# CityZen

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## Analysis of SCIAMACHY greenhouse gas retrievals over CityZen target regions

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# Analysis of SCIAMACHY greenhouse gas retrievals over CityZen target regions

## Abstract

IUP-UB's responsibility within Task 2.1 is to provide consistent time series of the two most important anthropogenic greenhouse gases $CO_2$ and $CH_4$ on the global scale derived from SCIAMACHY/ENVISAT nadir observations. To achieve this IUP-UB has improved the WFM-DOAS algorithm, which enables to retrieve column-averaged mole fractions of $CO_2$ and $CH_4$, denoted $XCO_2$ and $XCH_4$, and has used this algorithm for processing seven years of SCIAMACHY data (2003-2009). The data products have been compared with global models, gridded population density and emission data bases. While reasonable to good agreement with global models concerning large-scale features such as hemispheric annual increase and seasonal cycle has been found, correlation with population density and anthropogenic emissions at city scale ($0.5^o$ x $0.5^o$; ~50 km) is poor; the linear correlation coefficient is typically positive but less than 0.3 even for 7-year averages. In other words, elevated concentrations of $CO_2$ and/or $CH_4$ for individual cities, including megacities such as Mexico City, can typically not be detected. This finding is not unexpected. Due to the long lifetime of the two gases elevated concentrations are not restricted to areas close to the source and even a strong source is expected to typically only produce a very small (sub-percent) enhancement relative to the (variable) background concentration. In addition, strong non urban sources need to be considered, especially for methane, e.g., emissions from near-by wetlands. The expected (mega)city emission signal is typically much less than the noise level and detection requires averaging many individual measurements and (nearly) bias free retrievals. Due to the strict quality filtering of the SCIAMACHY retrievals, typically only a few hundred measurements per $0.5^o$ x $0.5^o$ grid cell are available for averaging even for a multi-year data set. Possible biases of the satellite retrievals, e.g. due to aerosols and residual clouds, may also contribute to the finding that elevated concentrations over individual cities can typically not be detected. As a result, the regional emission signal is typically below the SCIAMACHY detection limit. It was therefore not possible to derive trends and perform quantitative comparisons with bottom-up emission data as originally planned for this project.

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# 1. Greenhouse gas retrievals from SCIAMACHY

Within this project the SCIAMACHY $XCO_2$ and $XCH_4$ retrieval algorithm WFM-DOAS has been further improved and used to process all available SCIAMACHY nadir spectra for the time period 2003-2009. The resulting data products have been compared with global models and maps of population density and anthropogenic emissions. In the following this is explained and illustrated starting with the retrieval algorithm improvements. The findings are shortly summarized at the end.

## 1.1.Retrieval algorithm WFM-DOAS

The SCIAMACHY $XCO_2$ and $XCH_4$ CityZen data set covering 2003-2009 has been generated with version 2.0 of the WFM-DOAS retrieval algorithm (WFMDv2.0). The previous version of this algorithm, WFMDv1.0, is described in detail in Schneising et al., 2008, for $XCO_2$, and Schneising et al., 2009, for $XCH_4$. In the following a short overview about the algorithm is given including an overview about the major improvements of WFMDv2.0 compared to WFMFv1.0.

The retrieval of a long-lived and therefore relatively well-mixed gas such as carbon dioxide or methane is challenging as only the small variations on top of a large background yield information on its surface sources and sinks. Therefore, the retrieval algorithm has to meet the conflicting requirements of accuracy and speed in order to process the large amounts of data produced by SCIAMACHY. This is achieved by the Weighting Function Modified Differential Optical Absorption Spectroscopy (WFM-DOAS) retrieval technique developed at the University of Bremen for the retrieval of trace gases and optimised for $CO_2$, $CH_4$ and $O_2$ retrievals using a fast look-up table (LUT) scheme to avoid computationally expensive online radiative transfer calculations. WFM-DOAS is based on shifting or scaling pre-defined vertical profiles of atmospheric parameters (primarily trace gas concentrations and temperature). A linearized radiative transfer model is least-squares fitted to the observed spectra. A-priori information on the atmospheric composition is only used to get linearization point for the radiative transfer simulations but not to constrain the retrieval. The algorithm has been described in detail elsewhere (Schneising et al., 2008, 2009). We therefore focus on a discussion of the main differences between the current version 2.0, which has been used to generate the data set discussed here, and the previous version 1.0 (Schneising et al., 2008, 2009).

*Retrieval of carbon dioxide mole fractions:*

In order to convert the retrieved $CO_2$ columns into column averaged mole fractions the $CO_2$ columns are divided by the dry-air columns obtained from the simultaneously measured $O_2$ columns obtained from the $O_2$ A-band. The specific changes implemented for WFMDv2.0 compared to WFMDv1.0 (Schneising et al., 2008) are:

- Use of spectra with improved calibration: Level 1 version 6 instead of version 5.
- Use of improved spectroscopy: HITRAN 2008 (Rothman et al., 2009) instead of HITRAN 2004.
- Modified static detector pixel mask.

- Use of an improved look-up table scheme, i.e., use of an extended set of surface elevations (0 km, 1 km, 2 km, 3 km, 4 km, 5 km) and albedos (0.01, 0.03, 0.1, 0.3, 0.6,1.0) covering all naturally occurring surface types by means of interpolation.
- Use of a more realistic standard aerosol scenario in the forward model (OPAC background scenario described in Schneising et al. (2008, 2009)).
- Processing of a longer time series: 2003–2009 instead of 2003–2005.
- Use of SCIAMACHY Absorbing Aerosol Index v4.1 (Tilstra et al., 2007) instead of EarthProbe/TOMS AAI (Herman et al., 1997) to filter strongly aerosol contaminated scenes, in particular desert dust storms.
- Change of full width at half maximum (FWHM) used for the $O_2$ reference spectra calculation from 0.45 nm to 0.44 nm because of the associated improvement of fit quality.

*Retrieval of methane mole fractions:*

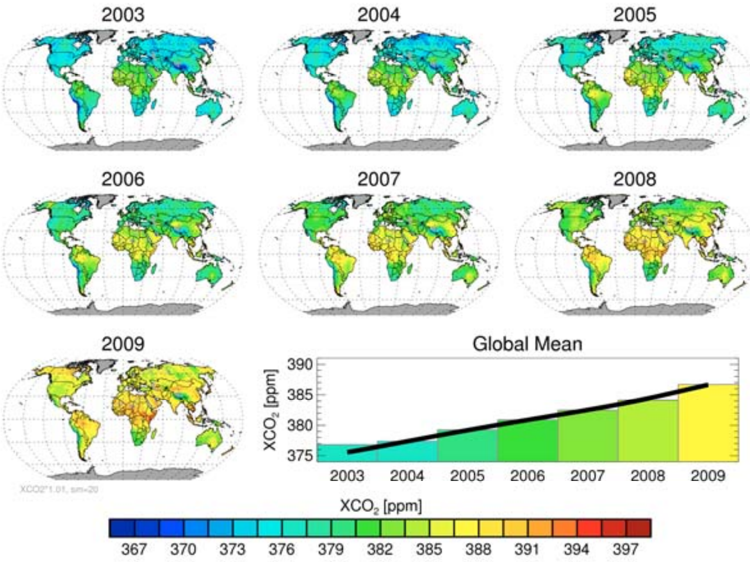
In order to convert the retrieved $CH_4$ columns into column averaged mole fractions the $CH_4$ columns are divided by the dry-air columns obtained from the simultaneously measured $CO_2$ columns correcting for $CO_2$ variability using CarbonTracker. The specific changes implemented for WFMDv2.0 compared to WFMDv1.0 (Schneising et al., 2009) are:

- Use of spectra with improved calibration: Level 1 version 6 instead of version 5.
- Use of improved spectroscopy: HITRAN 2008 (Rothman et al., 2009) plus Jenouvrier et al. (2007) $H_2O$ update in the methane fitting window instead of HITRAN 2004.
- Modified static detector pixel masks for three different time periods to get best possible results for the early years and stability until end of 2009 at the same time.
- Use of an improved look-up table scheme, i.e., use of an extended set of surface elevations (0 km, 1 km, 2 km, 3 km, 4 km, 5 km) and albedos (0.01, 0.03, 0.1, 0.3, 0.6, 1.0) covering all naturally occurring surface types by means of interpolation.
- Use of a more realistic standard aerosol scenario in the forward model (OPAC background scenario described in Schneising et al. (2008, 2009)).
- Processing of a longer time series: 2003–2009 instead of 2003–2005.
- Optimisation of filter criteria.
- Use of CarbonTracker version 2009 instead of 2007 (Peters et al., 2007, 2010) to correct the retrieved methane mole fractions for $CO_2$ variability.

Due to proceeding detector degradation in the spectral range used for the methane column retrieval, static detector pixel masks for three different time periods are used to get best possible results for the early years and stability until end of 2009 at the same time (Periode 1: 2003-2004; period 2: 01/2005-10/2005; period 3: 11/2005-12/2009). Since November 2005 only one remaining detector pixel in the Q-branch of the methane band around 1670 nm, which is the spectral region in the fitting window where the strongest absorption occurs, is serviceable. Therefore, the retrieval results since November 2005 are expected to be of reduced quality with regard to noise compared to the prior time period where more Q-branch detector pixels are available. Because of the increase of dead or bad pixels, the filter criteria have to be changed for data after October 2005 (third period) because the availability of considerably less detector pixels automatically increases the fit error. Therefore the filter criterion on the $CH_4$ column fit error is relaxed for the third period to get a comparable amount of retrievals classified as good. The quality filtering for this period is additionally extended to restrict to land only scenes and to filter out cloud contaminated scenes with a threshold algorithm like in the case of $XCO_2$ based on sub-pixel information provided by the SCIAMACHY Polarisation Measurement Device (PMD) channel A covering the spectral region 310–365 nm in the UV.

**Figure 1** shows the resulting global WFMDv2.0 $XCO_2$ and $XCH_4$ maps for the seven years 2003 – 2009 and the annually averaged values of $XCO_2$ and $XCH_4$. As can be seen, $CO_2$ has been increasing almost linearly in the considered time period whereas $CH_4$ was nearly constant except for recent years, where an increase can be observed.

**SCIAMACHY Carbon Dioxide**



**SCIAMACHY Methane**

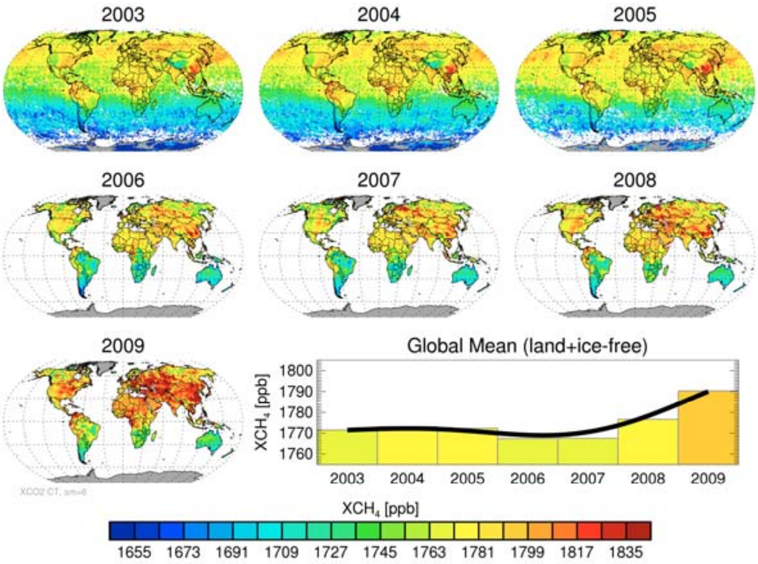


**Figure 1:** Top: Annual composite averages and global mean values of SCIAMACHY WFMDv2.0 $XCO_2$. Bottom: As top panel but for SCIAMACHY WFMDv2.0 $XCH_4$. Note that no data are shown for $XCH_4$ over water for 2006-2009 due to large noise caused by detector degradation.

## 1.2. Comparison with global models

### 1.2.1. Comparison of SCIAMACHY WFMDv2.0 $XCO_2$ with NOAA's CarbonTracker

To examine the $CO_2$ increase with time and the seasonal cycle more quantitatively the SCIAMACHY data are compared to the CarbonTracker release 2009 assimilation system (Peters et al., 2007, 2010) based on monthly data. The Carbon-Tracker $XCO_2$ fields as used for this study have been sampled in space and time as the SCIAMACHY satellite instrument measures. The SCIAMACHY altitude sensitivity has been taken into account by applying the SCIAMACHY $CO_2$ column averaging kernels to the CarbonTracker $CO_2$ vertical profiles. For comparisons with CarbonTracker the SCIAMACHY $CO_2$ data have been scaled by a constant factor of 1.01 to compensate a small systematic bias between the two data sets. As CarbonTracker data are only available up to 2008, the modelled column-averaged mole fractions for 2009 are obtained from the 2008 values by adding 2 ppm uniformly. As can be seen in **Figure 2**, the continuous increase with time on both hemispheres is consistent with CarbonTracker. However, the annual mean increase is somewhat smaller than in the model simulations, namely 1.5 ppm/yr compared to 2.0 ppm/yr. Results of the first half of 2003 are not shown in this figure due to changes of the instrument settings, e.g., detector temperatures and field of view (shift of nadir centre position and change of total scan width), during the first third of 2003 potentially introducing small offsets in the data adversely affecting this particular quantitative analysis with impact until midyear because of the smoothing window used.

For the northern hemisphere we also find good agreement of the phase of the $CO_2$ seasonal cycle with the model resulting in a pronounced correlation of the two data sets (r=0.94). In contrast to the northern hemisphere, systematic phase differences are observed on the southern hemisphere leading to a somewhat smaller correlation (r=0.84) which is, nevertheless, quite high due to the observed increase with time in both data sets. The discrepancy of the phases on the southern hemisphere can probably be ascribed to a large extent to the higher weight on ground scenes with occurrences of subvisual thin cirrus clouds (induced by the restriction to land and the smaller land fraction compared to the northern hemisphere) which are not explicitly considered in WFM-DOAS yet and are hence a potential error source leading to a possible overestimation of the carbon dioxide mole fractions for scenes with high subvisual cirrus fraction (Schneising et al., 2008). This could also be an explanation for the presumably unrealistic large cycle of the carbon dioxide growth rate on the southern hemisphere. First promising results for synthetic data obtained with a different approach which is based on (very time-consuming) online radiative transfer calculations demonstrate that accurate retrievals are also possible in the presence of thin cirrus clouds by taking selected cloud parameters specifically into account (Reuter et al., 2010). This indicates that cirrus can in principle also be considered explicitly in the future in a look-up table approach, even though it is not trivial to achieve sufficient processing speed for a global long-term analysis like the one presented in this manuscript. Although cloud parameters are not included in the state vector of the current WFM-DOAS version, the influence of subvisual cirrus on the retrievals is minimised resulting in much better agreement with CarbonTracker compared to the previous WFM-DOAS version especially on the southern hemisphere. The achieved reduction of the amplitude of the seasonal cycle is presumably a consequence of the interaction of the more realistic default aerosol scenario, improved spectroscopy, and the change-over to the SCIAMACHY Absorbing Aerosol Index. We find that for both hemispheres the mean amplitude of the retrieved seasonal cycle is about 1 ppm larger than for CarbonTracker deriving 4–5 ppm for the northern hemisphere and 1–2 ppm for the southern hemisphere from the SCIAMACHY data. The less pronounced seasonal cycle of CarbonTracker compared to the satellite data might be explainable to some extent by a too low net ecosystem exchange (NEE) between the atmosphere and the terrestrial biosphere; for example Yang et al. (2007) estimate that NEE in the northern hemisphere is about 25% larger than predicted by the CASA (Carnegie-Ames Stanford Approach) biogeochemical model which is also used in CarbonTracker. This underestimation is of the same order of magnitude as the scaling needed to fit the amplitude of the northern hemispheric seasonal cycle of CarbonTracker to SCIAMACHY.

According to NOAA/ESRL (http://www.esrl.noaa.gov/gmd/ccgg/trends/ ) the average surface growth rate for the considered time period is 1.92 ppm/yr with an estimated uncertainty in the global annual mean growth rate of 0.07 ppm/yr. Due to uncertainties in the transport, the application of the SCIAMACHY averaging kernels, and the consideration of the SCIAMACHY sampling in space and time, these surface results cannot be translated directly to the CarbonTracker column-averaged results used here leading to larger uncertainties of CarbonTracker and potentially to differing absolute values of the growth rate compared to the surface data. However, the assimilation of the surface data suggests that the actual uncertainty of the used CarbonTracker data is smaller than the stated standard deviation because the variability is assumed to be dominated by natural variations. Therefore, the mean annual growth rate derived from the SCIAMACHY data, which is systematically smaller than for CarbonTracker, possibly shows a low bias of a few tenths of 1 ppm. A low bias of this magnitude is potentially explainable in large part by known instrumental issues: SCIAMACHY light path monitoring results report small throughput losses which are slowly increasing with time in the spectral regions used for the $CO_2$ (channel 6) and $O_2$ (channel 4) column retrievals. This continuous decrease of the average detector signal is somewhat more pronounced in channel 6. Assuming that a throughput loss is linked to small low biases in the retrieved column growth rates this results in an underestimation of the $XCO_2$ growth rate. A first analysis with real data based on small subsets in 2003 and 2008 indicates that the additional consideration of the recently released complete consolidated SCIAMACHY m-factor data base in the calibration which was designed to compensate this degradation effect would actually increase the $XCO_2$ growth rate by up to 0.2–0.3 ppm/yr resulting in a total growth rate of about 1.7–1.8 ppm/yr. The improved calibration will be considered in future versions. The mean amplitude of the seasonal cycle is obtained by subtracting the deseasonalised trend from the smoothed curve and averaging the resulting amplitudes for all available years denoting the standard deviation as error. As can be seen, the amplitude of the seasonal cycle of the SCIAMACHY data is increasing on the northern hemisphere and decreasing on the southern hemisphere with higher latitudes in good qualitative agreement with CarbonTracker. Quantitatively, the differences of the amplitude values of the mid and high-latitudes ($30^o$–$90^o$) and the whole hemisphere are consistent for the SCIAMACHY and CarbonTracker data amounting to about 0.6 ppm for the northern and -0.3 ppm for the southern hemisphere. The absolute differences in the amplitudes of the retrieved and modelled seasonal cycle are on the order of 1–1.5 ppm for all specified zonal averages.

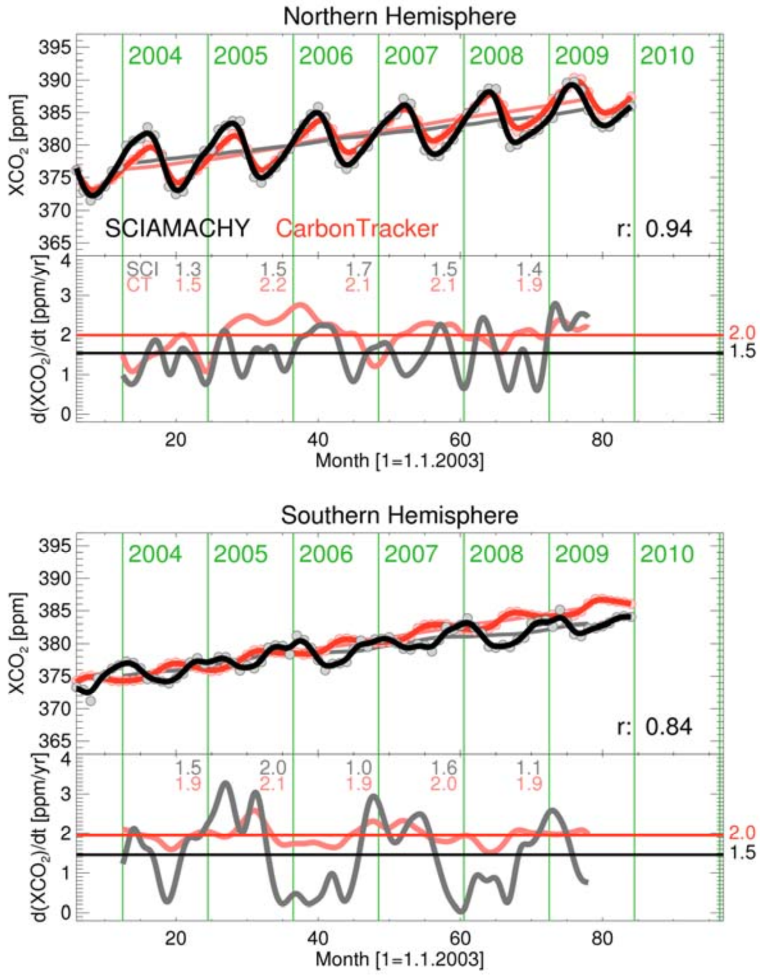


**Figure 2:** Comparison of the SCIAMACHY (black) and CarbonTracker (red) $XCO_2$ for the northern hemisphere (top) and the southern hemisphere (bottom) based on monthly means (coloured circles). The saturated solid lines have been smoothed using a four-month Hann window (which has a similar frequency response to a two-month boxcar filter but better attenuation of high frequencies). The pale solid lines represent the corresponding deseasonalised trends. Shown below are the derivatives of these deseasonalised curves corresponding to the current increase at the considered point in time. Also noted are yearly mean values of the derivatives (in pale colours) as well as the mean value of the whole time period on the right hand side.

### 1.2.1. Comparison of SCIAMACHY WFMDv2.0 $XCH_4$ with JRC's TM5 model

To examine this renewed methane increase in recent years more quantitatively **Figure 3** shows the temporal evolution of retrieved SCIAMACHY methane based on monthly means as well as the corresponding deseasonalised trend and its derivative for both hemispheres analogue to **Figure 2** for carbon dioxide. Larger scatter since November 2005 due to the lesser number of usable detector pixels is also observed in the monthly data. This is in particular true for the southern hemisphere where the stricter filter criteria accompanying the pixel mask change furthermore lead to a considerable less amount of measurements available for the computation of the monthly means because of the additional exclusion of scenes over oceans and the relative small land mass fraction. Nevertheless, the renewed methane growth is visible in **Figure 3** for both hemispheres. The anomaly since 2007 is derived from the difference of the mean values of the derivative of the deseasonalised trend after and before middle of 2006. To avoid a possible distortion of the analysed curves due to potentially introduced systematic regional biases caused by the alteration of the detector pixel mask at the end of October 2005, all values over a period of plus or minus 6 months from this date are not considered in the calculation of the mean value before 2007. It would be desirable to have model simulations for the whole time period 2003–2009 for a quantitative comparison of the respective anomalies. Here we use Scenario S1 of Bergamaschi et al. (2007) (based on the NOAA surface measurements) of the TM5 model. This TM5 model data set is currently only available for 2003. The corresponding values are repeated every year to study at least the seasonal behaviour. The TM5 fields as used for this study have been sampled in space and time as the SCIAMACHY satellite instrument measures. The SCIAMACHY altitude sensitivity has been taken into account by applying the SCIAMACHY $CH_4$ column averaging kernels to the model vertical profiles. Of course, the expected anomaly since 2007 for this yearly repeated model is zero. Nevertheless, a deviation from zero is a measure of the impact of sampling in particular due to the different filter criteria after October 2005; e.g., the restriction to land leads to a smaller seasonal cycle for the southern hemisphere being visible in both the satellite and model data. Hence, the anomaly of the model is a lower bound of the error of the observed SCIAMACHY anomaly. Neglecting other possible error sources the anomaly of SCIAMACHY $XCH_4$ amounts to 8.5 +/- 0.5 ppb/yr for the northern hemisphere and 5.3 +/- 0.7 ppb/yr for the southern hemisphere. The contribution of the restriction to land after October 2005 to the sampling error of the anomalies is smaller than 0.2 ppb/yr as estimated by additionally restricting to land for the whole time series and comparing the respective anomalies to the previously derived values. Hence, the potential error of the anomalies induced by using land and ocean scenes before and land only scenes after the end of October 2005 is small. This is a reasonable result because the amount of available data over the ocean is comparatively thin, anyway. To assess the expected absolute values and temporal evolution of the increase at least approximately, the SCIAMACHY data are also compared to a mean of surface measurements from the NOAA/ESRL network for the respective hemisphere using the 25 surface stations including all available stations providing monthly data without gaps for the period 2003–2008 and not being obviously influenced by local sources. Surface data for 2009 are not available yet. For the southern hemisphere two out of four Antarctic stations are also excluded in order to avoid giving too much weight on this region which is not sampled by SCIAMACHY data passing the quality filter after October 2005. As the phase of the seasonal cycle of methane can differ perspicuously for column-averaged and surface data, only the deseasonalised trends as well as the corresponding standard error (standard deviation divided by the square root of the number of used surface sites) for the hemispheric averages of the surface data are shown. The deseasonalised trends of the satellite and surface data are in reasonable to good agreement in particular for the northern hemisphere when considering the different absolute values of the data sets because surface data are always higher than column averaged data (note the different y-axes). The derived surface anomaly values of 10.1 ppb/yr for the northern hemisphere and 9.3 ppb/yr for the southern hemisphere are a coarse estimate of the magnitude of the expected hemispheric satellite anomalies. In conformity with expectations the actual corresponding SCIAMACHY values stated above are smaller than for the surface measurements. Additionally, the northern hemispheric anomalies for both data sets are higher than for the southern hemisphere further indicating that the values derived from the satellite data are consistent with the surface measurements and seem reasonable.

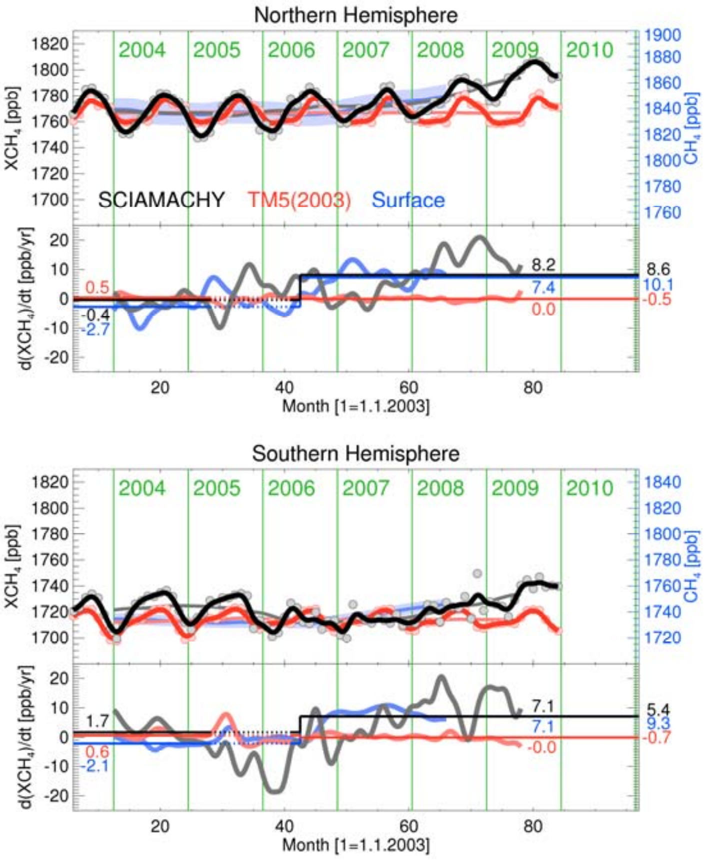


**Figure 3:** As **Figure 2** but for WFMDv2.0 XCH4. The anomaly (numbers on the right border) is defined as the difference of the mean values of the derivative of the deseasonalised trend after and before middle of 2006. Values one year around the detector pixel mask alteration end of October 2005 (indicated by the dotted lines) are not considered because systematic offsets due to the change are potentially distorting the deseasonalised trend (obtained by smoothing) and its derivative. As the TM5 model is only available for 2003, the corresponding values are repeated every year to study the amplitudes of the seasonal cycles and the impact of sampling on the observed anomaly because it is expected to be zero for the repeated model if there is no sampling influence. Hence, the anomaly of the model is a lower bound of the error of the observed SCIAMACHY anomaly. To assess the expected absolute values and temporal evolution of the increase at least approximately, the SCIAMACHY data are also compared to the deseasonalised trend of a mean of surface measurements from the NOAA/ESRL network for the respective hemisphere (blue) using 25 surface stations. Also shown is the corresponding standard deviation divided by the square root of the number of used surface sites in light blue.

## 1.3. Comparison with population density and emission maps

### 1.3.1. Previous results obtained with WFMDv1.0 (2003-2005)

Until now space-based measurements of greenhouse gases such as $CO_2$ and $CH_4$ have not yet been used to quantify anthropogenic emissions of mega cities or large urban areas.

The first potential detection of regionally elevated $CO_2$ over an anthropogenic source region has been reported in Schneising et al., 2008, using WFMDv1.0 retrievals for 2003-2005. The following has been written in the abstract of Schneising et al., 2008:

> “When averaging the SCIAMACHY $XCO_2$ over all three years (2003-2005) we find elevated $CO_2$ over the highly populated region of western central Germany and parts of the Netherlands (“Rhine-Main area”) reasonably well correlated with EDGAR anthropogenic $CO_2$ emissions. On average the regional enhancement is 2.7 ppm including an estimated contribution of 1-1.5 ppm due to aerosol related errors and sampling”.

The average year 2003-2005 SCIAMACHY WFMDv1.0 $XCO_2$ retrievals over the Rhine-Main area and along the US East Coast are shown in **Figure 4**. As can be seen, there is a reasonable agreement between the large scale features of the elevated $XCO_2$, shown in red and orange, and the large scale features of population density and anthropogenic $CO_2$ emissions.

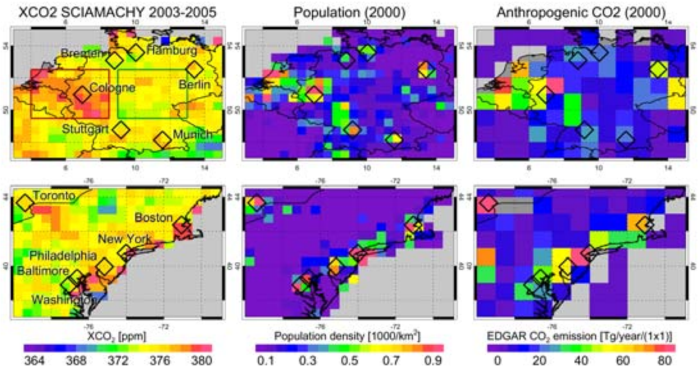


**Figure 4:** Left: SCIAMACHY WFMDv1.0 $XCO_2$ retrievals (year 2003-2005 average) over two anthropogenic source regions compared with population density (middle) and EDGAR anthropogenic $CO_2$ emissions. The gridded population density data were obtained from the Center for International Earth Science Information Network (CIESIN), Columbia University and Centro Internacional de Agricultura Tropical, available at http:://sedac.ciesin.columbia.edu/gwp. The anthropogenic $CO_2$ emissions have been obtained from the Emission Database for Global Atmospheric Research (EDGAR) (EDGAR 3.2 Fast Track 2000 (32FT2000), http://www.mnp.nl/edgar/). (Figure from: O. Schneising, PhD thesis, IUP, Univ. Bremen, Germany).

As discussed in Schneising et al., 2008, the expected $XCO_2$ enhancement over strong and extended anthropogenic source regions such as the Rhine-Main area, is on the order of 1-2 ppm. This is well below the single measurement retrieval precision of SCIAMACHY, which is about 2% (8 ppm). Assuming that the precision improves with the square root of the number of measurements added, more than 100 measurements have to be added to detect a 1 ppm $XCO_2$ enhancement. This is the typical number of SCIAMACHY “good” measurements passing the quality criteria per $0.5^o$x$0.5^o$ grid cell for Germany during 2003-2005. For the UK the number of “good” measurements per $0.5^o$x$0.5^o$ grid cell during 2003-2005 is typically 25 or less due to more frequent cloud cover. Therefore no clear correlation between the SCIAMACHY WFMDv1.0 $XCO_2$ retrievals and population density or anthropogenic $CO_2$ emissions over the UK has been found for the WFMDv1.0 2003-2005 $XCO_2$ data set (not shown here).

### 1.3.2. New results obtained with WFMDv2.0 (2003-2009)

The new SCIAMACHY WFMDv2.0 $XCO_2$ and $XCH_4$ data set covers seven years (2003-2009) compared to the three years covered by the previous WFMDv1.0 data set. As the precision (random error) improves with the number of measurements added one would expect that the correlation of the SCIAMACHY $XCO_2$ retrievals with population density improves if seven years of data are averaged as is possible now with the new WFMDv2.0 data set.

In the following results are shown for various regions relevant for CityZen. For each region multi-year averages of SCIAMACHY WFMDv2.0 $XCO_2$ and $XCH_4$ are shown and compared with population density and anthropogenic emissions.

For population density the CIESIN/CIAT gridded population of the world version 3 (GPWv3) data set for the year 2000 has been used (CIESIN/CIAT, 2005).

The anthropogenic $CO_2$ and $CH_4$ emissions have been obtained from the Emission Database for Global Atmospheric Research (EDGAR) version 4.0 (Source: EC-JRC/PBL, EDGAR version 4.0, http://edgar.jrc.ec.europa.eu/, 2009). Gridded data for the year 2005 have been used at $0.1^o\times0.1^o$(original $CO_2$ filename as downloaded: v40_CO2_2005_excl_organic_carbon.txt; original $CH_4$ filename as downloaded: v40_ch4_2005_all.txt). The EDGAR data have been re-gridded to $0.5^o\times0.5^o$ which is also the grid used for the SCIAMACHY data sets.

The basic idea for the presented analysis is the following: Even if the atmospheric lifetime of a gas emitted at the surface is very long and, therefore, the emitted gases can be transported over long distances, the concentrations are highest near the source. Because the expected enhancement over anthropogenic source regions such as cities is supposed to be very small (in comparison to the background concentration but also in comparison to the retrieval precision) several years of data have to be averaged to detect the small signal originating from the regional anthropogenic emissions. Therefore multi-year averages of the SCIAMACHY retrievals have been generated and compared to population density and emission maps. This has been done in order to find out if elevated GHG concentrations can be correlated with anthropogenic emission sources such as cities.

The SCIAMACHY $XCO_2$ and $XCH_4$ retrievals presented in the following are shown as regional anomalies. This means that for each regional map the corresponding mean value over the region has been subtracted. This has been done to remove the (variable) background concentration which permits to better focus on the spatial pattern over a given region. The spatial resolution of the maps is $0.5^o\times0.5^o$. Several $XCO_2$ and $XCH_4$ anomaly maps are shown per region: at least the year 2003-2005 average and the year 2003-2009 average. The best correlation with population density and anthropogenic emissions is expected to be present for the 2003-2009 data set because here the largest number of data have been averaged and, therefore, the noise should be lowest.

**Figure 5** shows the results for the CityZen region “BeNeLuxBig”. For $CO_2$ the spatial pattern for 2003-2005 is similar as has been obtained with the previous version WFMDv1.0. The $XCO_2$ is highest over the “Rhine-Main area” roughly corresponding to the region where also the EDGAR anthropogenic $CO_2$ emissions are highest. There is however no obvious correlation on the $0.5^o$x$0.5^o$ grid scale. Apparently SCIAMACHY cannot resolve the grid scale even if several years of data are averaged. Over the UK no correlation between atmospheric $XCO_2$ and $CO_2$ emissions is visible. This has already been found when investigating the WFMDv1.0 2003-2005 data set. This is very likely because of the very low number of “good” measurements over the UK due to more frequent cloud cover compared to the Rhine-Main area. When averaging all seven years (2003-2009) the largest $XCO_2$ is observed over a region which roughly corresponds to the region where also the anthropogenic $CO_2$ is highest but the correlation at the $0.5^o$x$0.5^o$ grid scale is only 0.16, i.e., close to zero. Over the UK the correlation does not improve for the 7 years average compared to the 3 years averages. The $XCO_2$ enhancement over the Rhine-Main area is about 1 ppm which is much less than the typical scatter of the $XCO_2$ over the entire BeNeLuxBig region which is about +/- 2 ppm as shown in the scatter plot (bottom middle panel).

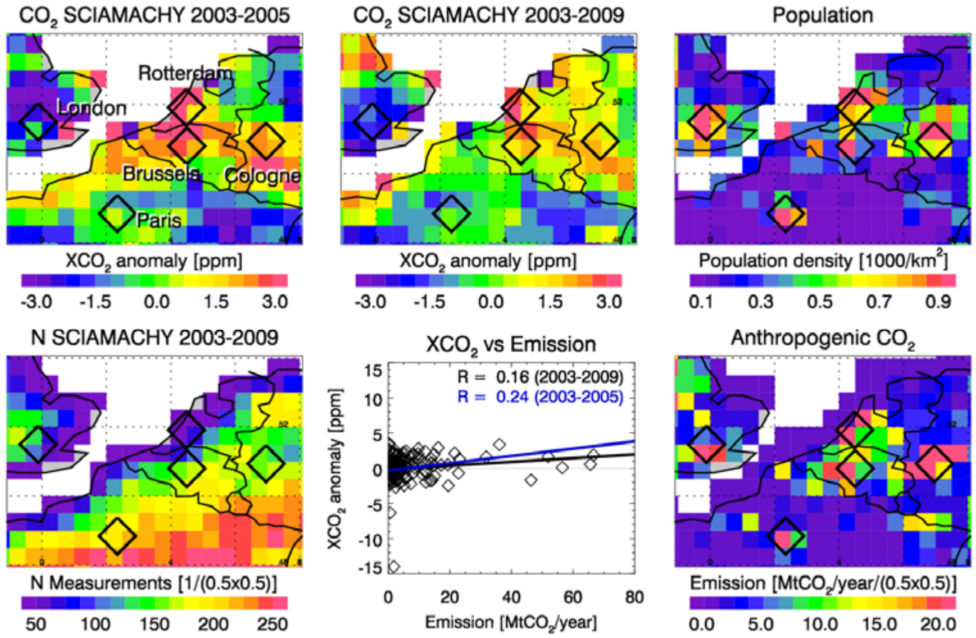
**Figure 5** also shows analogue results for $XCH_4$ over the BeNeLuxBig region. As can be seen, no clear correlation with population density or anthropogenic emission can be observed. This is likely due to the fact that cities or urban areas are less important methane sources compared to other strong non-urban area natural sources such as wetlands. It also has to be pointed out that the SCIAMACHY $XCH_4$ data product is the ratio of the retrieved $CH_4$ column and the retrieved $CO_2$ column. This means that this data product is only sensitive to methane sources which result in a relative methane column change which is significantly larger than the relative $CO_2$ column change due to emissions from the same source region. If for example a source produces a 1% $CH_4$ column enhancement and also a 1% $CO_2$ column enhancement (due to the emission of $CO_2$ and $CH_4$ molecules at a ratio of 200:1 (= 400 ppm : 2 ppm)), this emission source would not be visible in the SCIAMACHY $XCH_4$.

**Figure 6** - **Figure 12** show the corresponding results for the following regions: “PoValleyBig” (Italy), “Eastern Mediterranean”, “USEastCoast” (USA), “Mexico”, “India”, (mid/eastern) “China”, and “PearlRiverDelta” (China). The conclusions are similar as for the region BeNeLuxBig discussed above: Correlation of the atmospheric $XCO_2$ and $XCH_4$ as retrieved from the SCIAMACHY spectra using WFMDv2.0 with population density and/or anthropogenic emissions is low or absent.

**Table** 1 summarizes the results in terms of correlation coefficients. As can be seen, most of the correlation coefficients are positive but small.

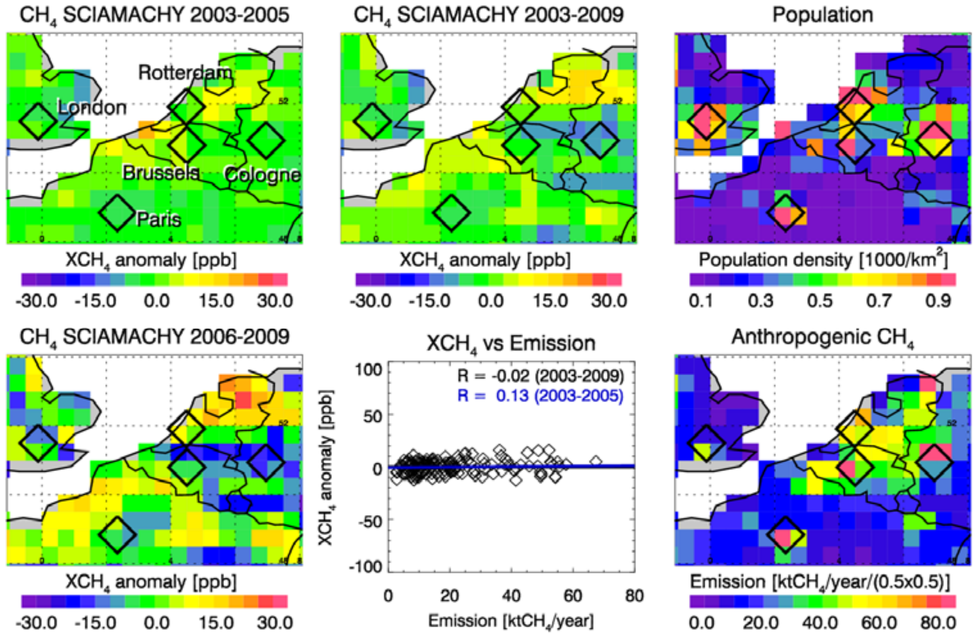
# Region: BeNeLuxBig

$CO_2$:



SCIAMACHY: WFMDv2 (sc:1.0,nsm=0) - Population: CIESIN/CIAT year 2000 - Emission: EDGAR v4.0 year 2005

$CH_4$:

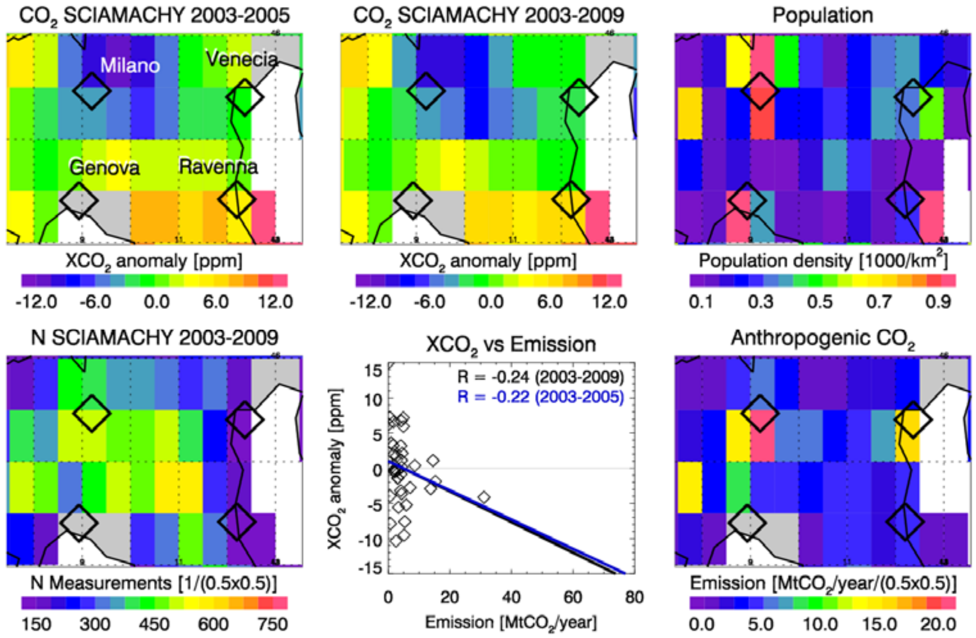


SCIAMACHY: WFMDv2 (ccs,sc:1.0,nsm=0) - Population: CIESIN/CIAT year 2000 - Emission: EDGAR v4.0 year 2005

**Figure 5:** SCIAMACHY $XCO_2$ (top) and $XCH_4$ (bottom) over the BeNeLuxBig CityZen region compared with population density and Edgar v4.0 anthropogenic emissions.

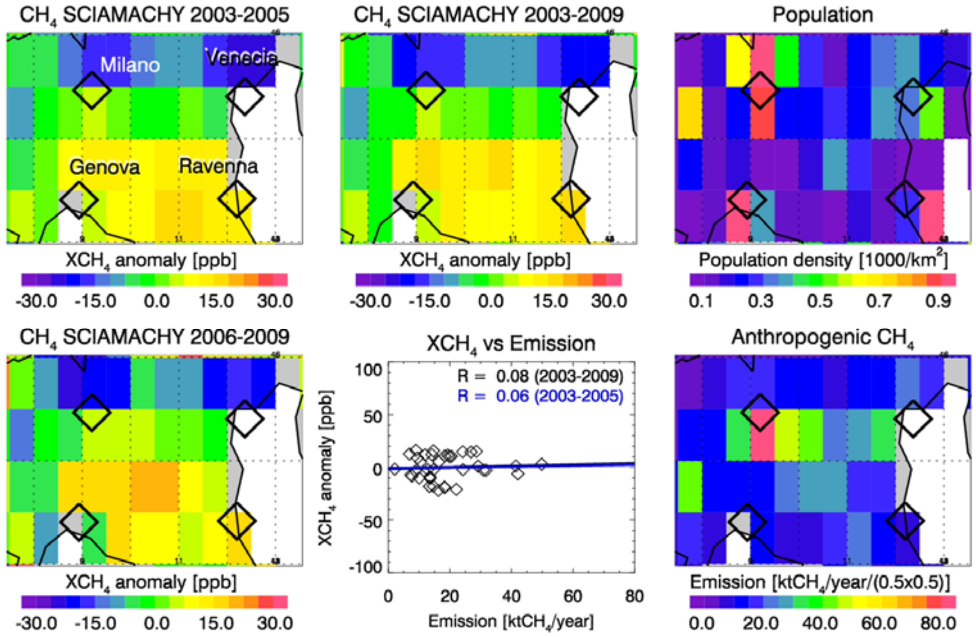
# Region: PoValleyBig

$CO_2$:



SCIAMACHY: WFMDv2 (sc:1.0,nsm=0) - Population: CIESIN/CIAT year 2000 - Emission: EDGAR v4.0 year 2005

$CH_4$:

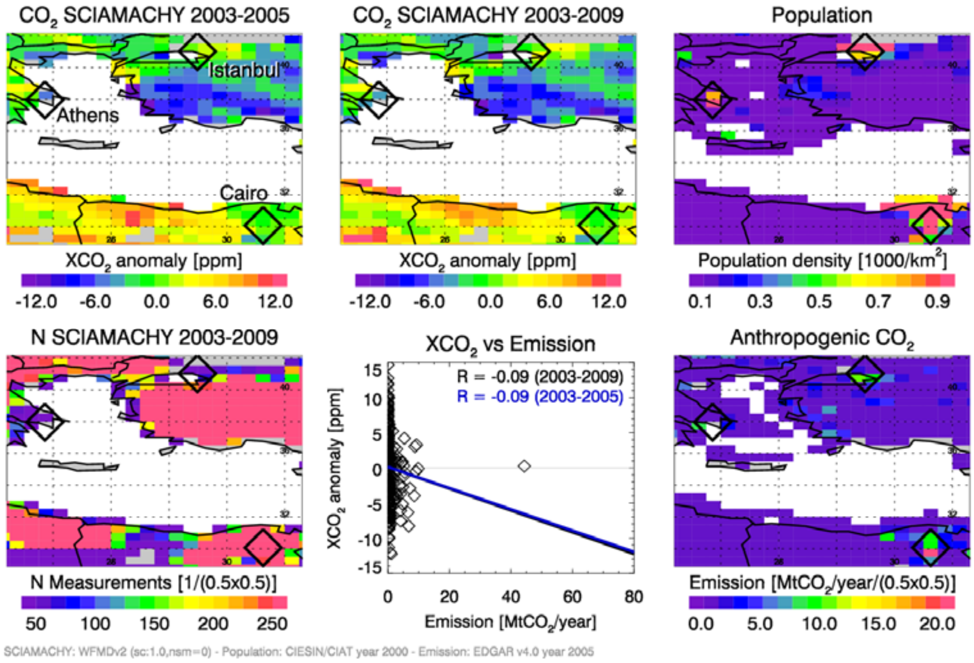


SCIAMACHY: WFMDv2 (ccs,sc:1.0,nsm=0) - Population: CIESIN/CIAT year 2000 - Emission: EDGAR v4.0 year 2005

**Figure 6:** As Figure 5 but for the region PoValleyBig.

# Region: EasternMediterranean

$CO_2$:



$CH_4$:

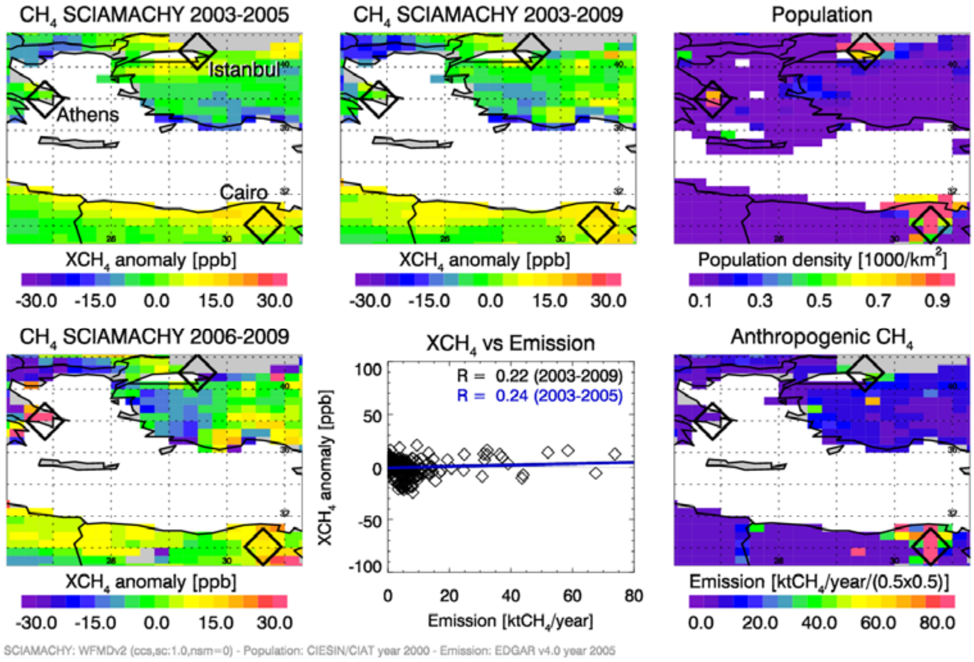
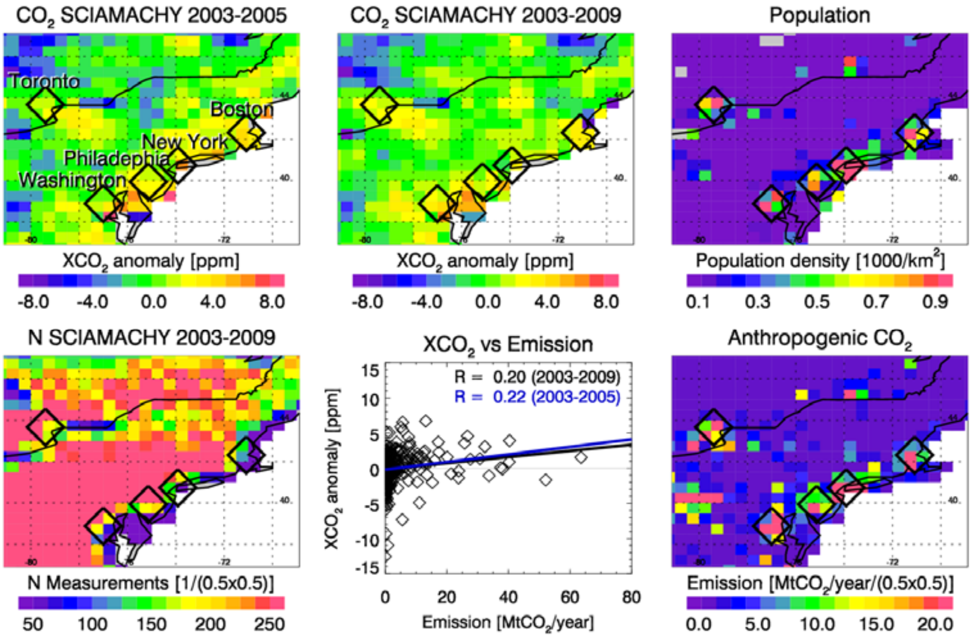


**Figure 7:** As Figure 5 but for the region EasternMediterranean.

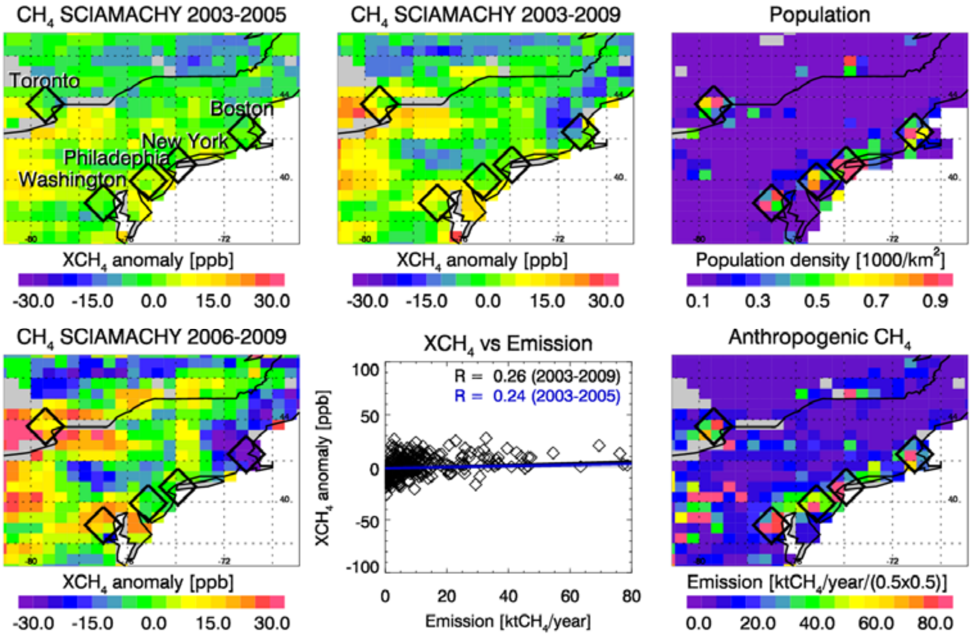
# Region: USEastCoast

$CO_2$:



SCIAMACHY: WFMDv2 (sc:1.0,nsm=0) - Population: CIESIN/CIAT year 2000 - Emission: EDGAR v4.0 year 2005

$CH_4$:

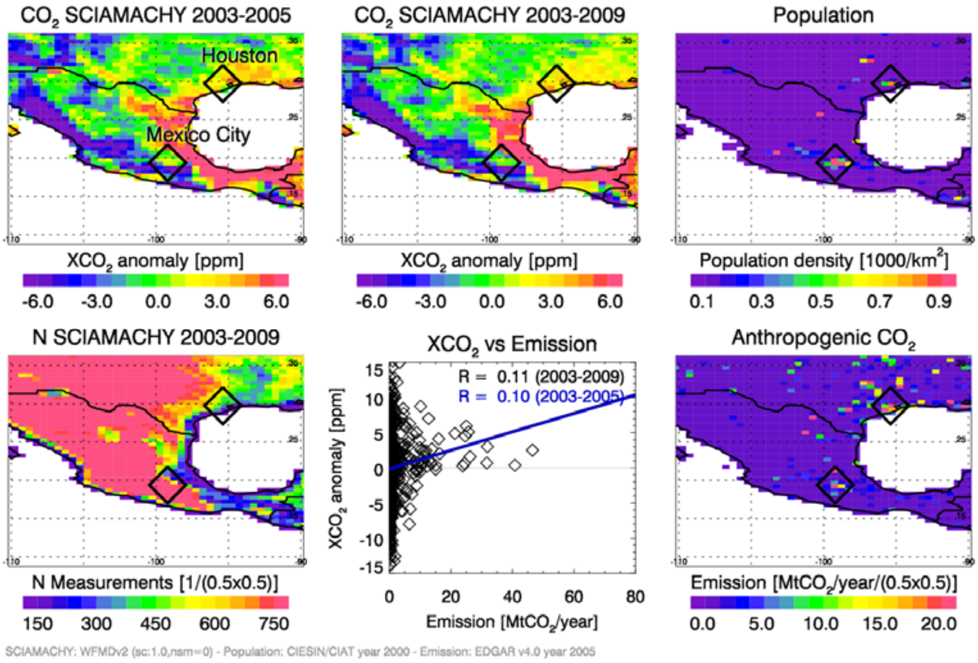


SCIAMACHY: WFMDv2 (ccs,sc:1.0,nsm=0) - Population: CIESIN/CIAT year 2000 - Emission: EDGAR v4.0 year 2005

**Figure 8:** As Figure 5 but for the region USEastCoast.

# Region: Mexico

$CO_2$:



$CH_4$:

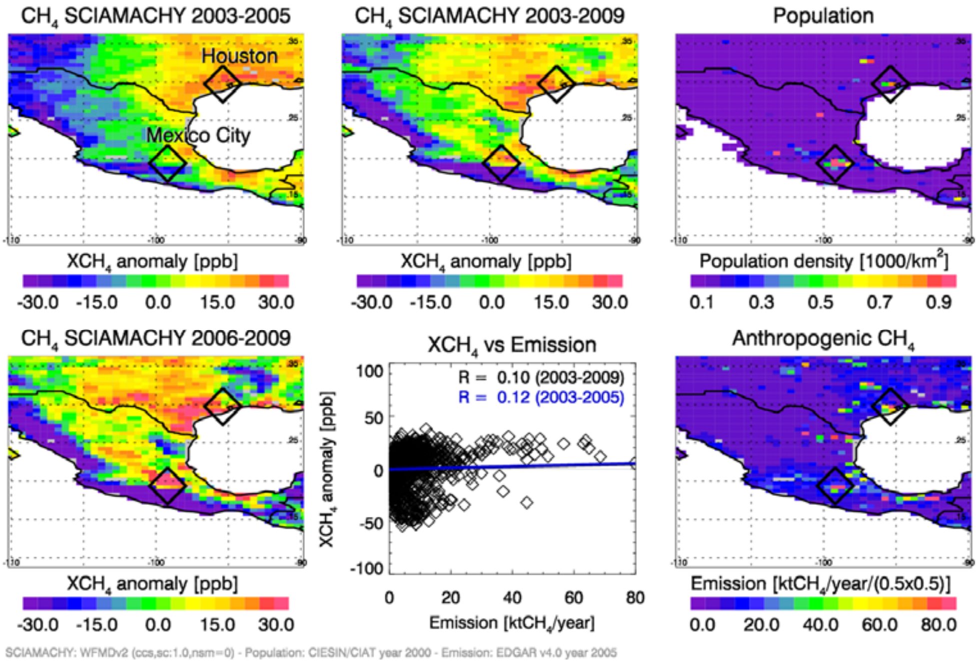
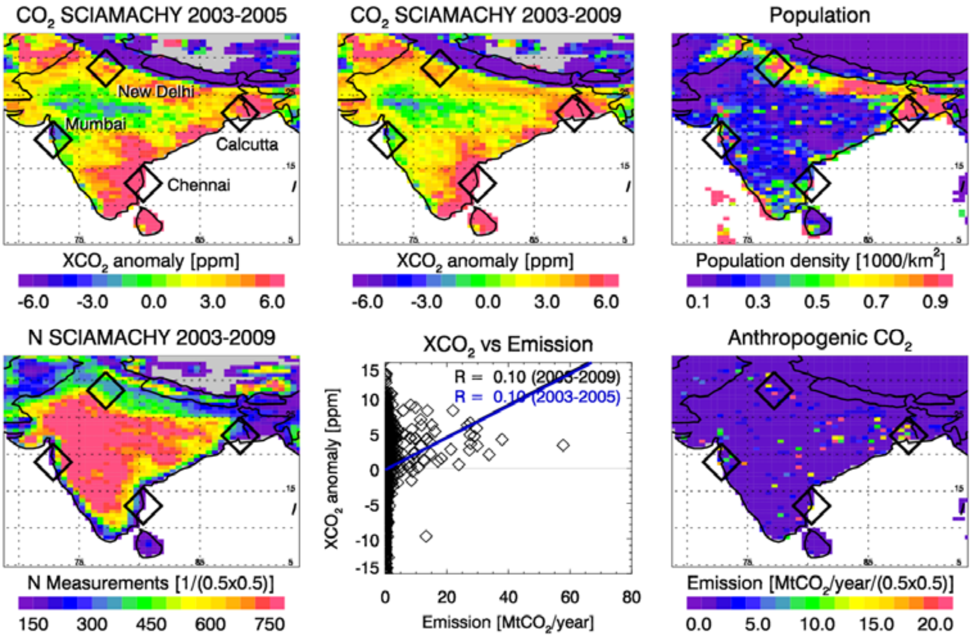


**Figure 9:** As Figure 5 but for the region Mexico.

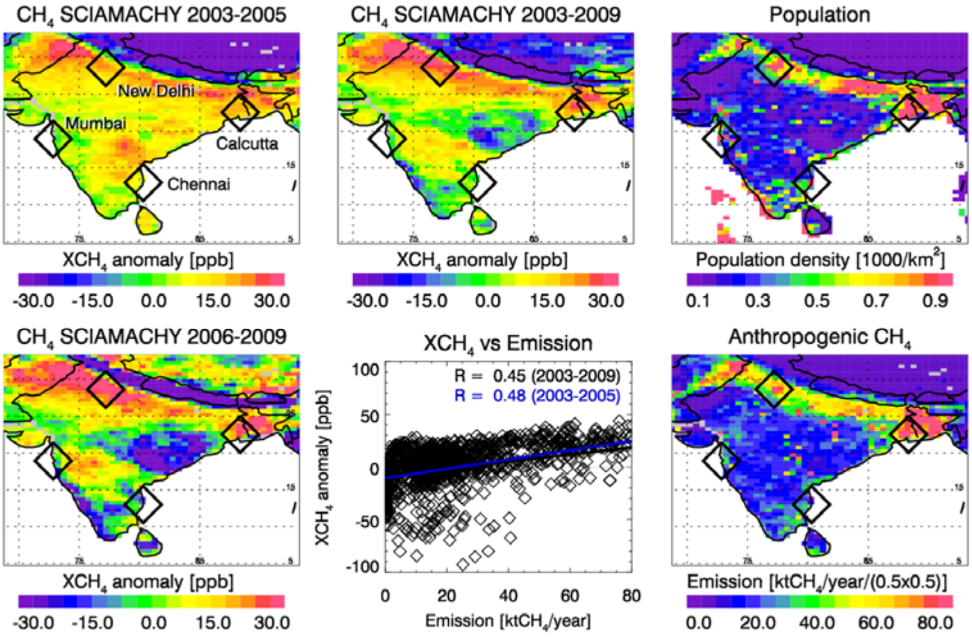
# Region: India

$CO_2$:



SCIAMACHY: WFMDv2 (sc:1.0,nsm=0) - Population: CIESIN/CIAT year 2000 - Emission: EDGAR v4.0 year 2005

$CH_4$:

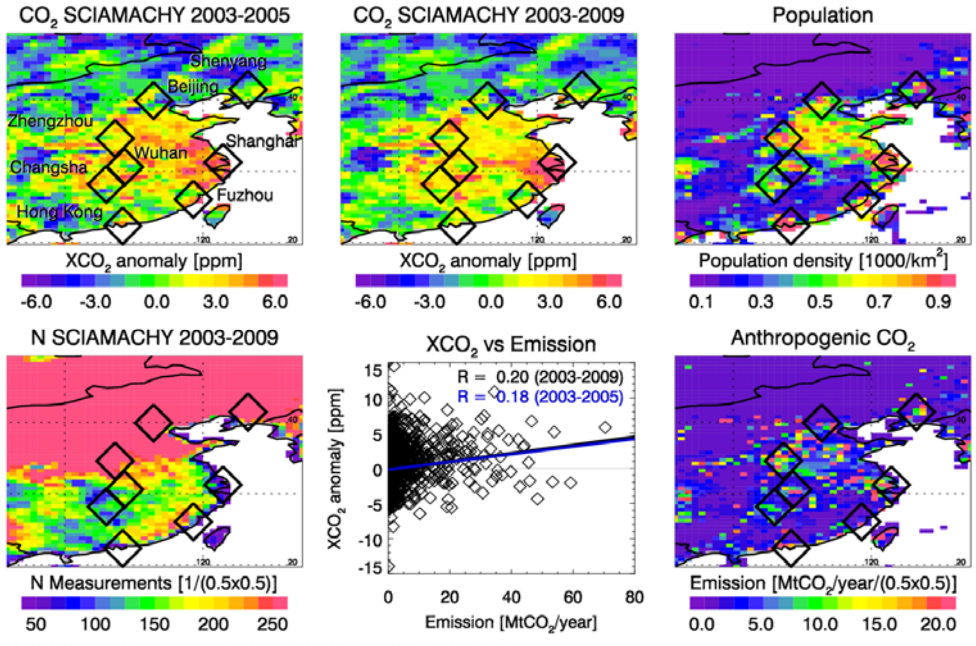


SCIAMACHY: WFMDv2 (ccs,sc:1.0,nsm=0) - Population: CIESIN/CIAT year 2000 - Emission: EDGAR v4.0 year 2005

**Figure 10:** As Figure 5 but for the region India.

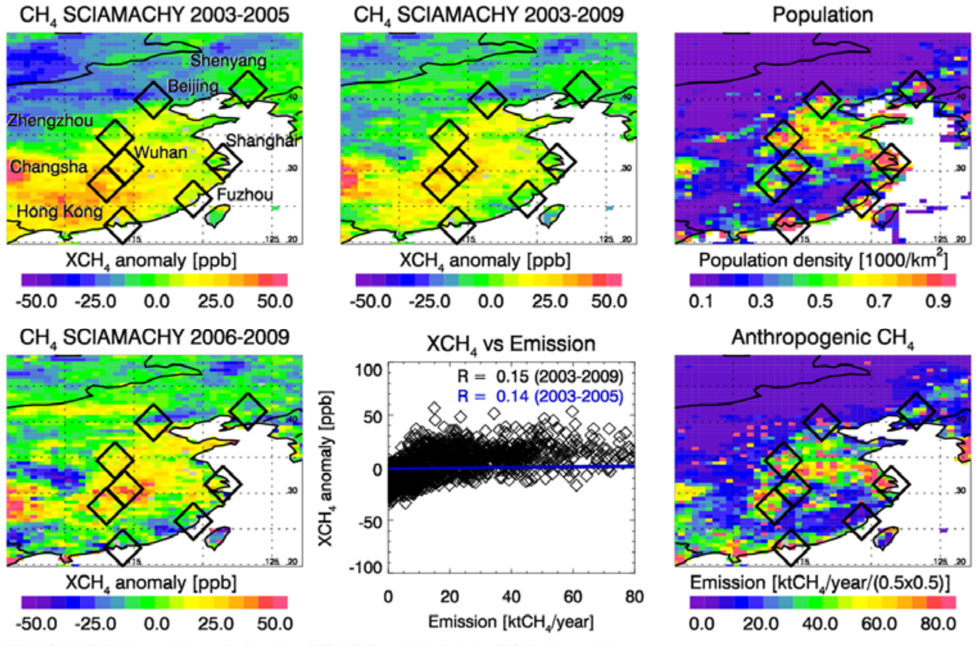
# Region: China

$CO_2$:



SCIAMACHY: WFMDv2 (sc:1.0,nsm=0) - Population: CIESIN/CIAT year 2000 - Emission: EDGAR v4.0 year 2005

$CH_4$:

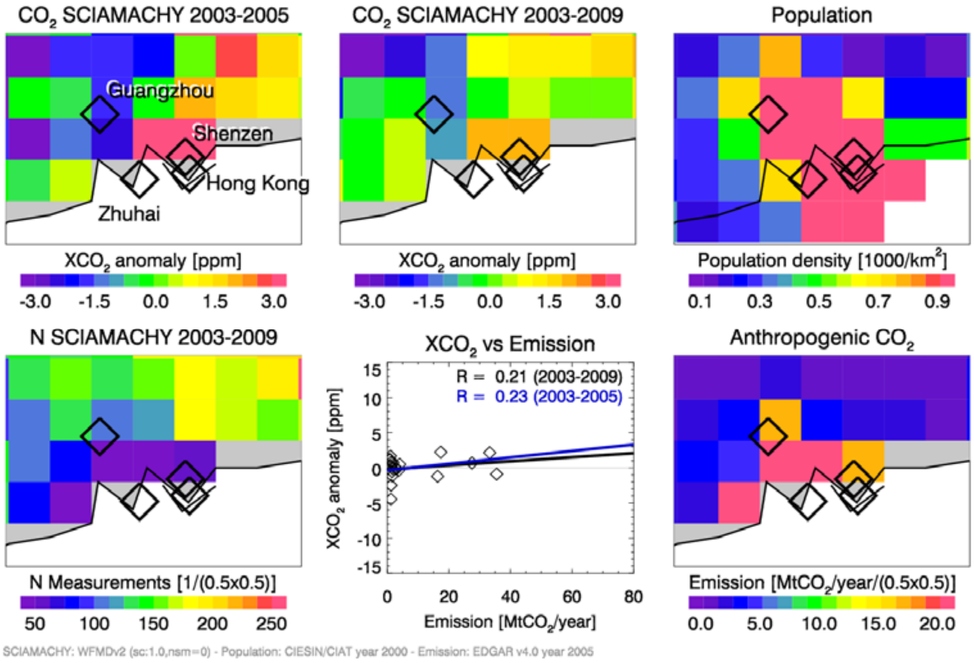


SCIAMACHY: WFMDv2 (ccs,sc:1.0,nsm=0) - Population: CIESIN/CIAT year 2000 - Emission: EDGAR v4.0 year 2005

**Figure 11:** As Figure 5 but for the region China.

# Region: PearlRiverDelta

$CO_2$:



$CH_4$:

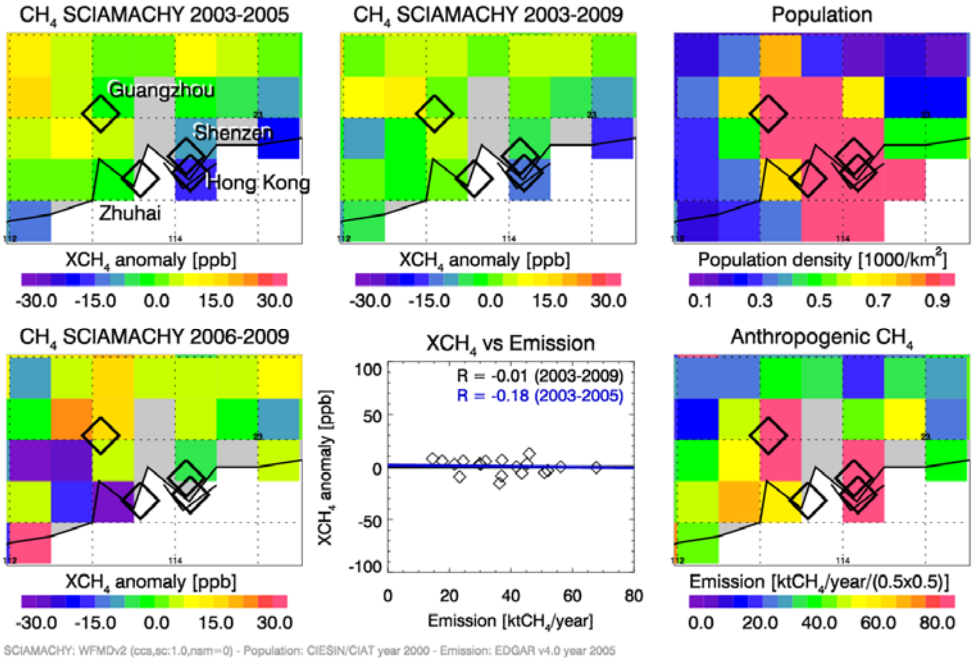


**Figure 12:** As Figure 5 but for the region PearlRiverDelta.

| | **Correlation coefficients**<br>**SCIAMACHY $XCO_2$/$XCH_4$ vs. Anthropogenic GHG Emissions at 0.5 deg x 0.5 deg resolution** | | | |
|---|---|---|---|---|
| | **$CO_2$** | | **$CH_4$** | |
| **Region** | **2003 -2005** | **2003 - 2009** | **2003 - 2005** | **2003 - 2009** |
| BeNeLuxBig | +0.24 | +0.16 | +0.13 | -0.02 |
| PoValleyBig | -0.22 | -0.24 | +0.06 | +0.08 |
| EasternMediterranean | -0.09 | -0.09 | +0.24 | +0.22 |
| USEastCoast | +0.22 | +0.20 | +0.24 | +0.26 |
| Mexico | +0.10 | +0.11 | +0.12 | +0.10 |
| India | +0.10 | +0.10 | +0.48 | +0.45 |
| China | +0.18 | +0.20 | +0.14 | +0.15 |
| PearlRiverDelta | +0.23 | +0.21 | -0.18 | -0.01 |

**Table 1:** Correlation between the gridded SCIAMACHY $XCO_2$ and $XCH_4$ retrievals and EDGAR anthropogenic emissions at $0.5^o$x$0.5^o$ spatial resolution for two different time periods expressed as linear correlation coefficient (for details see **Figure 5 - Figure 12**).

## 1.4.Greenhouse gases from SCIAMACHY: Summary and conclusions

IUP-UB’s responsibility within Task 2.1 is to provide consistent time series of the two most important anthropogenic greenhouse gases $CO_2$ and $CH_4$ on the global scale derived from SCIAMACHY/ENVISAT nadir observations. To achieve this IUP-UB has improved the WFM-DOAS algorithm, which enables to retrieve column-averaged mole fractions of $CO_2$ and $CH_4$, denoted $XCO_2$ and $XCH_4$, and has used this algorithm for processing seven years of SCIAMACHY data (2003-2009). The data products have been compared with global models, gridded population density and emission data bases. While reasonable to good agreement with global models concerning large-scale features such as hemispheric annual increase and seasonal cycle has been found, correlation with population density and anthropogenic emissions at city scale ($0.5^o$ x $0.5^o$; ~50 km) is poor; the linear correlation coefficient is typically positive but less than 0.3 even for 7-year averages. In other words, elevated concentrations of $CO_2$ and/or $CH_4$ for individual cities, including megacities such as Mexico City, can typically not be detected. This finding is not unexpected. Due to the long lifetime of the two gases elevated concentrations are not restricted to areas close to the source and even a strong source is expected to typically only produce a very small (sub-percent) enhancement relative to the (variable) background concentration. In addition, strong non urban sources need to be considered, especially for methane, e.g., emissions from near-by wetlands. The expected (mega)city emission signal is typically much less than the noise level and detection requires averaging many individual measurements and (nearly) bias free retrievals. Due to the strict quality filtering of the SCIAMACHY retrievals, typically only a few hundred measurements per $0.5^o$ x $0.5^o$ grid cell are available for averaging even for a multi-year data set. Possible biases of the satellite retrievals, e.g. due to aerosols and residual clouds, may also contribute to the finding that elevated concentrations over individual cities can typically not be detected. As a result, the regional emission signal is typically below the SCIAMACHY detection limit. It was therefore not possible to derive trends and perform quantitative comparisons with bottom-up emission data as originally planned for this project.

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