Emissions of trace gases and their atmospheric transport above Southern and Eastern Asia

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1. CO concentrations above Southern and Eastern Asia measured by the MOPITT instrument

1.1 Monthly means (annual cycle) – comparison with detected fires and population density

a) 850 hPa average CO values for January, April, July and October 2006:

→ clear differences between the months representing the four seasons
→ pronounced annual cycle in CO concentration structures and biomass burning (similar for 2005 – not shown here)

→ permanent high CO pollution over Eastern and South-Eastern Asia detected

January: highest pollution in South-Eastern China near Hong Kong and Guangdong (densely populated area [urban], detected fires in this area); high CO values also above the Indochinese Peninsula (due to biomass burning)

April: huge areas with very high CO concentrations (from the Indochinese Peninsula along the Chinese coast up to North-Eastern China and also above Siberia); much biomass burning in Southern Asia contributes to high CO values in the South-East; along the Chinese coast biomass burning is not the main contributor (rather urban areas); Siberian high CO concentrations mainly due to biomass burning

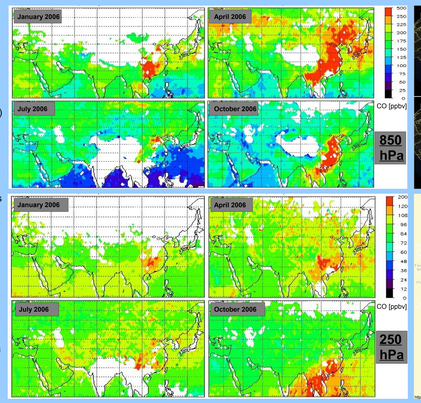
July: lowest CO concentrations in the annual cycle; still high concentrations of CO near Chinas megacities (Beijing, Tianjin, Chongqing); biomass burning generally low during summer

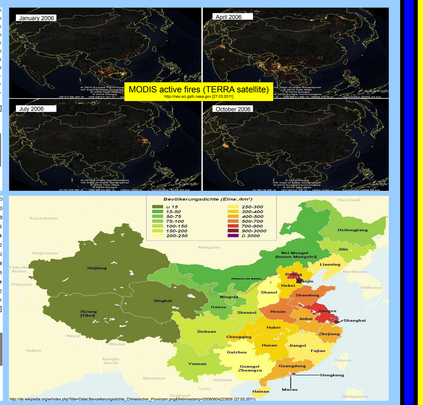
October: high CO maximum stretching from the Indochinese Peninsula in the south up to the Beijing metropolitan area in the north again along the densely populated coastal area – biomass burning with supposed low contribution
* Note the huge fire in the north-west of India which is investigated in the case study (→ part 3)

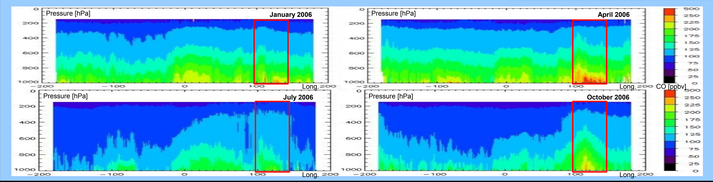
b) 250 hPa average CO values for January, April, July and October 2006:

→ evidence of local vertical transport of pollution (convection) – CO concentration patterns on 850 hPa and 250 hPa show structures which are alike to a big extent

→ evidence of transport of pollutants in direction of the Pacific Ocean (advection) (↔ Intercontinental transport)







1.2 CO concentrations compared on a global scale

Global cross sections (180°W to 180°E) for the months of January, April, July and October 2006 – indicated values are CO concentrations averaged along the degrees of longitude between 10°N (south of the Indochinese Peninsula) and 42°N (north of Beijing metropolitan area, including other megacities in the world) and they are averaged for the whole month

Examples of megacities and metropolitan regions which are located in that band (but outside of the red box in the diagram): Los Angeles (118°W), Mexico City (99°W), New York (74°W), Istanbul (29°E), Cairo (31°E), Baghdad (44°E), Tehran (51°E), Karachi (67°E), Mumbai (73°E), Delhi (77°E), Kolkata (88°E), Dhaka (90°E)

Megacities and metropolitan regions located within the red box: Chinese (Beijing, Shanghai, Hong Kong), Korean (Seoul, Incheon) and Japanese (Tokyo, Osaka) metropolitan regions, as well as Bangkok (Thailand) and Manila (Philippines)

Indochinese Peninsula with high emissions of CO due to biomass burning is also included in the red box

→ All year round the Eastern Asian region is the area on the whole globe with the strongest CO pollution, reaching highest into the troposphere

2. CARIBIC canister measurements of CO and hydrocarbons on flights Frankfurt ↔ Guangzhou ↔ Manila (whole year 2006)

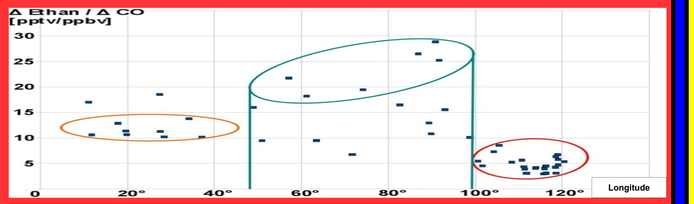
All canisters which are classified as 'polluted' (CO > 85 ppbv) and which have tropospheric characteristics (Ozone < 200 ppbv) are used in this analysis comparing enhancement ratios of Ethane to CO and Propane to Benzene. As reference a canister with an unpolluted sample taken over Central Asia in October 2006 is used. All the concentration levels of canisters which match the above mentioned characteristics and which are sampled on a height of above 8000 m are inserted into the following equation describing the enhancement ratios:

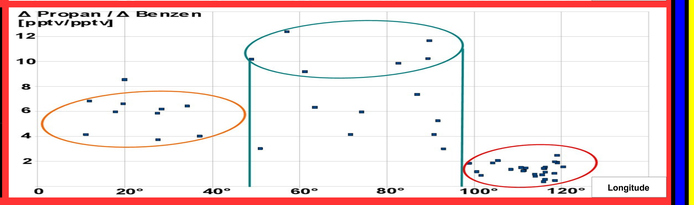
$$\frac{\Delta X}{\Delta Y} = \frac{X(can) - X(backgr)}{Y(can) - Y(backgr)}$$

Enhancement ratios of these 2006 canisters are plotted over the longitude of their sampling.

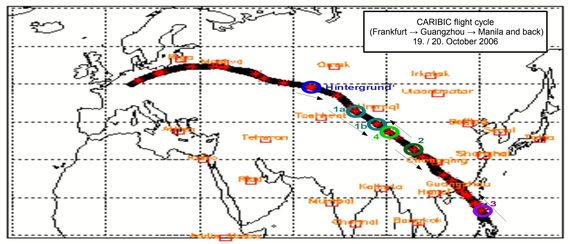
→ The result is that 3 regions with characteristic ratios can be observed:
- European emissions between Frankfurt and Moscow (orange coloured)
- Central Asian emissions between the Ural Mountains and Western/Central China (green coloured)
- Eastern Asian emissions between Central China and the Philippines (red coloured)

Part 1 (MOPITT) gives information about emission sources in Eastern Asia; CARIBIC observes regularly high concentrations of the alkanes above Central Asia and Western China leading to high enhancement ratios in Ethane to CO and Propane to Benzene (↔ investigated in the case study [part 3])





3. Casestudy on the CARIBIC flight cycle – 19th and 20th of October 2006



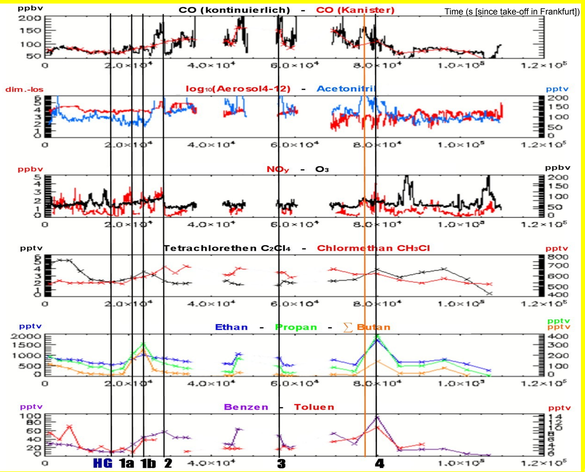
Numbers and 'Hintergrund' ('background') indicate the position of sampling of the canisters on the map. Additionally the canisters are marked in the time-trace gas concentrations diagram.

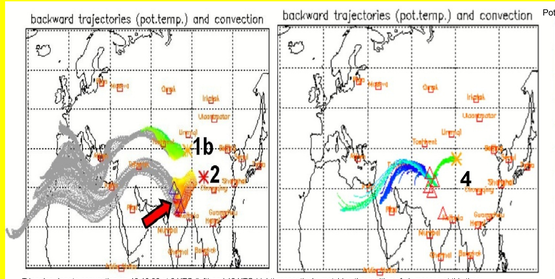
	Background	Canister 1a	Canister 1b	Canister 2	Canister 4
Time of Sampling (20.10.2006 UTC)	01:15	02:40	03:22	04:47	18:59
Ozone [ppbv]	54.86	63.24	81.28	71.83	74.55
Carbon Monoxide [ppbv]	71.56	83.14	87.57	107.86	109.76
Ethane [pptv]	543.95	860.32	1022.99	856.10	1717.22
Propane [pptv]	51.82	219.14	309.73	121.73	385.00
i-Butane [pptv]	4.38	53.01	84.43	15.85	51.01
n-Butane [pptv]	9.20	112.50	166.67	24.92	91.12
Σ Butane [pptv]	13.58	165.51	251.10	40.77	142.13
Benzene [pptv]	9.14	27.16	44.50	57.63	92.11
Toluene [pptv]	1.51	1.40	5.69	NaN	10.17

Enhancement rations (Equation → see part 2):

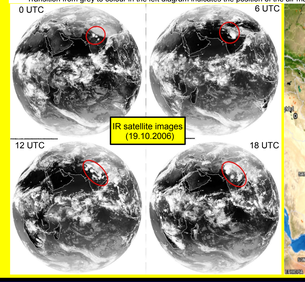
	Canister 1a	Canister 1b	Canister 2	Canister 4
Δ Ethane / Δ CO [pptv/ppbv]	27.32	29.92	8.60	30.71
Δ Propane / Δ CO [pptv/ppbv]	14.45	16.11	1.93	8.72
Δ ΣButane / Δ CO [pptv/ppbv]	13.12	14.84	0.75	3.37
Δ Benzene / Δ CO [pptv/ppbv]	1.56	2.21	1.34	2.17
Δ Ethane / Δ Propane [pptv/pptv]	1.89	1.86	4.47	3.52
Δ Propane / Δ Benzene [pptv/pptv]	9.29	7.29	1.44	4.02

→ Canisters 1a and 1b show similar characteristics indicating that both stem from the same pollution source; Canister 2 shows completely different characteristics; Canister 4 (collected on the flight back and regarding the location sampled between the positions of 1 and 2) exhibits values which lie between those of canisters 1 and canister 2





Triangles denote convection on 19.10.06 at 6 UTC (left) and 12 UTC (right) respectively, matching the position of air masses at this time.
Transition from grey to colour in the left diagram indicates the position of the air masses at the same time; in the right diagram the cross marks the current position



Canister 2: - backward trajectories cross a big area of strong convection which was persistent on the 19th of October in Northern India transporting air masses from the lower to the upper troposphere
- intense fires detected from 17th to 19th of October in the same location

	Canister 2 (M*)	Canister 2 (AC*)	ER BB*	ER BF*	MER BB/BF*
Δ Ethane / Δ CO [pptv/ppbv]	8.60	8.42	10.8	14.3	5.8 – 8.8
Δ Propane / Δ CO [pptv/ppbv]	1.93	2.03	0.9	4.1	1.0 – 1.6
Δ Benzene / Δ CO [pptv/ppbv]	1.34	2.24	1.3	4.4	No estimate

* (M: measured; AC: age corrected to emission ratio; ER BB: emission ratio biomass burning (Baker et al. 11); ER BF: emission ratio biofuel burning (Baker et al. 11); MER BB/BF: measured enhancement ratio in 1-2 days old plumes strongly influenced by both BB and BF (Lai et al. 10))

→ Canister 2 is strongly influenced by biomass burning with a possible contribution of biofuel burning

Canister 1 a/b: - very high concentrations in the short lived alkanes (butanes, but also pentane and hexane [not shown in the table])
→ fresh emissions (Central Asian or Western Chinese)
- biomass burning as main source can be excluded (not found along the backward trajectory, different signature as Can. 2)
- high enhancement ratios in i.e. ethane to CO and propane to benzene → the observed characteristic Central Asian emissions (from part 2)

- high emissions of alkanes, especially propane and butane, and observed enhancement ratios connected to petrochemistry, processing of natural gas, mineral oil and petroleum (Carmichael et al. 03; Wong et al. 07) – suits with industry structure and resource usage in that area

Canister 4: - shows characteristics of both, petrochemistry emissions and biomass/biofuel burning
- trajectories touch the convective complex associated with the fires

References: Baker et al. 11, Characterization of non-methane hydrocarbons in Asian summer monsoon outflow observed by the CARIBIC aircraft, Atmos. Chem. Phys., 11, 503-518; Carmichael et al. 03, Evaluating regional emission estimates using the TRACE-P observations, Journal of Geophysical Research, Vol. 108, No. D21, 8810; Lai et al. 10, Pollution events observed during CARIBIC flights in the upper troposphere between South China and the Philippines, Atmos. Chem. Phys., 10, 1649-1660; Wong et al. 07, Impact of Asian continental outflow on the concentrations of O3, CO, NMHCs and halocarbons on Jeju Islands, South Korea during March 2005, Atmospheric Environment 41 (2007) 2933-2944