CARBON MONOXIDE AND RELATED TRACE GASES AS RETRIEVED FROM SCIAMACHY OVER CO SOURCES REGIONS

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Summary

The spectral measurements of SCIAMACHY onboard ENVISAT contain information about various important atmospheric trace gases. Here we discuss simultaneous measurements of carbon monoxide (CO), formaldehyde (HCHO), and nitrogen dioxide (NO2), retrieved from the SCIAMACHY nadir observations using retrieval algorithms developed at the University of Bremen. We focus on the year 2004 and on major CO source regions such as Alaska (boreal forest fires during June), Indonesia (tropical forest fires during October-November), USA and Europe (mainly anthropogenic CO sources) and Asia (various sources). To get additional information for the various regions and their trace gas sources, we use AATSR fire counts, population density and ecosystem type maps as well as CO emissions from the EDGAR 3.2 database. The primary goal of this ongoing research activity is to increase our knowledge about the surface sources of the air pollutant carbon monoxide.

Introduction

CO is an important air pollutant and plays a key role in tropospheric chemistry. In general, the processes leading to the emission of CO in the troposphere are relatively well established. Its budget, however, is still uncertain (Petron et al., 2002). Remote sensing can be a helpful tool for CO global sources analysis. SCIAMACHY is a unique instrument for such an application, as SCIAMACHY is able to measure the CO vertical column with nearly equal sensitivity to all atmospheric layers, including the boundary layer, where the CO source signal is largest. Recently, three years (2003-2005) of SCIAMACHY CO columns have been retrieved using version 0.6 of the scientific retrieval algorithm WFM-DOAS (Buchwitz et al., 2007, Khlystova et al., 2006). Here we discuss this new CO data set focusing on the year 2004 and on various CO source regions. We also show simultaneous SCIAMACHY retrievals of formaldehyde (HCHO) total columns (Wittrock et al., 2006) and nitrogen dioxide (NO_2) tropospheric columns (Richter et al., 2005) as this provides additional information on the various sources.

Dils et al., 2006, have performed a first validation of the WFM-DOAS version 0.6 CO column data set by comparison with ground-based FTS measurements focusing on Europe. For the year 2004 they found on average good agreement between the SCIAMACHY and the FTS CO columns (average bias close to zero, standard deviation of the difference ~20% close to the noise of the SCIAMACHY data). The total error for HCHO comprises a number of different error sources, both, random and systematic (see Wittrock et al., 2006 and references given therein). The monthly statistical error for HCHO has been estimated to be 0.4×10^{16} molecules/cm^2 (~40%); the systematic error is about 0.1×10^{16} molecules/cm^2 (~10%). The total error for NO2 comprises additive (~$0.5\text{-}1 \times 10^{15}$ molecules/cm^2) and multiplicative (~10-20%) parts. The uncertainty for annually average NO2 is about 10% (Richter et al., 2005).

Atmospheric CO originates from (incomplete) combustion processes such as forest and savanna fires, fossil fuel burning, and biofuel burning but also from oxidation of methane and non-methane hydrocarbons (NMHC) such as HCHO. HCHO originates (photo)chemically from various pre-cursor gases emitted from vegetation (e.g., isoprene), fires, and anthropogenic activities (e.g., hydrocarbon emissions from industrial activities and traffic). Nitrogen oxides (NOx) are emitted by all combustion processes (Richter et al., 2005). In industrialized areas, NOx emissions are dominated by anthropogenic activities, such as road transportation (car engine), industrial combustion, electricity generation (turbines are driven by burning of fossil fuels like coal, natural gas, or petroleum), and heating systems; in rural regions NOx emissions are dominated by emissions from biomass burning, soil and lightning.

Here we focus on fire emissions and industrialized pollutions. Large fires emit significant amounts of CO and many other trace gases such as HCHO and NO2. Relatively oxidised components (such as NO_2) are emitted during the flaming stage (higher temperatures) of a fire, whereas more reduced components (such as CO and nonmethane hydrocarbons (e.g., HCHO)) occur during the smoldering stage (incomplete combustion at lower temperatures) (see Brasseur et al., 2003, and references given therein, Andreae and Merlet, 2000, Lobert et al., 1991). Over industrialized regions, the population density map shows the possible locations of anthropogenic pollution sources. This information can be used to separate the man-made sources from other kind of sources (such as, for example, biogenic sources).

Results

Figure 1 shows simultaneously measured CO, HCHO, and tropospheric NO_2 as retrieved from SCIAMACHY over Alaska and Canada during June 2004. During this period, several large boreal forest fires occurred in this region as shown by the AATSR fire counts (algorithm 2 data product obtained from http://dup.esrin.esa.it/ionia/wfa/index.asp). As can be seen, all three gases have elevated concentrations close to or near the fires. Spatially, the NO_2 and the HCHO show better correlation with the fire counts, whereas the maxima of the CO columns are located somewhat north of the fire counts. This may be due to atmospheric transport, the irregular sampling of the SCIAMACHY data, and due to the different lifetimes of the three gases (CO: typically 2 months, NO_2: hours to days (Richter et al., 2005), HCHO: few hours (Arlander, et al., 1995)). In this case, it is difficult to find exact correlation between CO and the fire counts. Nevertheless, the general similarity of the trace gas

pattern can be used as an indication on the CO origin. All three measured gases show the significantly elevated concentrations at or near the fires.

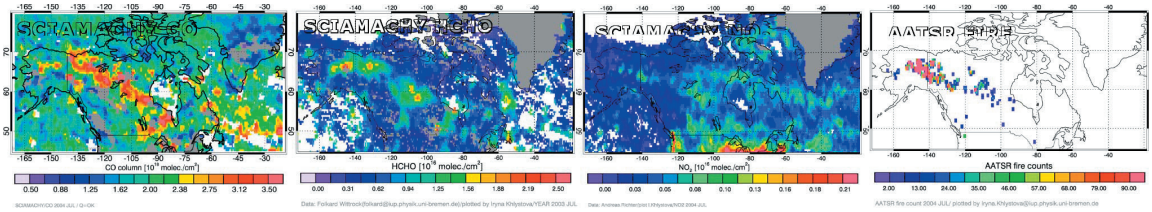

Figure 1. *Atmospheric trace gases over North America during July 2004. From the left to the right: SCIAMACHY CO and HCHO total columns, SCIAMACHY tropospheric NO_2 column, and AATSR fire counts.*

Figure 2 shows similar results but for Indonesia and northern Australia during October-November 2004. As can be seen, a large number of fires occurred in this region. The SCIAMACHY CO shows a good correlation with the AATSR fire counts, especially over northern Australia. HCHO is elevated over (from left to right) Java, Borneo, and New Guinea, well correlated with the AATRS fire counts. NO2 is high primarily over Java and parts of northern Australia, but not over Borneo and New Guinea, which may be due to different fuel types or combustion conditions. Moreover, despite large data gaps in the SCIAMACHY CO, an at least reasonable correlation between CO, fire counts and elevated concentrations of the other trace gases is visible in this figure.

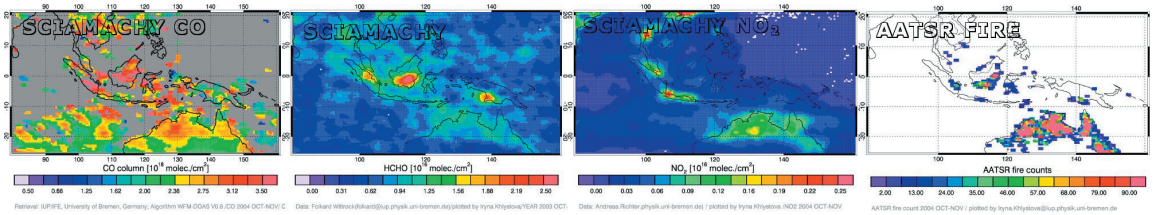

Figure 2. *As Fig. 1 but for Indonesia and northern Australia during October-November 2004.*

Figures 3 and 4 show simultaneously measured CO, HCHO, and tropospheric NO2 over the industrialized areas of North America and Europe during 2004. As can be seen from the pie plots (made using information obtained from the EDGAR 3.2 website), fossil fuel burning is a main source of CO for these regions. Over the US (Fig. 3), the CO is reasonably well correlated with population density. High CO occurs, for example, near highly populated areas along the west coast, as well as over most parts of the middle and eastern US. HCHO is high over the eastern US but not along the west coast. Tropospheric NO2 is highest over the north-eastern part of the US, well correlated with population density, also along the west coast, especially around San Francisco and Los Angeles. In contrast to NO2, HCHO is also high over large parts of south-east USA, very likely due to HCHO originating from biogenic emissions and corresponding oxidation processes. For Europe (Fig. 4), the results are similar as for the US. Generally, the SCIAMACHY CO columns over these industrialized regions are much smaller compared to region with strong biomass burning pollution.

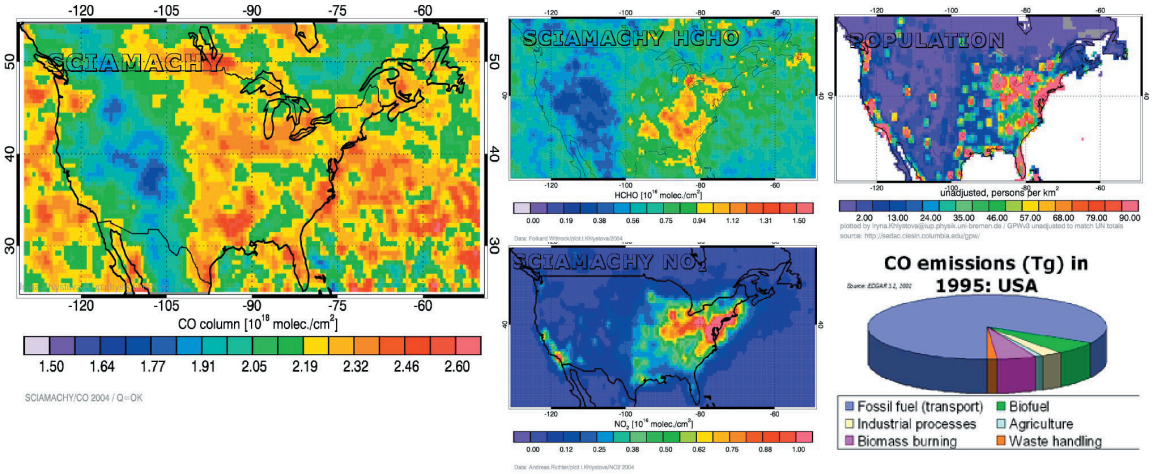

Figure 3. *SCIAMACHY year 2004 CO (left), HCHO (top middle), NO_2 (bottom middle), population density (CIESIN/CIAT, 2005) (top right) over North America, and pie plot of EDGAR USA CO sources (bottom right).*

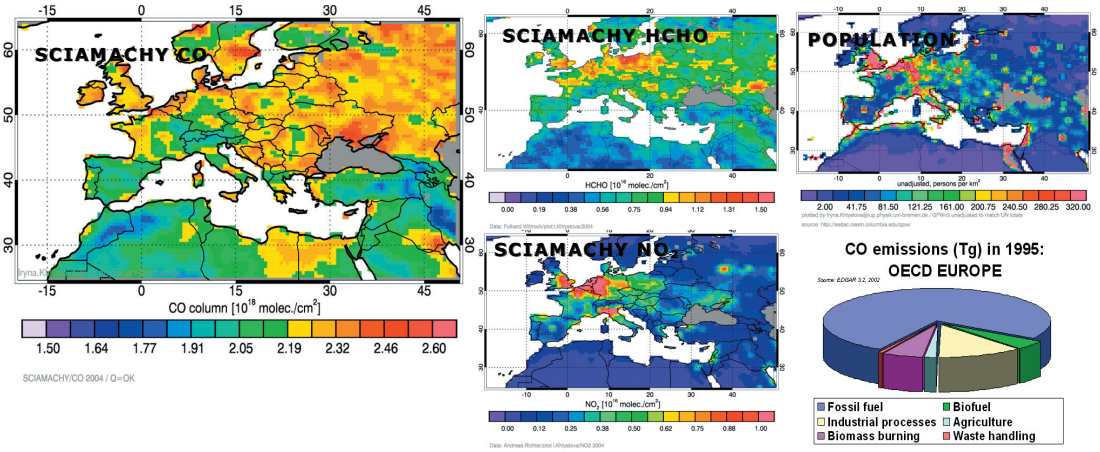

Figure 4. *As Fig. 3 but for Europe (over land).*

Figure 5 shows the distribution of pollutants over eastern Asia for 2004. As can be seen, China emits huge amounts of carbon monoxide. CO is highest over regions with high population density but also over Southeast Asia due to fires. NO2 over China is also well correlated with population density. This is also true for HCHO. HCHO however is, in contrast to NO2, also high over Southeast Asia, and reasonably well correlated with the fires. Shown is also CO from MOPITT, which is well correlated with the SCIAMACHY CO, although the SCIAMACHY CO appears to be somewhat higher, very likely due to the higher sensitivity of the SCIAMACHY measurements for the boundary layer. For eastern Asia two sub-regions with different CO main sources can be defined. The northern part around Beijing, with high population density and high anthropogenic CO emissions, and Southeast Asia, where population density and anthropogenic CO emissions are lower but a significant amount of CO is emitted from biomass burning (agricultural waste burning and tropical forest fires). Over the northern part around Beijing, all three gases are enhanced, whereas over Southeast Asia mainly CO and HCHO are high. This may indicate different conditions for burning processes or different fuel types. The variation in the ecosystem types can be an additional indication for the different burning conditions in the region. In addition, EDGAR data show, that the main CO sources in Asia are different for these two sub-regions. In the north-east fossil fuel and bio fuel use are the main sources and in the south-east biomass burning and bio fuel burning dominate.

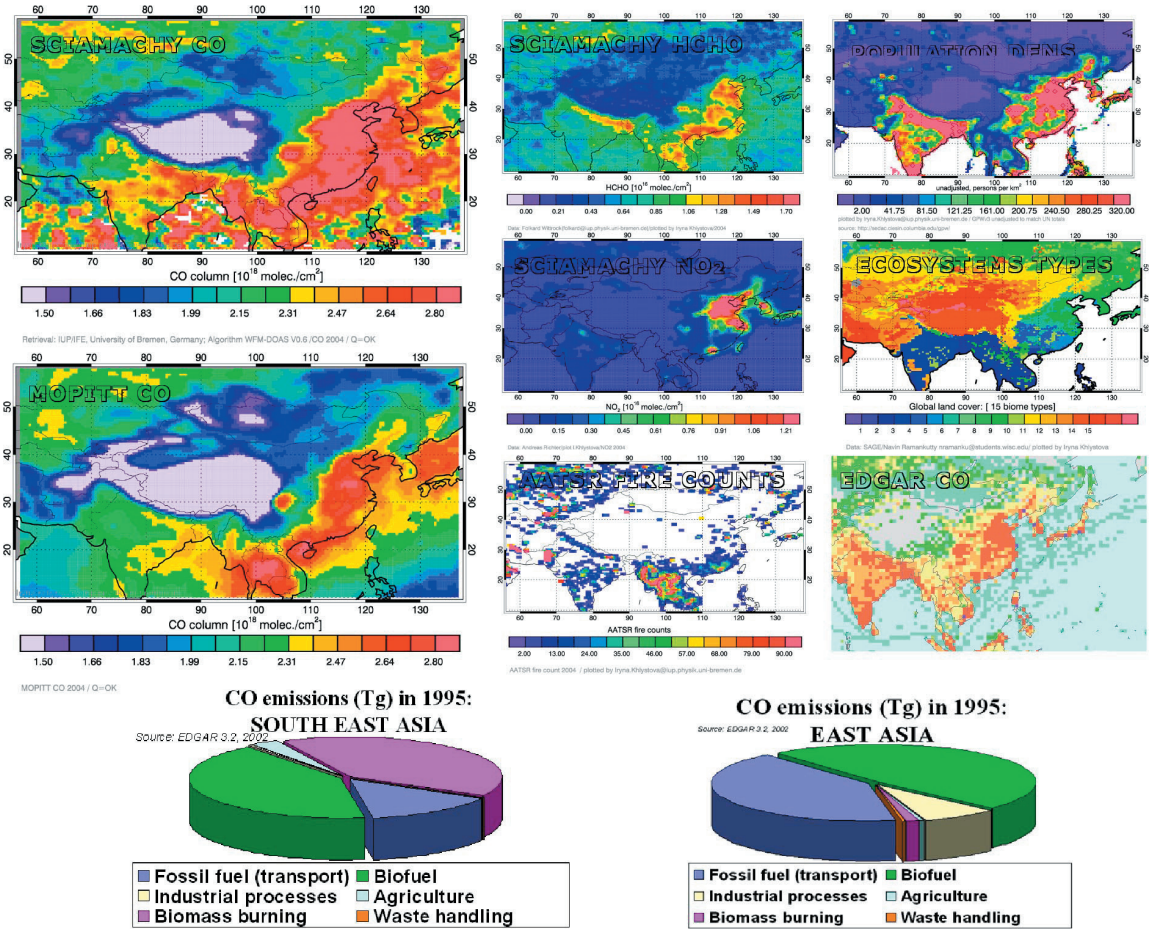

Figure 5. *SCIAMACHY year 2004 CO (top left) and MOPITT CO columns (bottom left) over Asia. Middle: SCIAMACHY HCHO vertical columns (top), SCIAMACHY NO_2 tropospheric column (middle), and AATSR fire count (bottom). Right: CIESIN/CIAT population density for 2005 (top), SAGE ecosystems types (middle), and EDGAR 3.2 CO emissions (bottom). EDGAR CO emissions are represented as pie plots for two regions.*

Conclusions

Simultaneous satellite measurements of the important air pollutants CO, HCHO, and NO2 over various source regions have been presented and discussed. Additional information such as fire counts and population density maps have been used to identify source regions and to discriminate between the various types of pollution sources. The goal of this ongoing research activity is to enhance our knowledge about the various air pollution sources, in particular carbon monoxide.

Acknowledgements

We thank ESA and DLR for providing SCIAMACHY calibrated level 1b spectra. The MOPITT level 2 CO data product has been obtained via the NASA Langley DAAC. The population density map has been obtained from http://veimages.gsfc.nasa.gov/116/popdensity.jpg. Exchange of information within the network of Excellence ACCENT is acknowledged. This work has been funded by the DLR-Bonn (SADOS), ESA (PROMOTE), the EU (AMFIC) and by the University and the State of Bremen.

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