Project:	**1104**
Project title:	**Atmosphärische Spurenstoffe (ATS)**
Project PI:	**Julian Kostinek**
Report period:	**1.1.2021 - 31.12.2021**

The focus of this project is on analyses of trace species measurements with high-resolution numerical simulations, mainly for long-lived anthropogenic greenhouse gases, e.g. carbon dioxide (CO_2), methane (CH_4) and nitrous oxide (N_2O) but also short-lived trace species, e.g. sulfur dioxide (SO_2) and nitrogen oxides (NO_x). Last years proposal was small and tailored to complement aircraft field campaigns (Methane-To-Go Europe, Methane-To-Go Africa) and a satellite program (CO2Image). Unfortunately, the Methane-To-Go Africa field campaign had to be postponed to the upcoming allocation period (2022) due to the Covid19 pandemic and the satellite program also suffered from delays. We therefore forfeited some resources.

Nevertheless, great progress has been made with respect to two out of three work packages involved in this project during the current allocation period 01.01 - 31.12.2021. The following sections will provide a short summary of each work package highlighting some of the key findings and scientific output, enabled by the DKRZ's HLRE-3.

WP1 - Quantifying regional and local greenhouse gas emissions in Europe and the U.S.

Methane (CH_4) emissions from coal production amount to roughly one-third of European anthropogenic CH_4 emissions into the atmosphere, with the Upper Silesian Coal Basin (USCB) in southern Poland being one of Europe's emission hot spots, mainly driven by mining and industrial activities. In this work package, FLEXPART-WRF (Brioude et al., 2013, Stohl et al., 2010) and WRF (Skamarock et al., 2019) were used to propagate CH_4 fluxes from known coal mine ventilation shafts in the USCB forward in time. The emission rates from each ventilation shaft are scaled a-posteriori in order to minimize residuals between model and airborne and ground-based measurements, acquired during the CoMet 2018 campaign. From airborne data and a Bayesian inversion approach, we find instantaneous emission estimates of 451/423 ± 77/79 $ktCH_4\ yr^{-1}$ for two flights on June 6th, 2018. These emission rates coincide with annual-average inventorial data from E-PRTR 2017 (6%) although they are distinctly lower (-28%/-32%) than values reported in the EDGAR v4.3.2 inventory. The findings have been published in Kostinek et al., 2021.

The work on high-resolution large eddy atmospheric simulations for anthropogenic CO_2 point sources, such as coal-fired power plants to support the determination of emission rates from integrated differential absorption lidar measurements (active airborne remote sensing) has been published in Wolff et al., 2021. Using these simulations enabled a qualitative assessment of turbulence on the established cross-sectional flux method. The optimization of the emission rates with an ensemble Kalman smoother (CarbonTracker Data Assimilation Shell, "CTDAS", van der Laan-Luijkx et al., 2017) is further investigated. A first test run, with reduced number of ensemble members, as well as two instead of three domains has been completed. In a next step the number of ensemble members will be increased and a third domain will be introduced. The computationally intensive simulations are thus envisaged within the current allocation period.

N_2O is the third most important long-lived anthropogenic greenhouse gas after CO_2 and CH_4 in terms of radiative forcing. Within this work package, work on N_2O emissions in the U.S. Midwest has been continued using an unique in situ N_2O dataset gathered during the ACT-America campaigns in 2017 and 2019. Appropriate scaling factors for the EDGAR inventory were deduced for two characteristic periods. We find scaling coefficients for agricultural EDGAR4.3.2 / EDGAR5.0 emissions of 6.3±4.6 / 3.5±2.7 for Oct. 2017 and 11.4±6.6 / 9.9±5.7 for June/July 2019, respectively. EDGAR reported N_2O emissions are thus significantly underestimated. These findings have been published in Eckl et al., 2021.

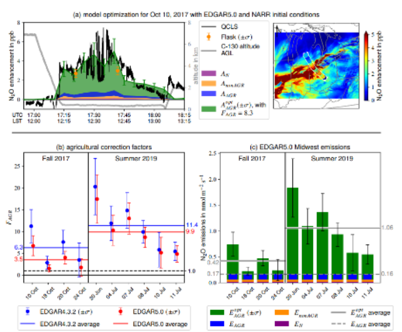


Figure 1 (a) Model optimization for Oct 10, 2017 with EDGAR5.0 and NARR initial conditions. (b) Agricultural correction factors. (c) EDGAR5.0 Midwest N2O emissions with optimized and non-optimized EAGR. (Eckl et al., 2020)

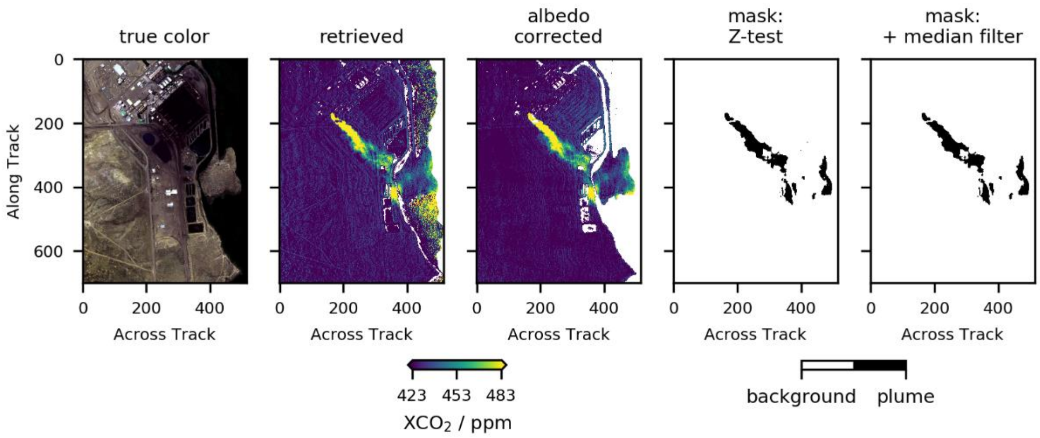
WP2 – Towards space-borne monitoring of localized CO_2 emissions

This work package delivered a concept and first performance assessment of a compact space-borne imaging spectrometer with a spatial resolution of 50 × 50 m^2. Through radiative transfer simulations with RemoteC (Butz et al., 2011), including a realistic instrument noise model it is concluded that an instrument noise error of 1.1 ppm (1σ) can be achieved for the retrieval of the column-averaged dry-air mole fraction of CO2 (XCO2).

In the current allocation period, we further specialized our CO2 retrieval capabilities with the RemoTeC radiative transfer modelling software to aircraft measurements from the airborne imaging sensor AVIRIS-NG (Chapman et al., 2019). These observations capture power plant emission signals at a spatial resolution of a few meters, which enables studies of emission rate inversions from an existing imaging sensor comparable to the concept described above.

We found that, at the coarse spectral resolution of AVIRIS-NG (FWHM=5 nm), albedo signals may interfere with molecular absorption features, making the retrieval challenging. Our work at DKRZ has allowed us to identify favourable retrieval configurations. For example, we found that XCO2 retrievals could be improved by constraining the interfering water vapor absorption through an additional spectral retrieval window at 1.8 um.

The best retrieval set-ups were applied to different power plant overflights and we subsequently developed plume masks, posterior correction methods and we estimated the emission rates of the facilities. The image below illustrates the steps taken to obtain the CO2 column enhancements due to the power plant from the AVIRIS-NG observation. Our work is currently being prepared for publication in a peer reviewed journal within the next months.



WP3 – Plume dispersion forecast and analyses for aircraft campaigns

This work package was intended for plume dispersion simulations with the HYSPLIT modelling system (Draxler et al., 1998, 2020) for short- and long-lived trace gases on the regional scale. Besides of actual weather forecasts, these tailored chemical forecasts are one of the main pillars supporting the planning of research aircraft flights. The simulations are particularly time-critical, since forecasts covering several days are needed daily. Although not as resource-intensive as WRF-type simulations, we have to resort to a supercomputer for timely and reliable HYSPLIT forecasts that would not be possible otherwise. Unfortunately, the requested resources could not be used in the current allocation period due to postponement of the aircraft campaign. We will re-request the resources for the postponed missions in the upcoming allocation period.

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