Distribution of CO_2 and other tracers in convective and non-convective regions during CRYSTAL-FACE.

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Abstract

Measurements of CO_2, CO, O_3, NO_x, water vapor and total H_2O (vapor + condensed phase) were made in the upper troposphere on board of the WB-57F during CRYSTAL FACE. Air in the outflow of deep convective storms, and in associated cirrus clouds, showed distinctly different composition from surrounding air. On average concentrations of CO_2 in clouds at ~12 km were 0.4 ppm lower than surroundings. The magnitude of the anomaly varied directly with condensed-phase water content. Concentrations of NO_x showed very large enhancements, but CO was rarely elevated and ozone was reduced, implying a major role for lightning-generated production of odd nitrogen. This preliminary look at CRYSTAL FACE data shows that tracer data can provide quantitative measures of transport in convective clouds.

Objectives

•The goal of our work in CRYSTAL-FACE (Figure 1 & Figure 2) was to obtain CO_2 measurements that can constrain the mass fluxes of convective transport (Figure 3). $[CO_2]$ is modulated mainly by variations in photosynthesis and respiration at the Earth surface. Atmospheric sources or sinks from oxidation of hydrocarbons and CO are small, and can generally be accounted for using data for $[CH_4]$ and [CO]. All these properties make CO_2 a unique tracer for global transport (e.g. , Andrews et al., 2001a&b ; Boering et al.1996 ; Hintsa et al., 1998 ; Wofsy et al.,1994). A suite of tracers such as CO_2, CH_4, CO, and halocarbons may give a "fingerprint" of source regions for air convected to the troposphere. For example, convection in equatorial Brazil will have relatively low levels of urban pollutants, whereas updrafts over Indonesia or the Himalayas may sweep up industrial effluents.

•CO_2 and other tracers measurements can also be used to improve our understanding of the chemistry in the middle world region. For example, airborne measurements of CO_2 have been used to separate lightning produced NOx (Figure 3) from anthropogenic NOx transported vertically from the mixed layer [Huntrieser et al, 2002 ; Holler et al, 1999]. This is possible due to the strong gradients in CO_2 between the mixed layer and the free troposphere outside of convective clouds.

CRYSTAL FACE: The Cirrus Regional Study of Tropical Anvils and Cirrus Layers - Florida Area Cirrus Experiment. Funded by NASA.

Mission description : 6 planes, based in Key West (FL), have realized about 15 flights in July 2002, mainly over the Florida peninsula.Onboard instruments have measured gases and aerosols within cumulus and cirrus clouds during the lifetime of a convective system. Our CO2 sensor flew on board the WB-57 F plane, sampling mainly the upper troposphere, middle world and lower stratosphere regions with an accuracy reaching 0.17ppm.

Figure 1. Planes flight patterns

Figure 2. The WB-57 F

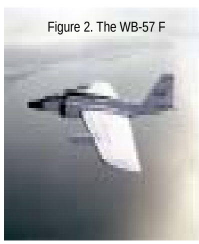
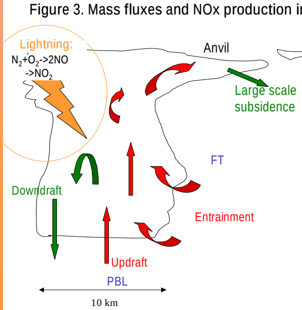


Figure 3. Mass fluxes and NOx production in a convective cloud.



• During its first stage, the convective system develops in the shape of cumulus clouds, entraining surrounding gases and moisture. In less than one hour, air from the planetary boundary layer (PBL) and from the free troposphere (FT) can be transported to the upper troposphere (UT) and even until the lower stratosphere (LS) within the developing convective systems. Once formed, the anvil later decomposes in cirrus clouds. Convective systems can thus play an important role in the transport of gases from the ground.

• NOy can be largely produced by lightning within the cloud. The quantification is poorly known.

Figure 4. Tracers altitude profiles averaged for July 2002. $[CO_2]$ measurements are compared to CMDL ground data (averaged between 15pm and 24pm): t=WLEF tower, TX ; h=mlo, Hawai ; b=rpb,Barbados ; w=bmw,West Bermudas ; k=key, Key Biscayne, FL.

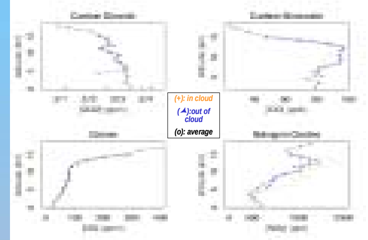
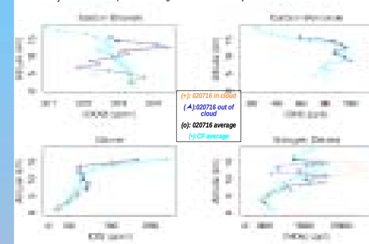


Figure 6. Tracers profiles in cloud and out of cloud for 16th of July (flight 020716). These are compared to averaged ones for whole July 2002.



Results

• CO_2, CO, O_3 and NO_y concentrations are in average not the same in cloud and out of cloud (Figure 4). A huge peak of [NOy] exists within the clouds.

• CO_2, CO and O_3 undergo slow chemistry compared to vertical transport in thunderstorms and can therefore be used as transport tracers to determine the amount of air that has been brought from the PBL and the FT by the convective system, and thus to quantify the NOy production by lightning only.

• Tracers ratios can give information on the likely sources of the air sampled within the cloud (easily characterized by water enhancement). The air sampled during CF within the anvils was usually coming from the marine PBL and from the free troposphere (Figure 5), and there is a clear in-situ NOy production by lightning. This is consistent with CMDL data (Figure 4).

• In the absence of stratospheric air masses, we can write for CO_2 and CO : $[Tracer]_{anvil}=f_{PBL}[Tracer]_{PBL} + f_{FT}[Tracer]_{FT}$. Plots of tracers concentration in function of total water amount (e.g. Figure 8) show asymptotic values for higher total water amount, that once stabilized is likely to be $[Tracer]_{anvil}$. $[Tracer]_{FT}$ can be associated to the tracer concentration outside of the cloud, allowing $[Tracer]_{FT}$ to be computed.

Figure 5. An air layer sampled on the 16th of July 2002.

➢$[O_3]$<100ppm => air layer does not come from the stratosphere ; it comes from the troposphere.

➢Negative correlation between [NOy] and [CO] => the air does not come from a polluted area.

➢Huge [NO] peak : indication of air freshness : it shows that at least part on the NOy peak has been produced within the cloud by lightning. Thus the other part comes from the marine PBL & the free troposphere.

A case study : anvil sampling, July 16th 2002

Figure 7 shows the cloud events encountered on July 16th. As shown on Figure 8, we make a rough hypothesis, that the "out of cloud" median $[CO_2]$ is equal to the FT $[CO_2]$, and that the "in cloud" asymptotic median $[CO_2]$ is representative of a mixture of FT air and PBL air concentrations ("anvil" value).

➢ Application to transport quantification from the PBL to the upper troposphere within convective systems.

•Satellite pictures of the flight track (Figure 9) show that the storm of events 1 to 3, sampled around 20:30 GMT, got formed in Miami area around 18:30 GMT and wind was blowing East. Thus the air sampled during "event 1" would likely partly come from the PBL around Miami.

•We used model results (Ann Fridlind et al., personal communication) to get the dilution factors between the PBL and the FT for the air sampled in the anvil, and to check consistency between our data and CMDL data : [anvil] = 0.4 [PBL]+0.6[FT].

•Application to $[CO_2]$ for events 2&3 : Anvil value is 373.3 ppm and FT value is 374.7 ppm => PBL value deduced is 371.2 ppm.

Conclusion 1: The combination of CF data and model results is consistent with Key Biscayne's CMDL data (Figure 10) two hours before the sampling (371.55 ppm), i.e. roughly when and where the storm was forming.

➢Application to NOx production by lightning.

•Using the dilution factors presented above, the NO_x budget within the convective system can be written : $[NO_x]_{anvil}=0.4\ [NO_x]_{PBL}+0.6\ [NO_x]_{FT}+[NO_x]_{lightning}$.

•NOx measurements do not exist for the PBL in areas relevant of non polluted and marine areas. From Figure 6, it seems reasonable to make the assumption that [NOx] in the PBL had an upper limit of 1000ppt.

•From Figure 8, we get respectively for event 2 and event 3 : $[NOx]_{anvil}$ ~ 5900 ppt to ~7100 ppt and $[NOx]_{FT}$ ~ 600 ppt to ~700 ppt.

⇒We can thus deduce : $[NOx]_{lightning}$ ~ 5140 ppt (event 2) to ~6280 ppt (event 3).

Conclusion 2: With these assumptions, on July 16th 2002, lightning contributed to produce ~88 % of the NOx within large anvils. This is consistent with the results obtained in huge thunderstorms over Europe (Huntrieser et al, 2002), for which NOx production by lightning has been evaluated to exceed 80%.

Figure 7. [CO2], total water and altitude time profiles for 020716 Tropospheric air only * (The cloud events are numerated in red).

* Filter used : $[O_3]$<100ppm, Water vapor>20ppm, Potential Temperature<355K

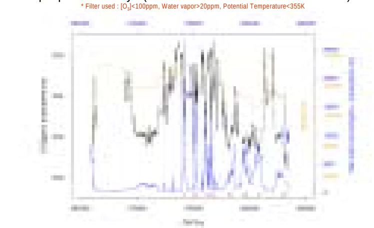
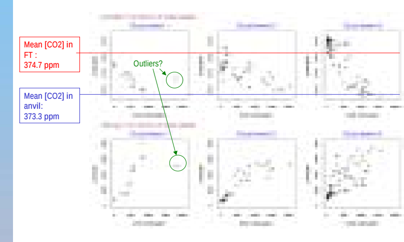


Figure 8. $[CO_2]$ and [NOy] in function of [total water] for events 1, 2 and 3.



Conclusion and perspectives

CRYSTAL FACE tracers concentrations are significantly different inside and outside of cloud. As showed in this preliminary work, these discrepancies should allow us to determine the origin and amount of the air masses updrafted by convection from the planetary boundary layer and from the free troposphere, as well as the related NOx sources, especially the NOx production by lightning. A case study for the 16th of July 2002 gave promising results by coupling data with preliminary modeling results. A deeper collaboration with modelers will be undertaken in the next weeks. CRYSTAL FACE CO_2 altitude profiles will as well be used to attempt to get information on the so-called "rectifier effect".

Figure 9. SATELLITE PICTURES for flight 020716 :

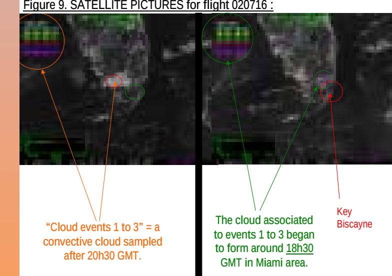


Figure 10. Map of Florida and Key Biscayne.

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