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

Tobias Borsdorff[1], Josip Andrasec[1], Joost aan de Brugh[1], Haili Hu[1], Ilse Aben[1], and Jochen Landgraf[1]

[1]SRON Netherlands Institute for Space Research, Utrecht, the Netherlands

Correspondence to: T. Borsdorff (t.borsdorff@sron.nl)

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 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 (1.5km-5km). As an example, we analyze CO enhancements over the 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 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 radial (OH) represents its major sink (Spivakovsky et al., 2000). Because of its moderate long lifetime 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 (Measure-

ments 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; Gloudemans et al. (2009),Frankenberg et al. (2005),Buchwitz et al. (2007), and Gimeno Garcia et al. (2011))).

5   The presence of clouds 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), and Borsdorff et al. (2016) suggest to consider only observations under clear-sky conditions or weakly cloud contaminated, assuming that the sensitivity to CO in the lower tropo-
10   sphere is sufficient to estimate the total CO column. A different approach is followed by Buchwitz et al. (2007), Gloudemans 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 reduced sensitivity when retrieving trace gas columns from cloud contaminated measurements. Alternatively, 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
15   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 cities. Also, the pronounced
20   enhancement of CO due to wildfires have been reported (e.g., Gloudemans 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, cloudy and clear-sky observations with different vertical CO sensitivities provide information on the
25   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). The mission objectives and requirements are provided by (Veefkind et al., 2012).

30   It measures Earth reflected radiances in the ultraviolet, visible, near infrared and shortwave infrared spectral range with a spatial resolution of about 7×7 km^2 at sub-satellite point and daily global coverage. Here, shortwave infrared (SWIR) observations in the 2.3 μm spectral region provide information on the CO total column amount. In recent years, the 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 CO total
35   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 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

5 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 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

10 type of cloud contamination.

We have applied the SICOR algorithm to the full SCIAMACHY 2.3 μm 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

15 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 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 μm measurements with their intrinsic vertical sensitivity for the detection of CO pollution from cities and wildfires. We separate CO retrievals under clear-sky, low (<1.5 km), and

20 medium-high (1.5-5 km) cloud conditions. As an example, we discuss CO pollution over Tehran, Paris, and Los Angeles as well as the wildfires in Mexico/Guatemala 2005 (Herrera, 2016) and Alaska/Canada 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 cities. Section 5 draws some conclusions on the upcoming TROPOMI CO dataset. The summary and conclusion is given in

25 section 6 and finally, section 7 states the availability of the data.

2   SCIAMACHY CO dataset and retrievals

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 120x30 km^2, 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

30 densities, retrieved from individual SCIAMACHY 2.3μm spectra for the entire period of the mission from January 2003 until April 2012. The CO data product consists of the estimates of the total column concentrations of CO, H$_2$O and HDO (c_{CO}, c_{H2O}, c_{HDO}), the corresponding retrieval noise (ϵ_{CO}, ϵ_{H2O}, ϵ_{HDO}), averaging kernels, effective cloud parameters (cloud optical thickness τ_{cld} and cloud height z_{cld}) and the SWIR Lambertian surface albedo. Auxiliary parameters like signal-to-

3

noise ratio SNR_{max} of the measurement and number of retrieval iterations (N_{iter}) are also provided. SICOR uses the profile scaling approach, discussed in detail by Vidot et al. (2012), Borsdorff et al. (2014), and Landgraf et al. (2016b). Here, cloud optical depth and height are estimated using prior knowledge about CH_4, ECMWF surface pressure, and the observed CH_4 absorption in the 2.3 μm spectral fit window (Landgraf et al., 2016a). The CH_4 data was taken from a TM5 model run (Williams et al., 2013, 2014) spanning the entire mission period of SCIAMACHY with global 3×2 degree^2 horizontal 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 CH_4 absorption line, which are beneficial for the retrieval of the effective cloud parameters. Due to an ice layer on the SWIR detectors and the 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 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. $N_{iter} < 15$

2. $\epsilon_{CO} < 4.5\,\sigma_{CO}$

3. $\epsilon_{H2O} < 4.5\,\sigma_{H2O}$

4. $\epsilon_{HDO} < 4.5\,\sigma_{HDO}$.

For the analysis of air pollution from 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 cities. It enables us to detect the relatively weak CO enhancement above cities after averaging the data over the entire mission period. 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

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

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 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 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.

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 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 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. 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 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 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 cloudy retrievals are in good agreement with the 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 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 (as shown in Fig. 4). The

6

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 and low cloud observations do not fully capture the

5   Alaska 2004 fire event. For medium-high clouds, the corresponding CO product shows much better coverage and so can detect 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

10   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

15   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 observations.

4   CO pollution from cities

In this section, we selected the three cities Paris, Tehran, and Los Angeles to discuss the relevance of cloudy observations for

20   the detection of urban pollution. We accumulated all SCIAMACHY observations from 2003 to April 2012 around these cities, distinguishing between clear-sky, low-cloud and medium-high cloud retrievals. 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 (Figs. 7, 8, and 9). 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

25   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 detect enhanced CO levels 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

30   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 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.

5 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

10 and is summaries in Tab. 2. Tehran and Los Angeles show a similar distribution with a high number of clear-sky and low cloud observations, whereas, for Paris cloudy measurements are more predominant. 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

15 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 7×7 km^2 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.

20 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

25 columns < 10 % for 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 retrievals from cloud contaminated SCIAMACHY 2.3 μm SWIR spectra to study pollution from cities and wild fires complementary to clear-sky soundings. For this purpose, we applied the SICOR algorithm to SCIAMACHY observations. SICOR was developed for the operational processing of the SWIR measurements of the TROPOMI instrument on ESA's Sentinel-5 Precursor. 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 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 cities and coincide with the MODIS urban area contours. For Paris, 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 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 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 (clouds are high reflectivity in the SWIR) and the temporal and spatial sampling.

Considering pollutions from cities, the CO retrieval performs equally well for clear-sky and low cloud measurements. 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 improves the spatial match of the CO enhancements with the corresponding MODIS urban areas of Tehran and Los Angeles. The low cloud retrievals of the 2005 wild fires in Mexico/Guatemala 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 SWIR spectral range.

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. 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 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

9

that CO 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 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, the corresponding CO data product will open 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 publication can be found at ftp://ftp.sron.nl/open-access-data/.

References

Borsdorff, T., Hasekamp, O. P., Wassmann, A., and Landgraf, J.: Insights into Tikhonov regularization: application to trace gas column retrieval and the efficient calculation of total column averaging kernels, Atmospheric Measurement Techniques, 7, 523–535, https://doi.org/10.5194/amt-7-523-2014, https://doi.org/10.5194%2Famt-7-523-2014, 2014.

Borsdorff, T., Tol, P., Williams, J. E., de Laat, J., aan de Brugh, J., Nédélec, P., Aben, I., and Landgraf, J.: Carbon monoxide total columns from SCIAMACHY 2.3 μm atmospheric reflectance measurements: towards a full-mission data product (2003-2012), Atmospheric Measurement Techniques, 9, 227–248, https://doi.org/10.5194/amt-9-227-2016, https://doi.org/10.5194%2Famt-9-227-2016, 2016.

Borsdorff, T., aan de Brugh, J., Hu, H., Nédélec, P., Aben, I., and Landgraf, J.: Carbon monoxide column retrieval for clear-sky and cloudy atmospheres: a full-mission data set from SCIAMACHY 2.3 μm reflectance measurements, Atmospheric Measurement Techniques, 10, 1769–1782, https://doi.org/10.5194/amt-10-1769-2017, https://www.atmos-meas-tech.net/10/1769/2017/, 2017.

Borsdorff, T., de Brugh, J. A., Hu, H., Aben, I., Hasekamp, O., and Landgraf, J.: Measuring Carbon Monoxide With TROPOMI: First Results and a Comparison With ECMWF-IFS Analysis Data, Geophysical Research Letters, 0, https://doi.org/10.1002/2018GL077045, https://agupubs.onlinelibrary.wiley.com/doi/abs/10.1002/2018GL077045, 2018.

Bovensmann, H., Burrows, J. P., Buchwitz, M., Frerick, J., Noël, S., Rozanov, V. V., Chance, K. V., and Goede, A. P. H.: SCIA-MACHY: Mission Objectives and Measurement Modes, Journal of the Atmospheric Sciences, 56, 127–150, https://doi.org/10.1175/1520-0469(1999)056<0127:smoamm>2.0.co;2, https://doi.org/10.1175%2F1520-0469%281999%29056%3C0127%3Asmoamm%3E2.0.co%3B2, 1999.

Buchwitz, M., de Beek, R., Bramstedt, K., Noël, S., Bovensmann, H., and Burrows, J. P.: Global carbon monoxide as retrieved from SCIAMACHY by WFM-DOAS, Atmospheric Chemistry and Physics, 4, 1945–1960, https://doi.org/10.5194/acp-4-1945-2004, https://doi.org/10.5194%2Facp-4-1945-2004, 2004.

Buchwitz, M., Khlystova, I., Bovensmann, H., and Burrows, J. P.: Three years of global carbon monoxide from SCIAMACHY: comparison with MOPITT and first results related to the detection of enhanced CO over cities, Atmospheric Chemistry and Physics, 7, 2399–2411, https://doi.org/10.5194/acp-7-2399-2007, https://doi.org/10.5194%2Facp-7-2399-2007, 2007.

Clerbaux, C., Edwards, D. P., Deeter, M., Emmons, L., Lamarque, J.-F., Tie, X. X., Massie, S. T., and Gille, J.: Carbon monoxide pollution from cities and urban areas observed by the Terra/MOPITT mission, Geophysical Research Letters, 35, n/a–n/a, https://doi.org/10.1029/2007GL032300, http://dx.doi.org/10.1029/2007GL032300, l03817, 2008.

de Laat, A. T. J., Gloudemans, A. M. S., Schrijver, H., van den Broek, M. M. P., Meirink, J. F., Aben, I., and Krol, M.: Quantitative analysis of SCIAMACHY carbon monoxide total column measurements, Geophysical Research Letters, 33, n/a–n/a, https://doi.org/10.1029/2005GL025530, http://dx.doi.org/10.1029/2005GL025530, l07807, 2006.

de Laat, A. T. J., Gloudemans, A. M. S., Aben, I., Krol, M., Meirink, J. F., van der Werf, G. R., and Schrijver, H.: Scanning Imaging Absorption Spectrometer for Atmospheric Chartography carbon monoxide total columns: Statistical evaluation and comparison with chemistry transport model results, Journal of Geophysical Research, 112, https://doi.org/10.1029/2006jd008256, https://doi.org/10.1029%2F2006jd008256, 2007.

de Laat, A. T. J., Dijkstra, R., Schrijver, H., Nédélec, P., and Aben, I.: Validation of six years of SCIAMACHY carbon monoxide observations using MOZAIC CO profile measurements, Atmospheric Measurement Techniques, 5, 2133–2142, https://doi.org/10.5194/amt-5-2133-2012, https://doi.org/10.5194%2Famt-5-2133-2012, 2012.

Deeter, M. N.: Operational carbon monoxide retrieval algorithm and selected results for the MOPITT instrument, Journal of Geophysical Research, 108, https://doi.org/10.1029/2002jd003186, https://doi.org/10.1029%2F2002jd003186, 2003.

Fioletov, V. E., McLinden, C. A., Krotkov, N., Moran, M. D., and Yang, K.: Estimation of SO2 emissions using OMI retrievals, Geophysical Research Letters, 38, n/a–n/a, https://doi.org/10.1029/2011GL049402, http://dx.doi.org/10.1029/2011GL049402, l21811, 2011.

5   Frankenberg, C., Platt, U., and Wagner, T.: Retrieval of CO from SCIAMACHY onboard ENVISAT: detection of strongly polluted areas and seasonal patterns in global CO abundances, Atmospheric Chemistry and Physics, 5, 1639–1644, https://doi.org/10.5194/acp-5-1639-2005, https://doi.org/10.5194%2Facp-5-1639-2005, 2005.

Gimeno Garcia, S., Schreier, F., Lichtenberg, G., and Slijkhuis, S.: Near infrared nadir retrieval of vertical column densities: methodology and application to SCIAMACHY, Atmospheric Measurement Techniques, 4, 2633–2657, https://doi.org/10.5194/amt-4-2633-2011, https: 10   //www.atmos-meas-tech.net/4/2633/2011/, 2011.

Gloudemans, A. M. S., Krol, M. C., Meirink, J. F., de Laat, A. T. J., van der Werf, G. R., Schrijver, H., van den Broek, M. M. P., and Aben, I.: Evidence for long-range transport of carbon monoxide in the Southern Hemisphere from SCIAMACHY observations, Geophysical Research Letters, 33, https://doi.org/10.1029/2006gl026804, https://doi.org/10.1029%2F2006gl026804, 2006.

Gloudemans, A. M. S., Schrijver, H., Hasekamp, O. P., and Aben, I.: Error analysis for CO and CH4 total column retrievals from 15   SCIAMACHY 2.3 μm spectra, Atmospheric Chemistry and Physics, 8, 3999–4017, https://doi.org/10.5194/acp-8-3999-2008, https: //doi.org/10.5194%2Facp-8-3999-2008, 2008.

Gloudemans, A. M. S., de Laat, A. T. J., Schrijver, H., Aben, I., Meirink, J. F., and van der Werf, G. R.: SCIAMACHY CO over land and oceans: 2003–2007 interannual variability, Atmospheric Chemistry and Physics, 9, 3799–3813, https://doi.org/10.5194/acp-9-3799-2009, https://doi.org/10.5194%2Facp-9-3799-2009, 2009.

20   Herrera, G. V.: Mexican forest fires and their decadal variations, Advances in Space Research, 58, 2104 – 2115, https://doi.org/https://doi.org/10.1016/j.asr.2016.08.030, http://www.sciencedirect.com/science/article/pii/S0273117716304835, space and Geophysical Research related to Latin America - Part 2, 2016.

Holloway, T., Levy, H., and Kasibhatla, P.: Global distribution of carbon monoxide, Journal of Geophysical Research: Atmospheres, 105, 12 123–12 147, https://doi.org/10.1029/1999jd901173, https://doi.org/10.1029%2F1999jd901173, 2000.

25   Krijger, J. M., Aben, I., and Schrijver, H.: Distinction between clouds and ice/snow covered surfaces in the identification of cloud-free observations using SCIAMACHY PMDs, Atmospheric Chemistry and Physics, 5, 2729–2738, https://doi.org/10.5194/acp-5-2729-2005, https://doi.org/10.5194%2Facp-5-2729-2005, 2005.

Krijger, J. M., Tol, P., Istomina, L. G., Schlundt, C., Schrijver, H., and Aben, I.: Improved identification of clouds and ice/snow covered surfaces in SCIAMACHY observations, Atmospheric Measurement Techniques, 4, 2213–2224, https://doi.org/10.5194/amt-4-2213-2011, 30   https://www.atmos-meas-tech.net/4/2213/2011/, 2011.

Landgraf, J., aan de Brugh, J., Borsdorff, T., Houweling, S., and O., H.: Algorithm Theoretical Baseline Document for Sentinel-5 Precursor: Carbon Monoxide Total Column Retrieval, Atbd, SRON, Sorbonnelaan 2, 3584 CA Utrecht, The Netherlands, 2016a.

Landgraf, J., aan de Brugh, J., Scheepmaker, R., Borsdorff, T., Hu, H., Houweling, S., Butz, A., Aben, I., and Hasekamp, O.: Carbon monoxide total column retrievals from TROPOMI shortwave infrared measurements, Atmospheric Measurement Techniques, 9, 4955– 35   4975, https://doi.org/10.5194/amt-9-4955-2016, https://doi.org/10.5194%2Famt-9-4955-2016, 2016b.

Liu, C., Beirle, S., Butler, T., Hoor, P., Frankenberg, C., Jöckel, P., Penning de Vries, M., Platt, U., Pozzer, A., Lawrence, M. G., Lelieveld, J., Tost, H., and Wagner, T.: Profile information on CO from SCIAMACHY observations using cloud slicing and compar-

ison with model simulations, Atmospheric Chemistry and Physics, 14, 1717–1732, https://doi.org/10.5194/acp-14-1717-2014, https://www.atmos-chem-phys.net/14/1717/2014/, 2014.

Logan, J. A., Prather, M. J., Wofsy, S. C., and McElroy, M. B.: Tropospheric chemistry: A global perspective, Journal of Geophysical Research, 86, 7210, https://doi.org/10.1029/jc086ic08p07210, https://doi.org/10.1029%2Fjc086ic08p07210, 1981.

5 McMillan, W. W.: Daily global maps of carbon monoxide from NASA's Atmospheric Infrared Sounder, Geophysical Research Letters, 32, https://doi.org/10.1029/2004gl021821, https://doi.org/10.1029%2F2004gl021821, 2005.

Pfister, G., Hess, P. G., Emmons, L. K., Lamarque, J.-F., Wiedinmyer, C., Edwards, D. P., PÈtron, G., Gille, J. C., and Sachse, G. W.: Quantifying CO emissions from the 2004 Alaskan wildfires using MOPITT CO data, Geophysical Research Letters, 32, https://doi.org/10.1029/2005GL022995, http://dx.doi.org/10.1029/2005GL022995, l11809, 2005.

10 Pommier, M., McLinden, C. A., and Deeter, M.: Relative changes in CO emissions over megacities based on observations from space, Geophysical Research Letters, 40, 3766–3771, https://doi.org/10.1002/grl.50704, http://dx.doi.org/10.1002/grl.50704, 2013.

Randerson, J., van der Werf, G., Giglio, L., Collatz, G., and Kasibhatla, P.: Global Fire Emissions Database, Version 4.1 (GFEDv4), https://doi.org/10.3334/ornldaac/1293, https://daac.ornl.gov/cgi-bin/dsviewer.pl?ds_id=1293, 2017.

Rinsland, C. P., Luo, M., Logan, J. A., Beer, R., Worden, H., Kulawik, S. S., Rider, D., Osterman, G., Gunson, M., Eldering, A., Goldman,
15 A., Shephard, M., Clough, S. A., Rodgers, C., Lampel, M., and Chiou, L.: Nadir measurements of carbon monoxide distributions by the Tropospheric Emission Spectrometer instrument onboard the Aura Spacecraft: Overview of analysis approach and examples of initial results, Geophysical Research Letters, 33, https://doi.org/10.1029/2006gl027000, https://doi.org/10.1029%2F2006gl027000, 2006.

Rodgers, C. D.: Inverse methods for atmospheric sounding : theory and practice, vol. 2 of *Series on atmospheric, oceanic and planetary physics*, World Scientific, Singapore,, River Edge, N.J., réimpression : 2004, 2008, 2000.

20 Schneider, A., Friedl, M. A., and Potere, D.: A new map of global urban extent from MODIS satellite data, Environmental Research Letters, 4, 044 003, http://stacks.iop.org/1748-9326/4/i=4/a=044003, 2009.

Spivakovsky, C. M., Logan, J. A., Montzka, S. A., Balkanski, Y. J., Foreman-Fowler, M., Jones, D. B. A., Horowitz, L. W., Fusco, A. C., Brenninkmeijer, C. A. M., Prather, M. J., Wofsy, S. C., and McElroy, M. B.: Three-dimensional climatological distribution of tropospheric OH: Update and evaluation, Journal of Geophysical Research: Atmospheres, 105, 8931–8980, https://doi.org/10.1029/1999jd901006,
25 https://doi.org/10.1029%2F1999jd901006, 2000.

Turquety, S., Hadji-Lazaro, J., Clerbaux, C., Hauglustaine, D. A., Clough, S. A., Cassé, V., Schlüssel, P., and Mégie, G.: Operational trace gas retrieval algorithm for the Infrared Atmospheric Sounding Interferometer, Journal of Geophysical Research: Atmospheres, 109, n/a–n/a, https://doi.org/10.1029/2004jd004821, https://doi.org/10.1029%2F2004jd004821, 2004.

Veefkind, J., Aben, I., McMullan, K., Förster, H., de Vries, J., Otter, G., Claas, J., Eskes, H., de Haan, J., Kleipool, Q., van Weele, M.,
30 Hasekamp, O., Hoogeveen, R., Landgraf, J., Snel, R., Tol, P., Ingmann, P., Voors, R., Kruizinga, B., Vink, R., Visser, H., and Levelt, P.: TROPOMI on the ESA Sentinel-5 Precursor: A GMES mission for global observations of the atmospheric composition for climate, air quality and ozone layer applications, Remote Sensing of Environment, 120, 70–83, https://doi.org/10.1016/j.rse.2011.09.027, https://doi.org/10.1016%2Fj.rse.2011.09.027, 2012.

Vidot, J., Landgraf, J., Hasekamp, O., Butz, A., Galli, A., Tol, P., and Aben, I.: Carbon monoxide from shortwave infrared reflectance
35 measurements: A new retrieval approach for clear sky and partially cloudy atmospheres, Remote Sensing of Environment, 120, 255–266, https://doi.org/10.1016/j.rse.2011.09.032, https://doi.org/10.1016%2Fj.rse.2011.09.032, 2012.

Williams, J. E., van Velthoven, P. F. J., and Brenninkmeijer, C. A. M.: Quantifying the uncertainty in simulating global tropospheric composition due to the variability in global emission estimates of Biogenic Volatile Organic Compounds, Atmospheric Chemistry and Physics, 13, 2857–2891, https://doi.org/10.5194/acp-13-2857-2013, https://doi.org/10.5194%2Facp-13-2857-2013, 2013.

Williams, J. E., Bras, G. L., Kukui, A., Ziereis, H., and Brenninkmeijer, C. A. M.: The impact of the chemical production of methyl nitrate from the NO CH_3O_2 reaction on the global distributions of alkyl nitrates, nitrogen oxides and tropospheric ozone: a global modelling study, Atmospheric Chemistry and Physics, 14, 2363–2382, https://doi.org/10.5194/acp-14-2363-2014, https://doi.org/10.5194%2Facp-14-2363-2014, 2014.

Yurganov, L. N., Duchatelet, P., Dzhola, A. V., Edwards, D. P., Hase, F., Kramer, I., Mahieu, E., Mellqvist, J., Notholt, J., Novelli, P. C., Rockmann, A., Scheel, H. E., Schneider, M., Schulz, A., Strandberg, A., Sussmann, R., Tanimoto, H., Velazco, V., Drummond, J. R., and Gille, J. C.: Increased Northern Hemispheric carbon monoxide burden in the troposphere in 2002 and 2003 detected from the ground and from space, Atmospheric Chemistry and Physics, 5, 563–573, https://doi.org/10.5194/acp-5-563-2005, https://doi.org/10.5194%2Facp-5-563-2005, 2005.
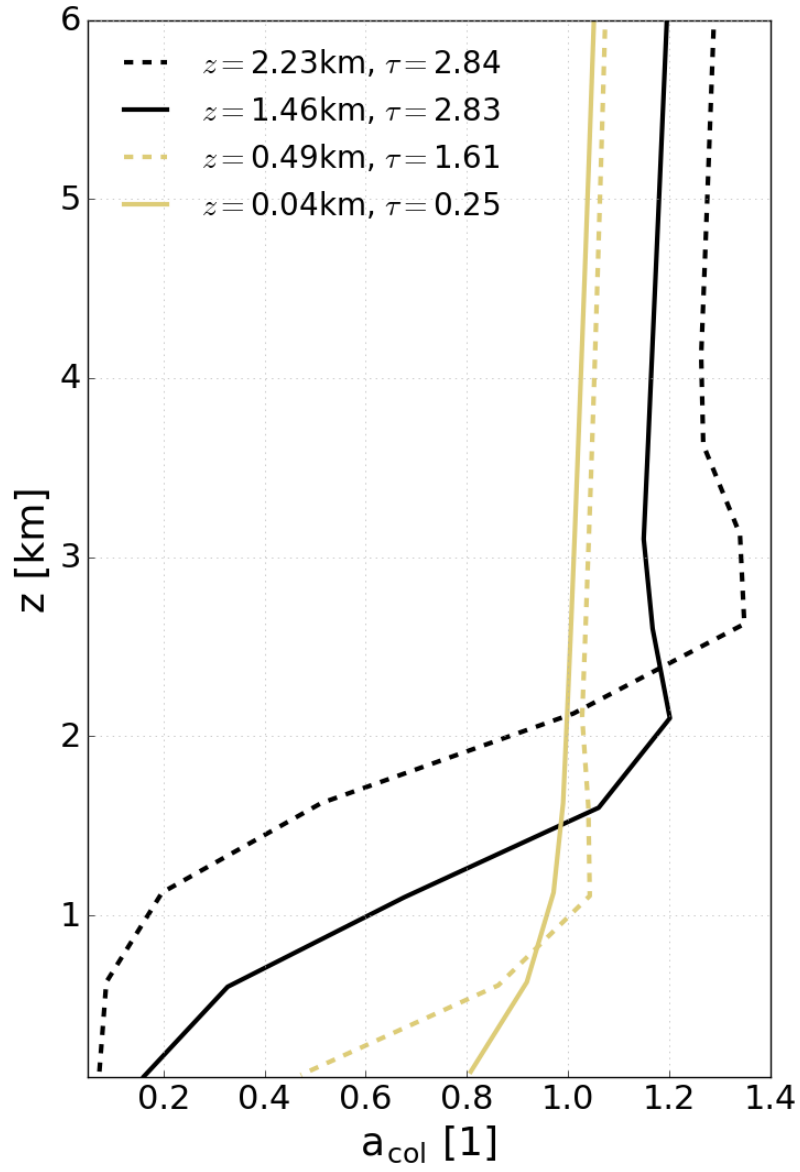
Figure 1. SCIAMACHY CO total column averaging kernels for different cloud centre heights (z_{cld}) and cloud optical thicknesses (τ_{cld}). Here, the solid yellow line is representative for clear-sky conditions. The figure shows typical cases of the vertical retrieval sensitivity of the SCIAMACHY CO retrievals over Paris.
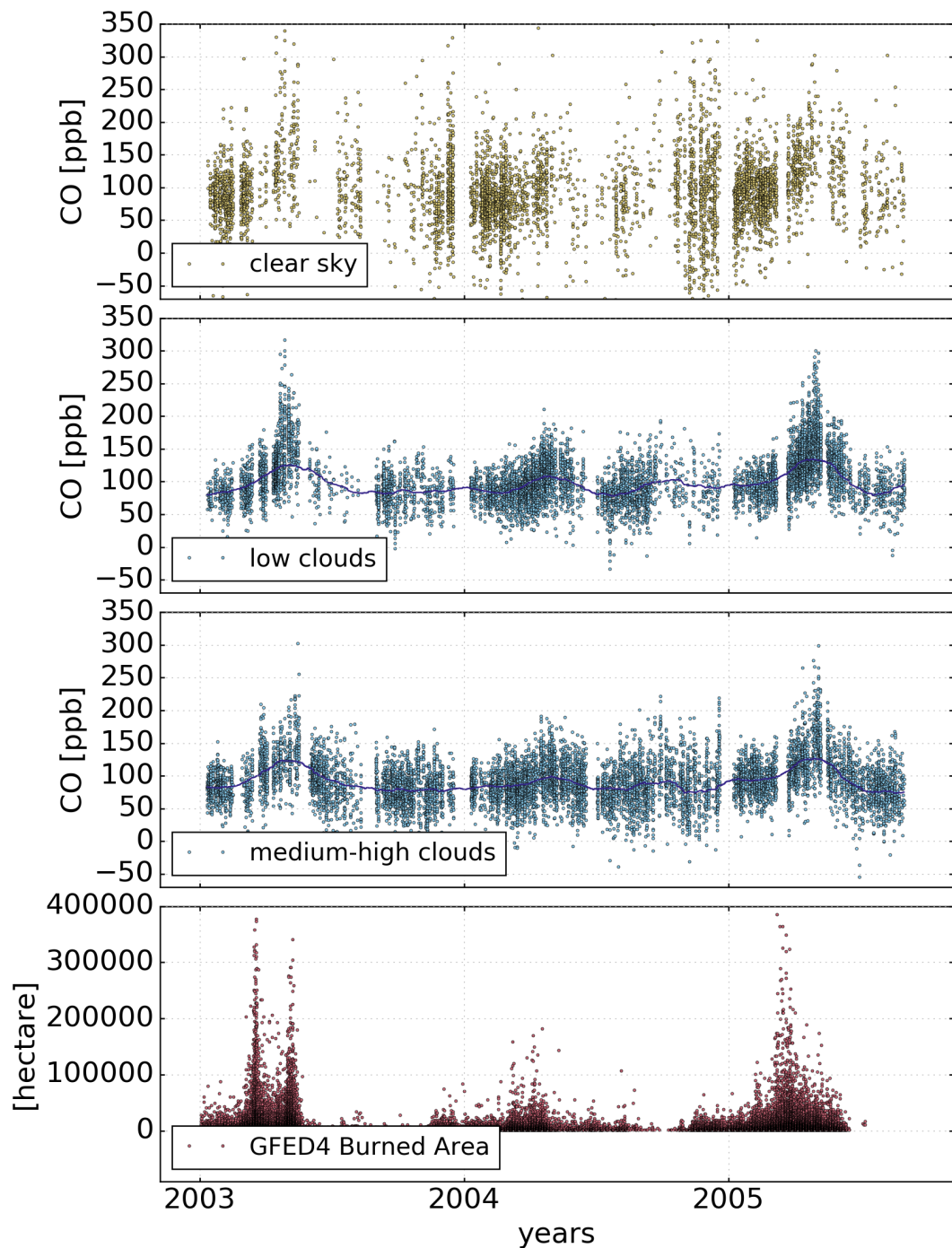
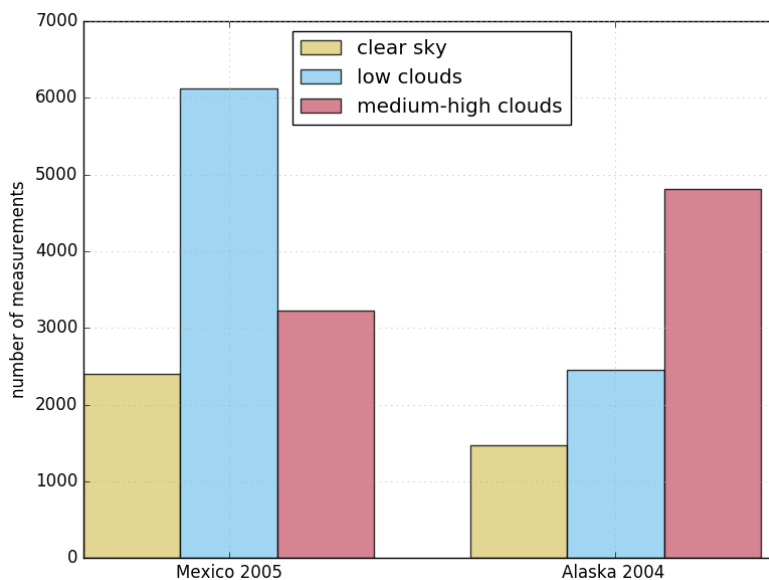**Figure 2.** Individual SCIAMACHY CO retrievals under clear-sky, low cloud, and medium-high cloud atmospheric conditions as well as daily GFED4 Burned Area over Mexico/Guatemala in the latitude/longitude box [(22.5°N,100.0°W), (10.0°N,80.0°W)]. The blue line is a running median with half width of 30 days.
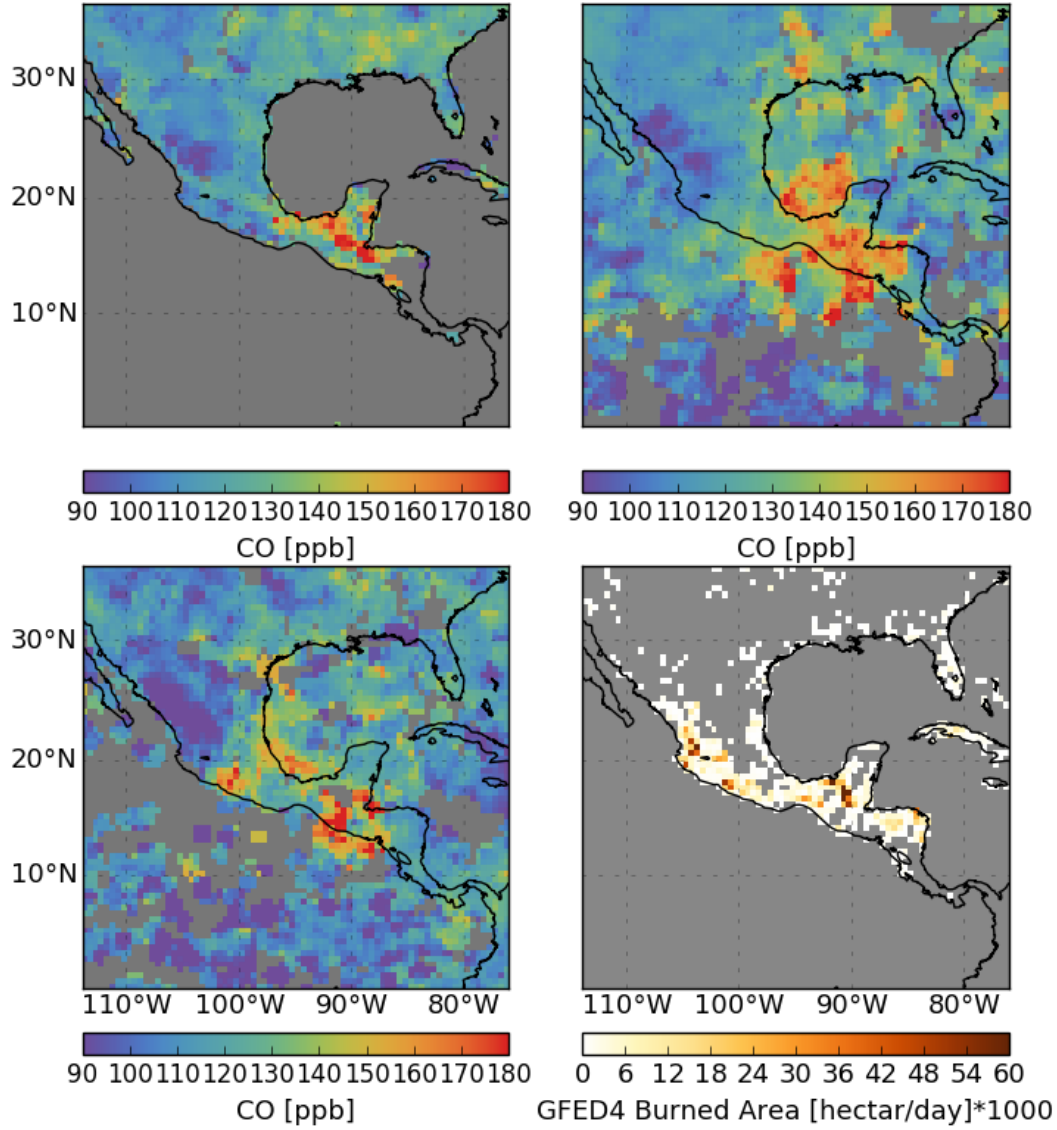
Figure 3. SCIAMACHY CO retrievals under clear-sky (top left), low cloud (top right), medium-high cloud (bottom left) atmospheric condition as well as daily GFED4 Burned Area (bottom right) averaged from 15th March to 15th May 2005 over Mexico and Central America. The resolution of the plot is 0.5 degree in latitude and longitude and the data is oversampled using a radius of 90km.
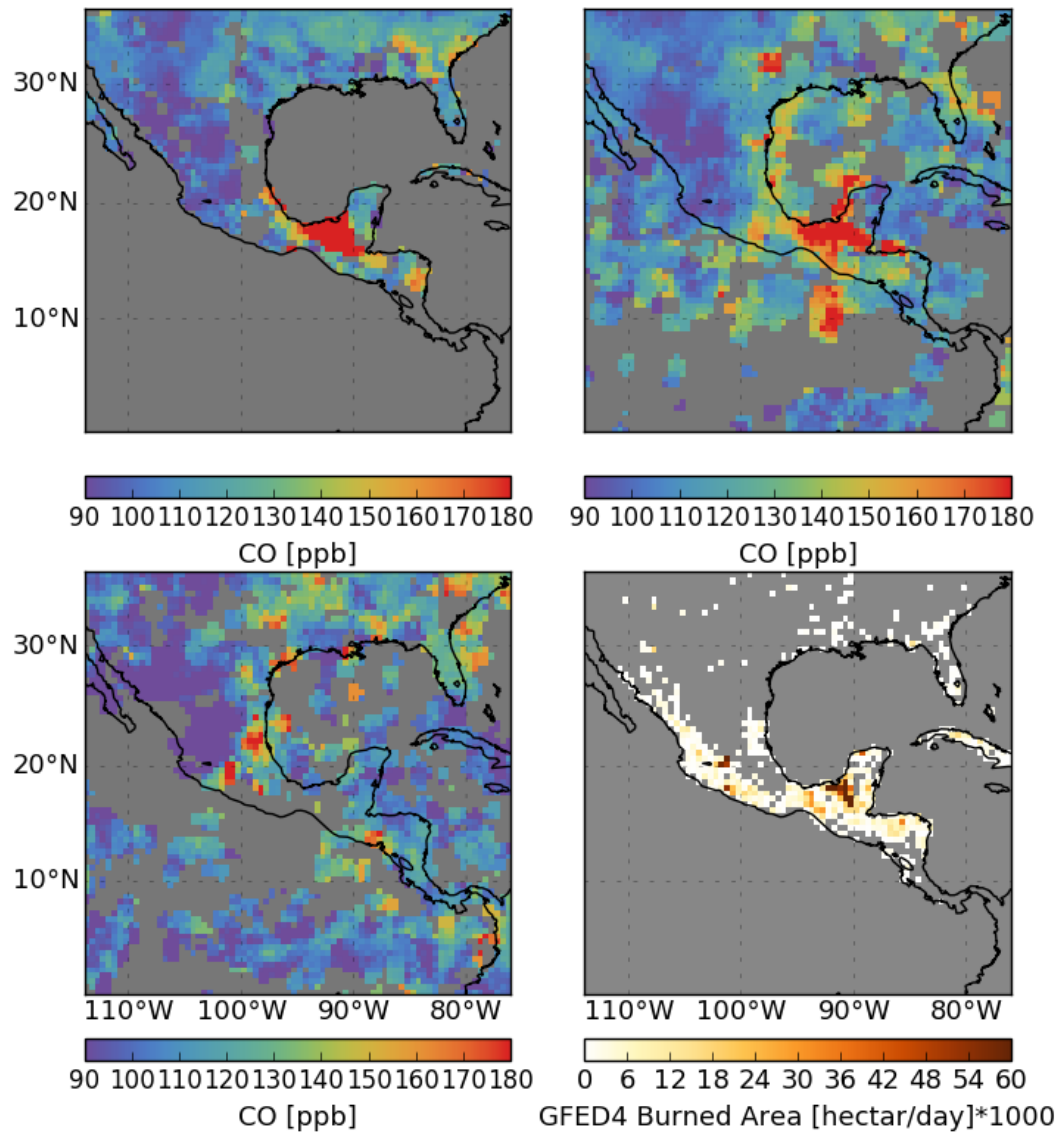
Figure 4. Same as Fig. 3 but the data is averaged from 1st April to 15th May 2003.

18
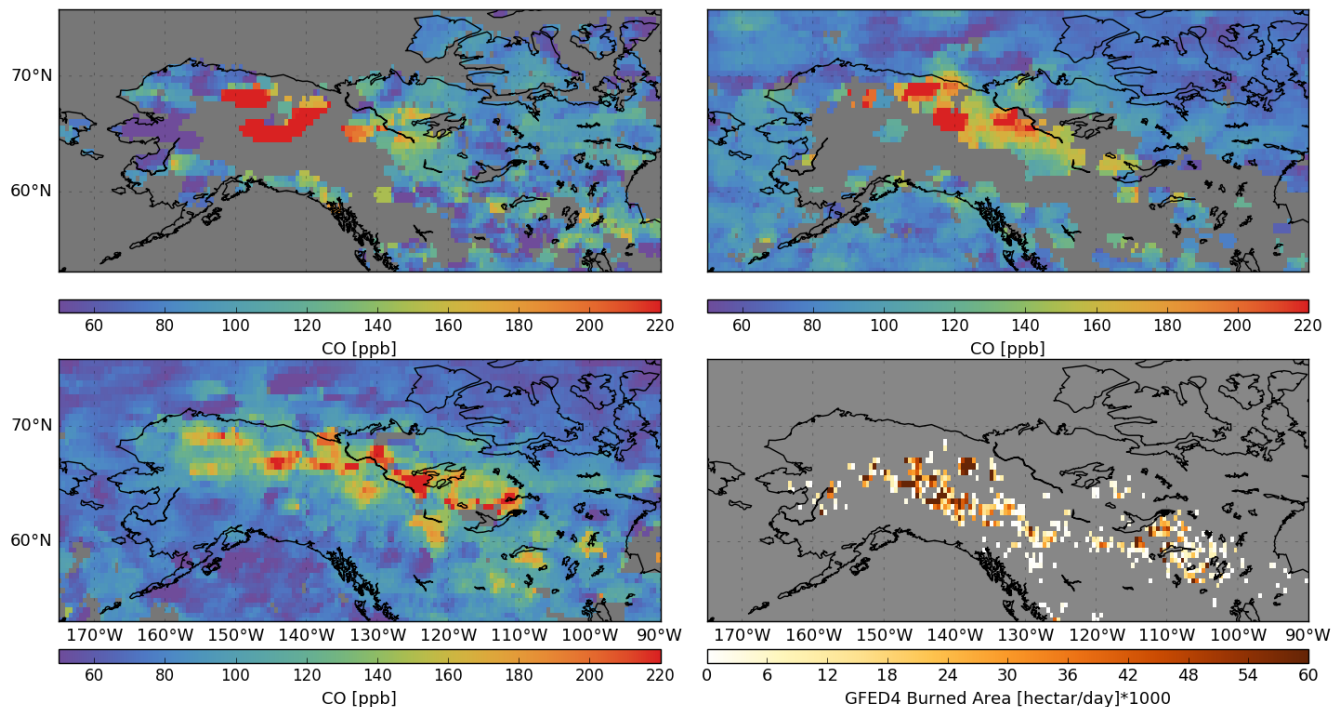
Figure 5. Same as Fig. 3 but for Alaska/Canada. The data is averaged from 1st July to 1st August 2004.

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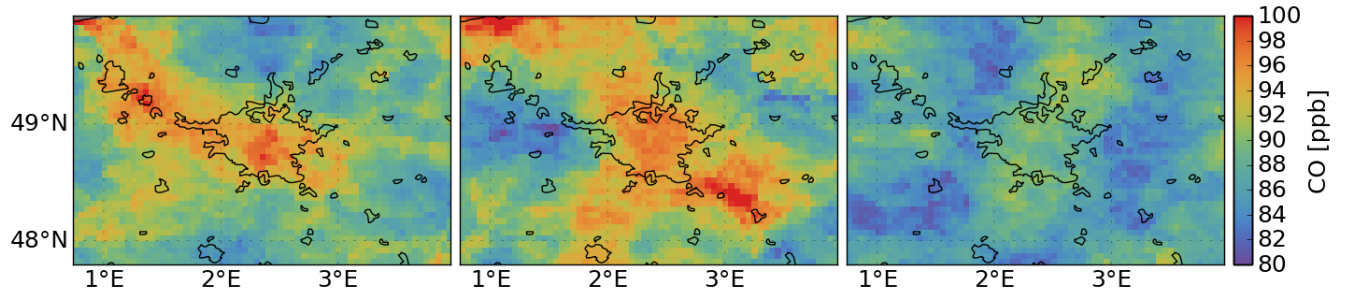
Figure 7. SCIAMACHY CO column mixing ratio averaged from January 2003 to April 2012 under clear-sky (left panel), low cloud (middle panel), and medium-high cloud (right panel) atmospheric conditions above Paris. The spatial sampling of the plot is $\delta = 0.05$ degree in latitude and longitude and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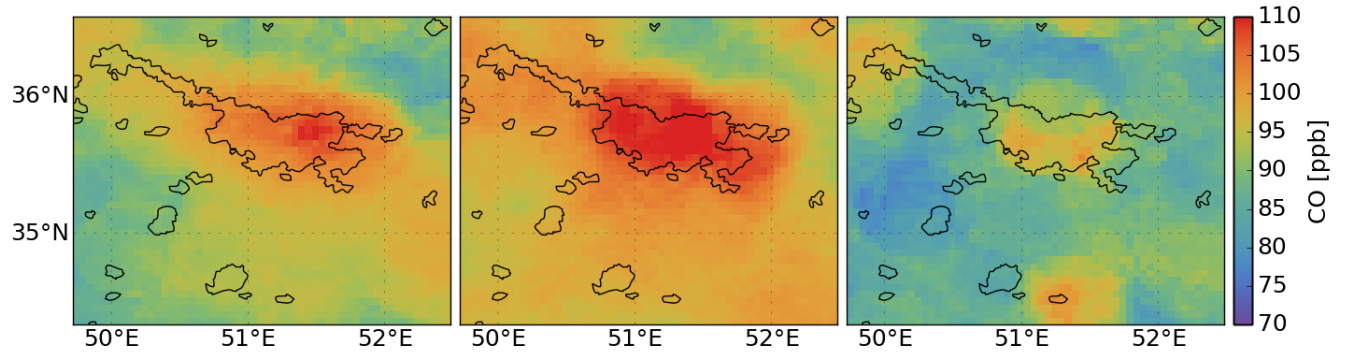

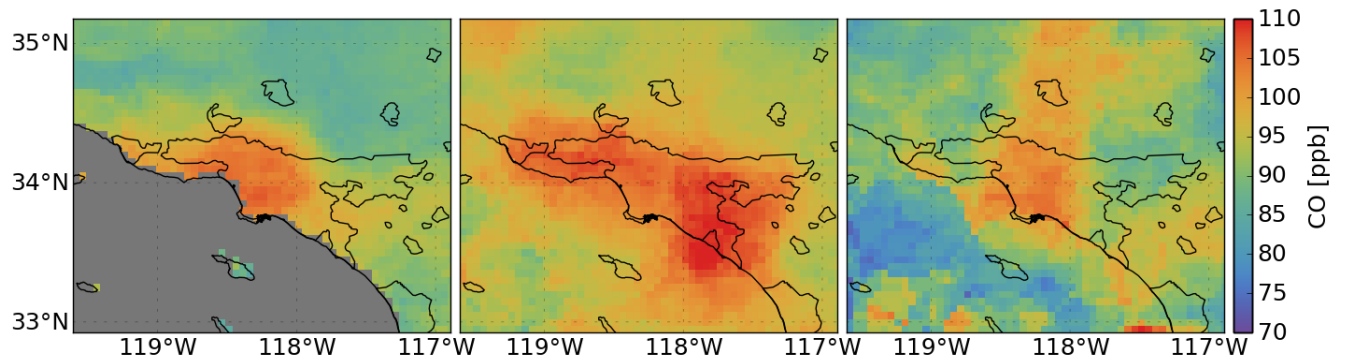**Figure 8.** Same as Fig. 7 but for Tehran.

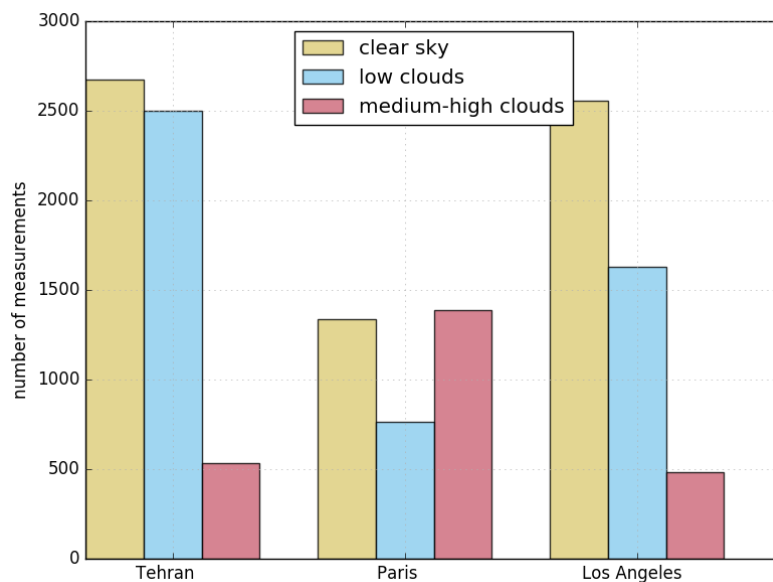**Figure 9.** Same as Fig. 7 but for Los Angeles.

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.
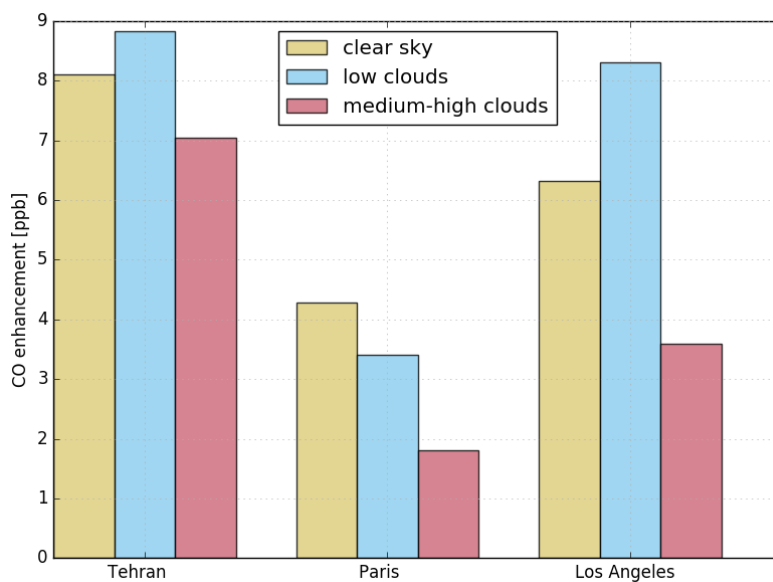

Figure 11. CO enhancement over cities shown in Fig. 7, 8, and 9 relative to the background concentration under clear-sky (yellow), low cloud (blue), and medium-high cloud (pink) atmospheric conditions. The difference of the median CO concentration inside and outside the urban area contours of each city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

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

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.

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

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