Combining a receptor-oriented framework for tracer distributions with a cloud-resolving model to study transport in deep convective clouds: Application to the NASA CRYSTAL-FACE campaign

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[1] Quantitative models of deep convection play a central role to improve understanding of weather, trace gas distributions, and radiative regime of the upper troposphere. Cloud-resolving models of deep convection are useful tools to simulate relevant processes. Observations of tracers such as CO_2 can provide critical constraints on mass transport within these models. However, such measurements do not span the entire four-dimensional domain in space and time. We introduce a new method to improve tracer constraints on such models, combining a Receptor-Oriented Atmospheric Modeling (ROAM) framework with airborne and ground-based CO_2 data. We illustrate the application of ROAM in generating initial and boundary conditions of CO_2 for cloud-resolving model simulations, for a case study in the CRYSTAL-FACE campaign. Observations and model results were compared for CO_2 profiles from the surface up to 16 km, inside and outside of a deep convective cloud. ROAM generated concentration fields that agreed within 0.5 ppm (1σ) of observations outside the cloud. When ROAM-derived initial and boundary CO_2 concentrations were fed to a state-of-the-art cloud-resolving model (DHARMA), the combined modeling system successfully reproduced observed concentration differences, 0.2–0.8 ppm, between in-cloud and out-of-cloud air at $9 \sim 14$ km. Results suggest that ~25% of air at 14 km was lifted through the convective system from the PBL. This study demonstrates the potential of the receptor-oriented framework to constrain redistribution of air within convective systems using CO_2, and it points to the need for better coordinated tracer measurements in future field missions. *INDEX TERMS:* 0322 Atmospheric Composition and Structure: Constituent sources and sinks; 0368 Atmospheric Composition and Structure: Troposphere—constituent transport and chemistry; 0341 Atmospheric Composition and Structure: Middle atmosphere—constituent transport and chemistry (3334). **Citation:** Xueref, I., et al. (2004), Combining a receptor-oriented framework for tracer distributions with a cloud-resolving model to study transport in deep convective clouds: Application to the NASA CRYSTAL-FACE campaign, *Geophys. Res. Lett.*, *31*, L14106, doi:10.1029/2004GL019811.

1. Introduction

[2] Convection plays a major role in determining properties of the atmosphere, linking air from the planetary boundary layer (PBL) with the upper troposphere. For example, convection redistributes water in its various forms, strongly affecting formation of cirrus clouds and the upper tropospheric radiative regime [*Eagleson*, 2000]. There is wide theoretical and experimental evidence that convection plays a major role in transporting pollutants [e.g., *Dickerson et al.*, 1987]. The large variety of interacting nonlinear processes and extensive range of spatio-temporal scales make development of cloud-resolving models (CRMs) that accurately simulate deep convective clouds difficult, and render parameterizations used in large-scale models highly uncertain [*Lu et al.*, 2000]. In this sense, profile data obtained by aircraft, balloon and ground measurements are usually used to constrain CRMs [*Pickering et al.*, 1988], as will satellite data be more and more in the future. In particular, tracer measurements can help to assess the transport of air masses within convective systems, since atmospheric concentrations of these species depend on the sources of the air masses they belong to [*Dickerson et al.*, 1987]. Here we argue that tracer studies using atmospheric CO_2 can potentially provide detailed information on transport rates and mechanisms [*Andrews et al.*, 2001], due to the special characteristics of CO_2: spatially and temporally varying surface fluxes, extensive surface observations [*Wofsy and Harriss*, 2002], and absence of chemical transformations in the atmosphere. Contrasts between CO_2 in the PBL and the free troposphere typically develop over land during the growing season, and these gradients provide signals to constrain rates of convective transport of air from the PBL to higher altitudes. If the 3-dimensional field of tracer concentrations is known prior to onset of convection, and concentrations are measured during and after convection, the observations provide integral constraints on the magnitudes of the transport fluxes.

[3] In this paper we use observations of atmospheric CO_2 to study convective transport during the NASA-led CRYSTAL-FACE campaign that occurred over Florida during summer 2002 [*Jensen et al.*, 2004]. A modeling framework is required that simulates tracer redistribution

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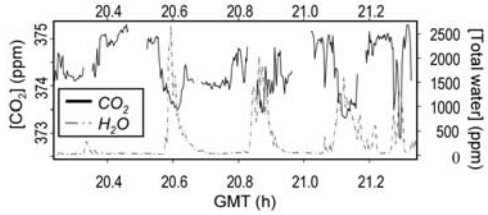

Figure 1. Observed time series of CO_2 and total water, from the 16 July 2002 flight.

within the convective system (e.g., by a cloud resolving model) and also properly represents the atmospheric distribution of CO_2 before convection and outside of the convective system afterwards. Direct measurements alone are insufficient to describe the spatial distribution, since CO_2 exhibits significant spatial variability during summer, over continents [*Gerbig et al.*, 2003a] (hereinafter referred to as G03a), and in-situ airborne measurements cannot span the entire domain.

[4] Here we use a receptor-oriented to extend observations in order to provide CO_2 initial fields and time-varying boundary conditions for a cloud-resolving model. An influence function is computed by tracking Lagrangian particles emitted in the mesh backward in time from the domain of the CRM, combined with estimates of the upstream tracer boundary condition, fossil-fuel derived emissions, and biospheric fluxes to simulate CO_2 fields to serve as input to the CRM. The CRM then redistributes the tracers through convective transport to produce concentration fields that can be compared to observations. The receptor-oriented framework is designed to resolve gradients in distributions of tracers like CO_2 [*Gerbig et al.*, 2003b] (hereinafter referred to as G03b) by predicting tracer concentrations at the receptors' point locations, minimizing the discrepancy between model gridcell-averaged values and concentrations at point locations (''representation errors'') [G03a] that can introduce biases into derived results.

[5] The method is appraised by: 1) comparing ROAM-simulated CO_2 distributions to measurements outside of clouds, and 2) evaluating simulations from the cloud-resolving model, driven with ROAM-derived initial and boundary conditions, against observed differences in CO_2 between air found inside and outside of clouds, to assess the fidelity of transport processes represented in the model.

2. CRYSTAL-FACE Observations

[6] CRYSTAL-FACE was a mission to study the transport and radiative effects of tropical convective and cirrus cloud systems. From 3 to 29 July 2002 sampling over the Florida region was carried out with in situ instruments onboard the NASA WB-57F and other aircrafts. The Harvard CO_2 sensor [*Daube et al.*, 2002] reported data throughout the duration of nearly each flight of the WB-57F (~8 hours), spanning altitudes from the PBL to the stratosphere, achieving precision of 0.17 ppm (1σ) traceable within 0.2 ppm to world standards maintained by CMDL. Water measurements were made using the Harvard total water and water vapor instruments [*Weinstock et al.*, 1994].

[7] Figure 1 shows part of (11~15 km) the time series of CO_2 and total water concentrations measured on 16 July 2002, a day with a large convective system. The CO_2 signal decreases during most cloud penetrations, as can be seen from the negative correlation with total water. Table 1 shows the measured differences between CO_2 concentrations in clouds (CO_{2cloud}) versus those outside of clouds (CO_{2out}), averaged over altitude bins for all flights. Clouds were identified as periods with more than 150% relative humidity or when total water reached more than 30% above the ice condensation threshold. For cases where insufficient water data were available, we used liquid water derived from the Cloud Aerosol and Precipitation Spectrometer [*Baumgardner et al.*, 2002]. Cloud-free data were identified as having relative humidity less than 90% or no detectable liquid water. To derive differences between CO_{2cloud} and CO_{2out}, periods of out-of-cloud measurements were aggregated into 1-km altitude bins, averaged for each bin, and linearly interpolated to in-cloud time periods. The average differences (CO_{2cloud} − CO_{2out}) shown in Table 1 are relatively small, but easily resolved by the measurement. Values are negative between 9 and 15 km (with peak difference of ~1 ppm at 10–11 km), indicating convective vertical transport from lower altitudes with depleted CO_2 from biospheric uptake. At altitudes above 15 km, a sign reversal is observed.

Table 1. Average Differences in CO_2 Concentrations (CO_{2cloud} − CO_{2out}) for All Flights During the CRYSTAL-FACE Campaign

Altitude (km)	9–10	10–11	11–12	12–13	13–14	14–15	15–16
$[CO_2]$ (ppm)	−0.44	−0.95	−0.36	−0.69	−0.24	−0.16	+0.35

3. Application of the Receptor-Oriented Atmospheric Modeling Framework

[8] The Receptor-Oriented Atmospheric Modeling (ROAM) framework [G03b] includes (Figure 2): (1) a backward-time Lagrangian particle transport model— the Stochastic Time-Inverted Lagrangian Transport (STILT) Model [*Lin et al.*, 2003] driven with assimilated meteorological fields (e.g., EDAS [*Black*, 1994]); (2) lateral tracer boundary conditions based on remote marine surface data; (3) fossil fuel fluxes of CO_2 from inventories; and (4) a simple biospheric flux model using eddy covariance data [*Baldocchi et al.*, 2001].

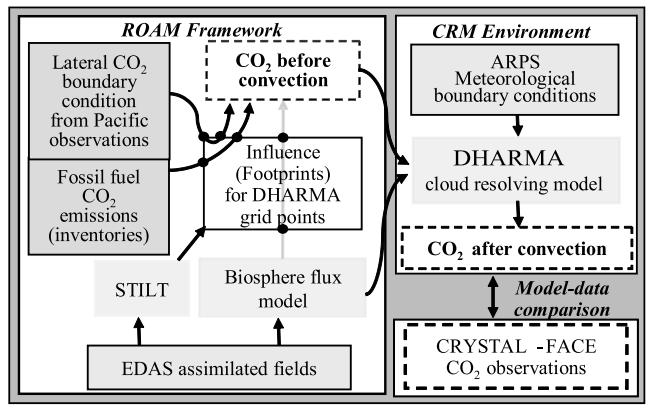

Figure 2. Schematic diagram showing the information flow between the Receptor-Oriented Atmospheric Modeling (ROAM) framework and the cloud-resolving model (DHARMA).

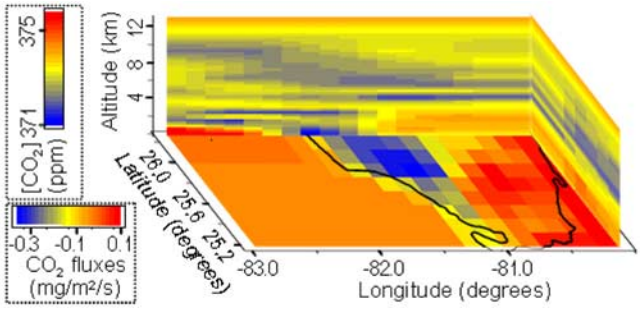

Figure 3. CO_2 boundary conditions provided by ROAM for the domain covered by the cloud-resolving model DHARMA, and surface fluxes (biospheric and fossil) within the domain (for 16 July 2002 at 18 GMT).

[9] ROAM computes CO_2 below 12 km based on inventory-derived fossil fuel emissions and optimized biospheric fluxes from G03b, as well as lateral tracer fields over remote marine regions. Because these lateral fields could be constructed only for the troposphere [G03b] (i.e., excluding stratospheric data), we obtained concentrations above 12 km for this study by simple linear interpolation of measured CO_{2out} profiles. ROAM was used to calculate hourly CO_2 mixing ratio at 300 locations (20 vertical, 5 longitudinal, 3 latitudinal; horizontal spacing ~50 km) within the CRM domain on 16 July (1500–2300 GMT). In addition, hourly surface fluxes were calculated for the CRM domain as surface boundary condition on a grid with spacings of 1/6° latitude and 1/4° longitude based on surface fluxes used in ROAM. Figure 3 shows these surface fluxes and the tracer boundary condition at 1800 GMT, interpolated to the CRM gridpoints.

[10] CO_2 fluxes show significant gradients related to variations in vegetation type (cropland, forest, shrubland, wetland, and open water) and fossil fuel emissions. Higher CO_2 emissions in the eastern half of the domain correspond to significant combustion in the Miami region. Note that in contrast to biospheric fluxes, the resolution of the fossil fuel inventory is only 1° by 1°, causing some emissions to appear over the ocean. Mixing ratios at the northern boundary of the CRM domain show influence from biospheric fluxes: e.g., CO_2 in the lower atmosphere at ~81.5°W is depleted after easterly winds transported air over an area with stronger photosynthetic uptake. To assess the quality of CO_2 mixing ratios calculated with this framework, we extracted the ROAM values at points closest in space and time to the observations made outside of clouds. The comparison between modeled and measured CO_2 shown in Figure 4 reveals good general agreement between 4 and ~12 km. Above 12 km discrepancies are expected, due to the lack of a stratospheric representation for CO_2. Below this level the root-mean-square error is only 0.46 ppm, suggesting that CO_2 values simulated by ROAM over the domain are realistic.

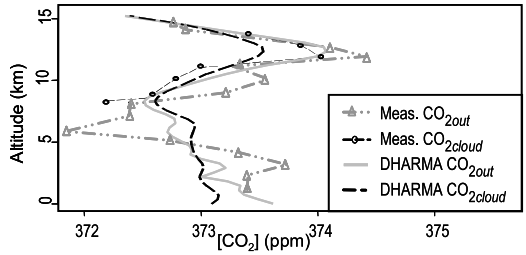

Figure 5. Profiles of CO_{2cloud} and CO_{2out} (18–21 GMT): comparison between measurements and DHARMA outputs.

4. Results From the Cloud-Resolving Model, Using ROAM-Derived Initial and Boundary Fields

[11] A cloud-resolving model with size-resolved microphysics, the Distributed Hydrodynamic-Aerosol-Radiation-Microphysics Application (DHARMA) [*Fridlind et al.*, 2004], was run to simulate transport within the 16 July convective system during its main stage of development (1500–2100 GMT). Meteorological initial and boundary conditions were derived from the Advanced Regional Prediction System (ARPS) [*Xue et al.*, 2003], a mesoscale forecasting system running at 15 km resolution. Initial and boundary CO_2 fields were taken from the ROAM runs. Figure 5 shows measured profiles of CO_{2cloud} and CO_{2out}, together with the corresponding model output averaged between 1800 and 2100 GMT and horizontally averaged over the entire model domain. A rigorous point-to-point comparison is not possible, since in this instance DHARMA did not place the convective system at the exact observed location, and cloud development in the model occurs ~1 hour prior to actual occurrence. The general shape of profiles within and outside of clouds is reproduced, however, with some inconsistencies near the tropopause (probably due to improper representation of stratospheric CO_2). If we compare the simulated (CO_{2cloud} − CO_{2out}) with the measured differences, the agreement is remarkably good (Figure 6). Measured differences have a minimum of −0.75 ppm at ~13.2 km, coinciding with the simulated

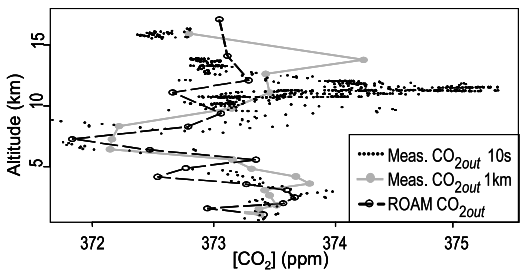

Figure 4. Altitude profiles of CO_2 outside of clouds (CO_{2out}): comparison between CRYSTAL-FACE (CF) observations and simulations by ROAM.

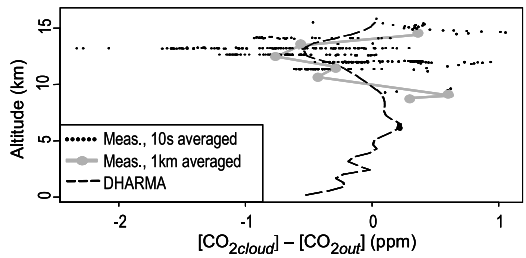

Figure 6. Profiles of CO_2 concentrations differences between in-cloud and out-of-cloud regions (18–21 GMT): comparison between observations and DHARMA results.

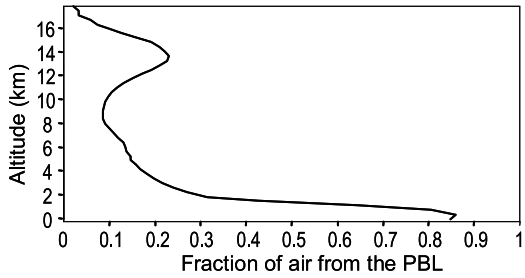

Figure 7. Fraction of in-cloud air at each elevation that originates from the planetary boundary layer (0–1.5 km elevation), as calculated by DHARMA for fully-developed deep convection on 16 July 2002 (18–21 GMT).

minimum of −0.53 ppm at 13 km. To diagnose the convective redistribution of air we introduced an artificial tracer within the cloud-resolving model. An artificial PBL tracer added to the July 16 simulation and redistributed with the modeled transport indicates that the predominant altitude of detrainment is 14 km, where ~25% of the cloudy air originated in the PBL (Figure 7).

5. Discussion

[12] Using atmospheric backtracking to determine initial and boundary conditions for cloud-resolving models provides a means to utilize information on convective transport contained in distributions of tracers such as CO_2. The receptor-oriented framework generally captures the spatio-temporal distribution of CO_2 outside of clouds, and the cloud-resolving model redistributes these fields in a way that is consistent with measured CO_2 differences between cloudy areas and cloud free ones.

[13] In order to better use tracer information in future missions, two improvements to the airborne sampling methodology are needed: (1) Characterizing the variability of tracers in airmasses upwind of the convective system, particularly in the PBL. Note that few data points were obtained outside of clouds during the campaign, and none at lower altitudes, within the PBL. (2) Choosing times and locations that exhibit large vertical gradients in tracer concentrations prior to convection to increase the signal available for constraining transport processes—e.g., in summertime continental airmasses for CO_2 [cf G03b].

[14] A receptor-oriented framework like ROAM can be driven using forecast meteorology to guide airborne sampling prior to convection, to optimize the constraints provided by tracers on transport. When forecast fields include convective mass fluxes, Lagrangian backtracking of anvil air can be used to guide measurements upstream of a storm system. Direct measurement of upstream CO_2 concentrations would significantly reduce uncertainties in ROAM simulations, which here used upstream tracer fields estimated from distant CMDL stations and past vertical soundings. Even when convective mass fluxes are not available in forecast fields, ROAM can still be used to forecast vertical tracer gradients in the environment outside of clouds, so that locations with enhanced gradients can be identified prior to the flights in order to provide larger signals to constrain vertical transport. Additional tracers such as CO, CH_4, and other long-lived species can be readily incorporated into ROAM, enhancing the information available to infer transport.

6. Conclusion

[15] We have shown that combining a receptor-oriented approach for tracer fields with a cloud resolving model for convective transport provides a framework for using tracer data to diagnose the distribution of vertical transport in convective clouds, overcoming the limited coverage of field data. The receptor-oriented framework can be strategically employed in field campaigns to predict locations of strong tracer gradients for planning sampling locations upstream of and within a convective system. A strong effort is currently being made by the carbon cycle community to utilize information available in atmospheric CO_2 measurements by using data assimilation systems [*Wofsy and Harriss*, 2002]. As these systems become available, estimates of surface fluxes become more reliable, and the resulting knowledge on the distributions of long-lived tracers like CO_2 will be highly beneficial to the interpretation of detailed measurements and to their utilization to infer transport processes.

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