Large Scale Variations of Greenhouse Gas Concentrations Related to Meteorological Conditions during the Airborne Atmospheric Composition Measurements

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Abstract: Concentrations of CH_4, CO_2, O_3, CO, H_2, H_2O were measured continuously at the altitude of about 7 km during long distance (up to 3000 km) horizontal transit flights on board the IL-18 research airplane in July 1993. Tracer concentration changes as airplane crosses different air masses, it appeared to be possible to distinguish at least two types of air masses: one from stratosphere is characterized by high ozone, low humidity, carbon monoxide, and methane. Another type is characterized by lower ozone, hydrogen, carbon dioxide and higher methane concentration and humidity. Small difference can be observed in the carbon monoxide concentration in the air masses originating from lower troposphere. In addition to tropopause folding event on July 18 north of Yakutsk, an upward transport event was observed in July 28, 1993 during meridianal flight along the 60°N latitude. In those cases simultaneous change in the concentrations of different tracers clearly shows the origin of the air mass - either from stratosphere or from the lower troposphere. The air mass origin could be decided based on positive and negative correlation between different tracer concentrations and meteorological parameters. Atmospheric composition observations agree well with the variation of meteorological parameters derived from JMA global analyses (large scale features) and those measured on board.

1. Introduction

Airborne atmospheric composition measurements appear to be an effective tool for studying tracer transport by dynamic processes, such as stratosphere -tropospheric exchange rate (Danielsen, 1968). Those studies were focused largely on ozone, at the same time cycles of greenhouse gases like methane and nitrous oxide are also affected by the upper atmospheric dynamics. *In situ* measurements

of greenhouse gases distribution in the upper troposphere are valuable reference sources for species transport studies and models of atmospheric composition. As for the large scale upper air composition data only a few studies has been reported so far which involve measurements of methane and other greenhouse gases. Methane and other tracers were observed during STRATOZ (Marenco, 1988, Schmidt, 1982) and ABLE3 experiments (Harris, 1992). As a result of those experiments it was established, that the tracer concentration are subject to a great influence by dynamic processes. At high latitudes the stratospheric air intrusions affect about one third of air samples (Browell, 1993). It appeared rather difficult even to differentiate between air masses of different origin without extensive meteorological analysis and comprehensive atmospheric composition measurements (Marenco, 1988). Perturbation of the tracer concentration field by mesoscale dynamic processes usually complicate derivation of generalized meridianal and latitudinal profiles. Generalized or average profiles are necessary for verification and adjustment of GCM based chemical tracer transport models. With the development of three-dimensional models which are based on observed winds (Taylor et al., 1993), high spatial and temporal resolution data become more valuable.

Main objectives of joint Russian-Japanese expedition on airborne measurements of greenhouse gases over Siberia were to obtain surface flux data and receive large scale concentration distributions. During the expedition 5 long-distance flights were made with full set of chemical and meteorological parameters measured on board. Simultaneous acquisition of several atmospheric tracer concentrations has been proved to be extremely useful by a number of airborne observation studies in upper troposphere. Treated separately, atmospheric tracers are subject to different source, sink and transport influences. Thus it is difficult to assign major factor which led to observed concentration variation. As it can be seen from the results of this study, several tracers taken together become more reliable indicators of transport, emission and transformation processes.

Transit flights were performed along the following route: Sendai (38°N, 141°E) - Khabarovsk - Yakutsk (62°N, 129°E) - July 15; Yakutsk - Tiksi (72°N, 128°E) and back - July 18; Yakutsk - Nizhnevartovsk (61°N, 76°E) - July19; Nizhnevartovsk -Yakutsk, July 28; Yakutsk - Sendai, July 30. Also, carbon dioxide measurements were performed, on the way from Moscow (56° N, 38° E) to Sendai through Nizhnevartovsk and Yakutsk (July 6 - 8) and back (August 2 - 3). On July 18th tropopause folding event was observed, it is discussed in below.

2. Concentration Distributions Obtained during Long Range Transit Flights

Measurements and data corrections were done by principal investigators and are reported in accompanying papers. Here we present summary of horizontal profiles obtained by several instruments. Latitudinal and longitudinal distributions taken in two long- range flight transects are presented here as illustration: *a*) Yakutsk-Tiksi-Yakutsk, July 18, and *b*) Nizhnevartovsk-Yakutsk, July 28.

Tracer gas distribution during the first flight are shown on Fig. 1. Negative correlation between humidity and ozone concentration is apparent at several moments of high ozone concentration. High ozone and low humidity usually correspond to air masses of stratospheric origin (Danielsen, 1968). At the same time negative correlation can be seen between methane and ozone concentrations. Low CO_2 and high CH_4 correspond to air masses coming from lower troposphere, where methane is produced and carbon dioxide is consumed by vegetation. That consideration is also backed up by ozone concentration which goes down when methane is up. A horizontal scale of the unhomogeneity is around 0.5 - 1.0 degree latitude. Generally this ozone folding event shows several typical features of tropopause folding. Japan Meteorological Agency 1.875 degree global analysis data set was used for preparation of pressure, temperature, potential vorticity and adiabatic vertical velocity maps. Also, those values were interpolated in time and space to the points along the flight route. Maps of potential temperature, surface pressure and back trajectory analysis are in a good agreement with observations given the low dataset resolution. Meteorological dataset proved to be helpful in understanding larger scale features that are discussed with respect to next episode. A correlation between methane and ozone concentration during the folding phenomena can be expressed as

$$\delta CH_4 = -0.6 \bullet \delta O_3$$

Tracer fields on Fig. 2 suggest that there exists strong upward flow from lower troposphere around the 90 - 100°N with horizontal scale of about 500 km. At that time concentrations of methane, ozone, hydrogen and carbon dioxide had were close to the values that are typical for lower troposphere. The scale of that event is larger than the resolution of the JMA global analysis data, so the data are suitable for comparison and model verification. A number of large scale features is in qualitative agreement.

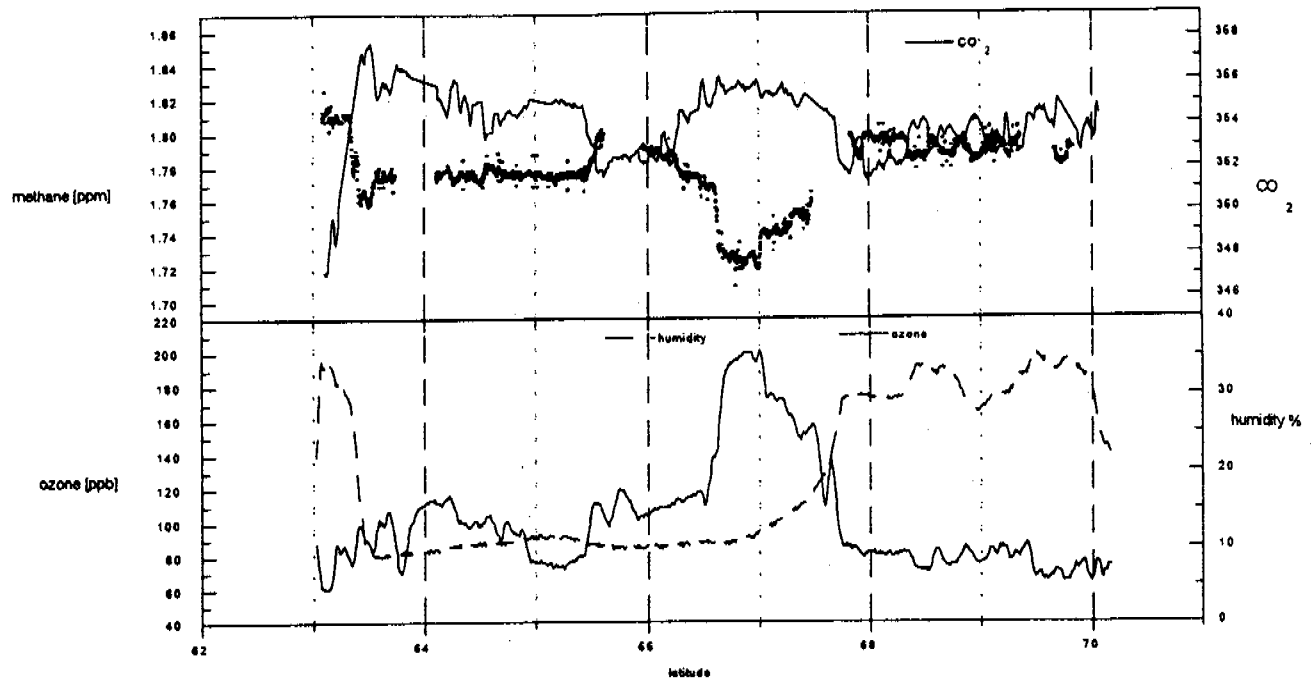

Fig. 1 Methane, carbon dioxide, ozone and humidity along flight route with respect to latitude. Flight from Tiksi area to Yakutsk, July 18, 1993. Straight lines on methane and carbon dioxide plots correspond to calibration periods, when the atmospheric concentration is not measured.

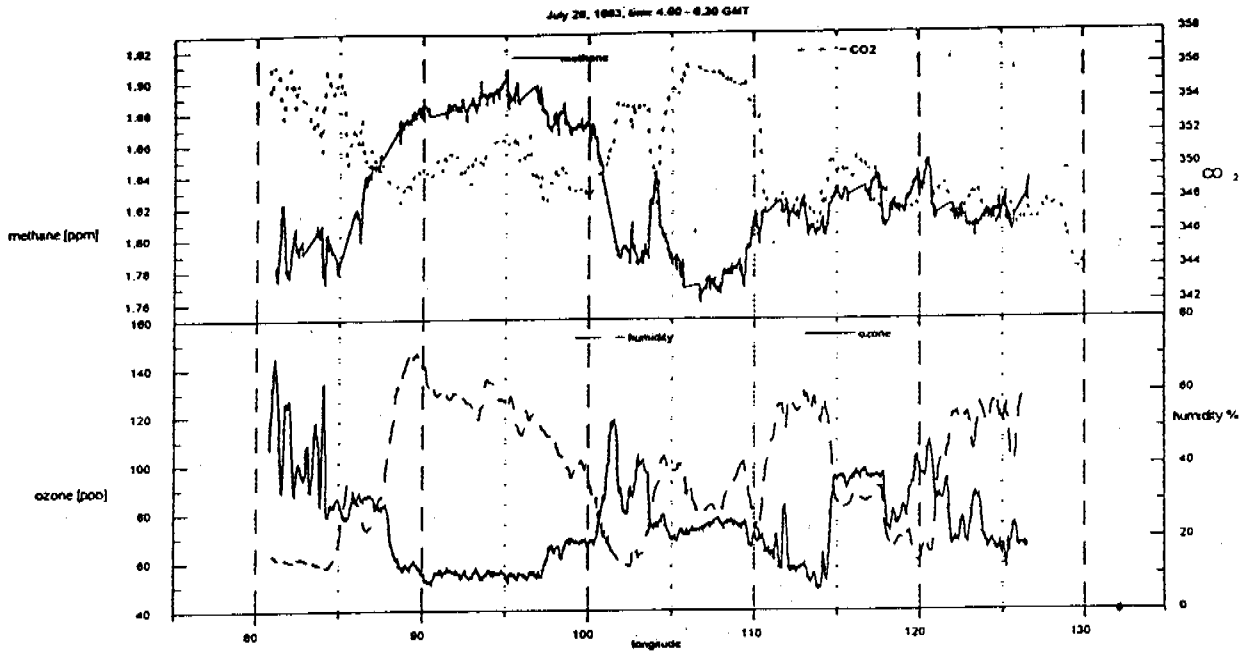

Fig. 2 Methane, carbon dioxide ozone and humidity along flight route with respect to longitude. Flight from Nizhnevartovsk to Yakutsk, along 61°N latitude line, July 28, 1993.

Vertical wind velocity fields derived from the global JMA analyses for 12 GMT on July 28 show two maxima along the flight route between 90 and 120° N occupying large area, so it is evident that the air mass did ascend to the area of measurement from the surface layer below it. Potential vorticity profile has two maxima on Fig. 2, while ozone (accompanied by low humidity) has two peaks shifted 5 degree to the west. A discrepancy between time and position of potential vorticity maxima and ozone peak could probably be explained by more detailed analysis or higher resolution meteorological model.

3. Conclusions

Methane, carbon dioxide and other tracer's distributions show significant correlation within separate air mass volumes. That makes it possible to discriminate air masses into at least three categories: air of stratospheric origin, well mixed upper tropospheric air and air transported from the lower troposphere and PBL. Large scale atmospheric concentration unhomogenieties caused by atmospheric dynamics where observed, the scale being large enough for using that data in the global tracer transport model experiments for verifying the emission and transport mechanism.

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