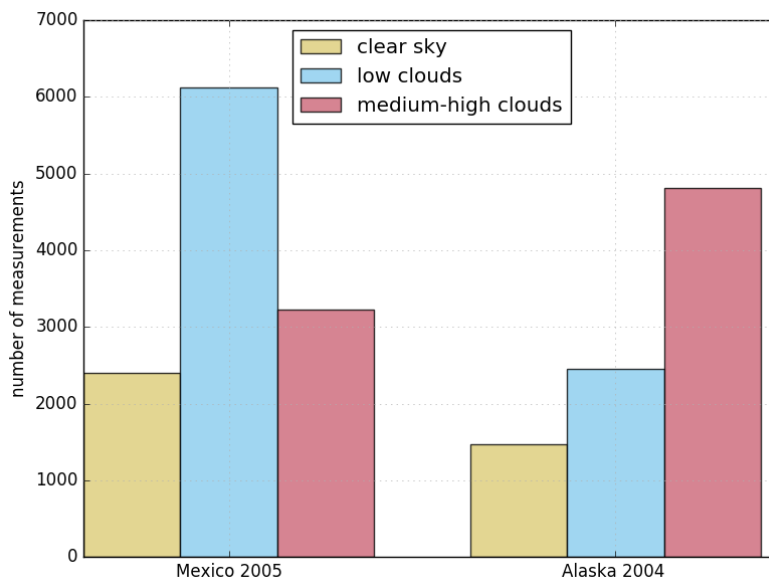


Figure 6. Number of individual SCIAMACHY CO retrievals under clear-sky (yellow), low cloud (blue), and medium-high cloud (pink) atmospheric conditions for the time range and latitude/longitude box specified in Fig. 3 and Fig. 5.

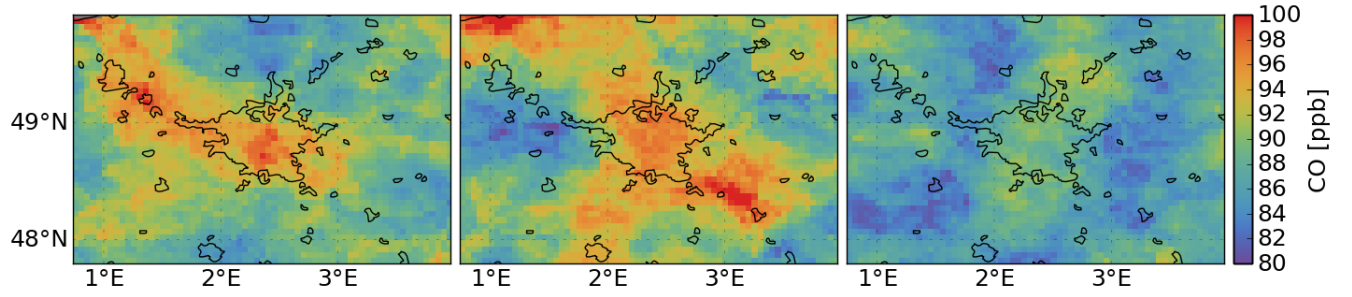


Figure 7. SCIAMACHY CO column mixing ratio averaged from January 2003 to April 2012 in the latitude/longitude box under clear-sky (49.9°N,0.7°E left panel), low cloud (47.8°N middle panel), 4.0°E and medium-high cloud (right panel) atmospheric conditions above Paris. The resolution-spatial sampling of the plot is 0.05- $\delta = 0.05$ degree in latitude and longitude and the data is oversampled using a are averaged with radius of 40km $r = 40$ km. The urban area contours are based on MODIS measurements Schneider et al. (2009).

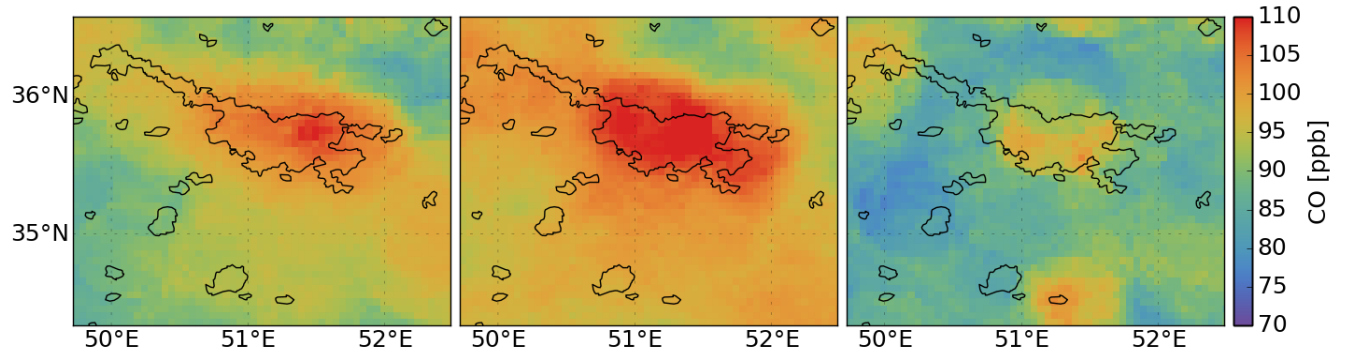


Figure 8. Same as Fig. 7 but for Tehran using the latitude/longitude box (36.6°N,49.7°E), (34.3°N,52.5°E).

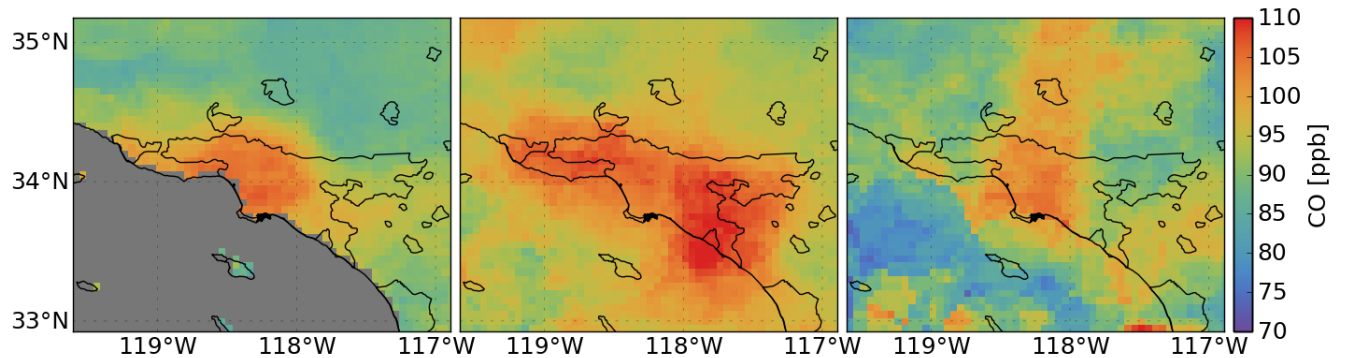


Figure 9. Same as Fig. 7 but for Los Angeles using the latitude/longitude box (35.2°N,119.6°W), (32.9°N,116.9°W).

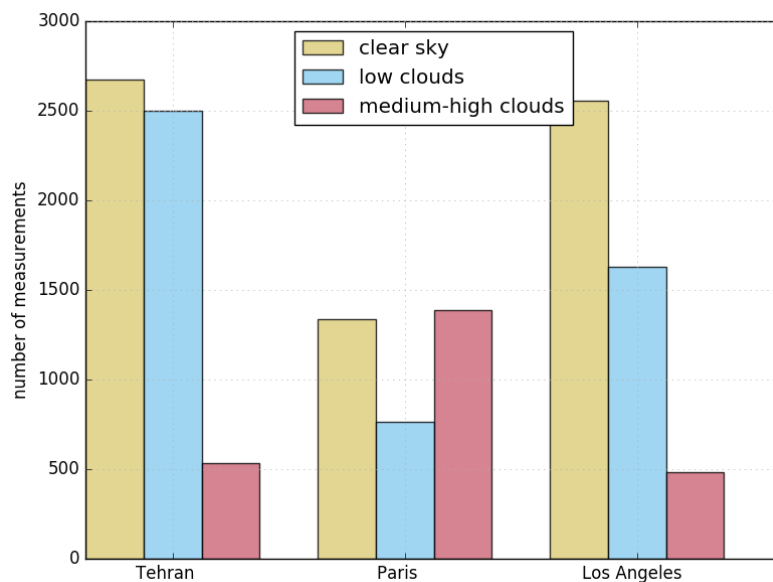


Figure 10. Number of individual SCIAMACHY CO retrievals under clear-sky (yellow), low cloud (blue), and medium-high cloud (pink) atmospheric condition for the time range and latitude/longitude box specified in Fig. 7, 8, and 9.

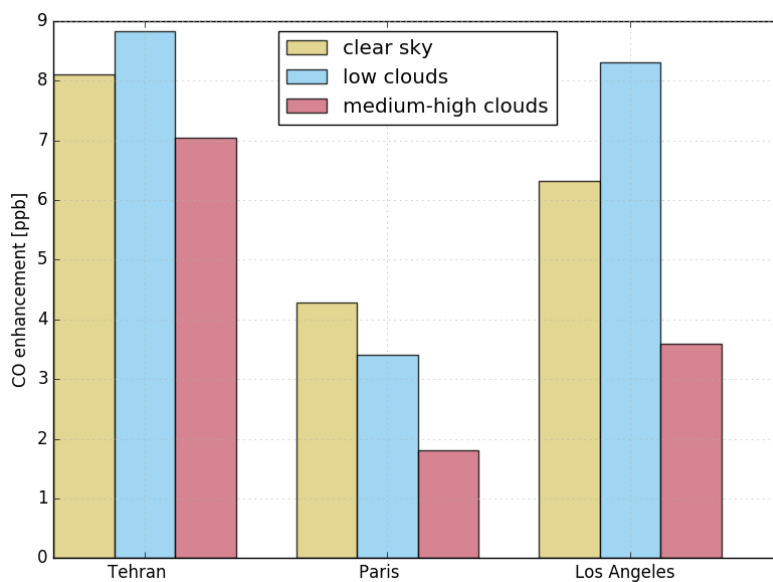


Figure 11. CO enhancement over cities shown in Fig. 7, 8, and 9 relative to the background concentration under clear-sky (yellow), low cloud (blue), and medium-high cloud (pink) atmospheric conditions. The difference of the median CO concentration inside and outside the urban area contours of each city is shown.

category	optical depth	cloud height	SNR
clear-sky observations	$\tau_{cld} < 2$	$z_{cld} < 0.5\text{km}$	$\text{SNR}_{\text{max}} > 15$
observations with low clouds	$\tau_{cld} > 2$	$z_{cld} < 1.5\text{km}$	$\text{SNR}_{\text{max}} > 100$
observations with medium-high cloud	$\tau_{cld} > 2$	$1.5\text{km} < z_{cld} < 5\text{km}$	$\text{SNR}_{\text{max}} > 100.$

Table 1. Categories of cloudy observations defined by the retrieved cloud optical depth τ_{cld} , the cloud height z_{cld} and the spectral maximum of the measurement SNR.

<u>cities</u>	<u>clear-sky</u>	<u>low cloud</u>	<u>medium-high cloud</u>
<u>Tehran</u>	<u>47% (2674)</u>	<u>44% (2501)</u>	<u>9% (537)</u>
<u>Los Angeles</u>	<u>55% (2557)</u>	<u>35% (1630)</u>	<u>10% (482)</u>
<u>Paris</u>	<u>38% (1338)</u>	<u>22% (766)</u>	<u>40% (1388)</u>

Table 2. Number of clear-sky and cloud contaminated measurements for the cities Tehran, Paris, and Los Angeles. The absolute number of observations are given in brackets