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VERTICAL PROFILE OF KRYPTON-85 IN THE STRATOSPHERE

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Abstract. A vertical profile of Krypton-85 from 12 to 40 km height was obtained from a balloon-borne experiment. The results display a clear concentration gradient between the troposphere and 22 km. Above this altitude, the concentration varies very little, in agreement with theoretical predictions.

Introduction

Stratospheric ozone is directly affected by changes in the concentrations of trace gases, which are injected into the lower atmosphere by human activities. The concentrations of these gases result from both atmospheric transport and chemical reactions, in connection with solar rays. The kinetics of these reactions are far from being known with the desirable accuracy. However parametrizations of the vertical transport are generally not introduced in the models as independent data, but are calculated from the distribution of chemical species. Therefore, it should be particularly useful to make use of an independent tracer which is not involved in the whole chemical system. This is precisely the interest of Krypton-85 measurements.

Remember that Krypton-85 is injected into the lower troposphere essentially by the reprocessing of nuclear fuels. As a noble gas, it is chemically inert and disappears from the atmosphere only by radioactive decay, with a half-life of 10.76 years. Its concentration has been monitored in the troposphere since 1950 (Weiss et al., 1983; Dvorak and Wilhelmsova, 1984). The annual increase is presently of the order of $2.15 \times 10^{-2} \text{ Bq.kg}^{-1}\text{yr}^{-1}$, i.e. about 3.5% per year (Weiss et al., 1986). At the end of 1986 it was approximately 0.70 Bq.kg^{-1} in the troposphere.

Some rare measurements of Kr-85 were obtained from time to time in the lower layers of the stratosphere by Pannetier (1968), Telegadas and Ferber (1975), and recently by Weiss and collaborators (unpublished results). However, to our knowledge, no measurement has ever been published at altitudes of 28-40 km, for which vertical transport data are particularly lacking. The aim of this paper is to report one Kr-85 vertical profile in this altitude range.

Sampling and measurements

A balloon-borne experiment was launched from Aire-sur-l'Adour (France) on November 16th 1986, during a period of very strong winds in the upper stratosphere.

The sampled air was first compressed by a mechanical booster pump, and then by a one stage

rotary pump. Oil and water vapours were removed in a liquid nitrogen cooled trap, and the air was blown through one trap filled with activated charcoal, and maintained at 120 K by liquid nitrogen. The sampling device comprised five such independent traps selected by valves operated by remote control. At the end of the line the air pressure was automatically controlled at a value of about 10^6 Pa for which the sampling efficiency is optimal.

After recovering the sampler the air, already enriched in rare gases, was desorbed by heating and the krypton of the air was again trapped in cold charcoal and then separated and measured by gas chromatography. This krypton was subsequently introduced into a beta counter whose volume was completed by a classical Argon-Methane mixture, in order to measure the Kr-85 activity. The background of this counter is 0.55 pulse per minute and its efficiency is 78%. As the krypton mixing ratio of 1.14×10^{-6} is constant in air throughout the middle atmosphere, the knowledge of the volume of krypton introduced into the counter gives the amount of air to which the Kr-85 activity must be ascribed.

Four samples were collected during the flight. A fifth trap was maintained closed during the flight as a blank, in order to detect the presence of any contamination.

Discussion and conclusion

The results of the measurements are shown in Table 1. As a comparison, the values calculated from the vertical transport coefficients proposed by Chang (1976) are also indicated.

The activity of the blank was not detectable. The decay of the Kr-85 concentration versus altitude is very clear above the tropopause situated at 11.58 km height as well as from 15 to 22 km (fig.1). The concentration is practically constant between 30 and 40 km, which implies either very rapid vertical mixing at these altitudes as proposed by Chang (1976) and Crutzen et al (1976), or an advection from other parts of the stratosphere in agreement with Telegadas and Ferber (1975) and the theoretical considerations of Holton (1986). However between the tropopause and 22 km height, the concentration decrease is significantly more rapid than expected from the 1D model proposed by Cariolle (1980).

Because of the small number of experimental results, it is impossible to calculate transfer coefficients, but only an "apparent" age of the air masses, defined as the time span since their entry into the stratosphere. This age varies from 2.3 year in the lower stratosphere to about 8 years in the upper stratosphere.

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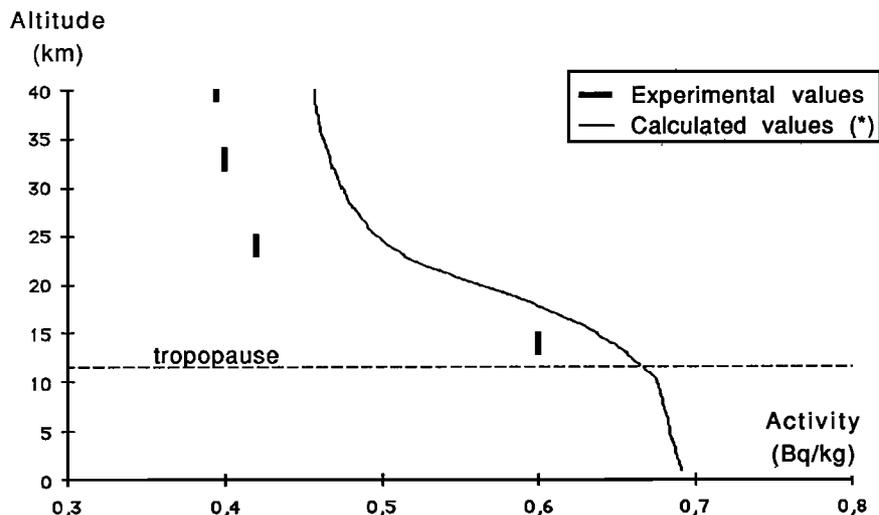
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TABLE 1. Vertical profile of krypton-85

Altitude hPa (km)	Air volume sampled TPN litre	Activity Bq/kg of air ($\pm 1\sigma$)	Apparent age (year)	Calculated activity (*) Bq/kg of air
186 - 117 (12.26 - 15.2)	10.0	0.60 ± 0.02	2.3	0.65
38.4 - 26.2 (22.35 - 24.8)	28.3	0.42 ± 0.01	7.8	0.51
10.07 - 6.64 (30.75 - 34)	8.4	0.40 ± 0.02	8.6	0.47
3.77-2.84 (38 - 40)	18.1	0.395 ± 0.005	8.8	0.46

(*) Values calculated from the vertical transport coefficients proposed by Chang (1976) and from the lower tropospheric Kr-85 concentration since 1950 (Dvorak and Wilhelmova, 1984; Weiss et al., 1986) with the 1D model proposed by Cariolle (1980).



(*) Values calculated from the vertical transport coefficients proposed by Chang (1976) and from the lower tropospheric Kr-85 concentration since 1950 (Dvorak and Wilhelmova, 1984; Weiss et al., 1986) with the 1D model proposed by Cariolle (1980).

Fig. 1. Observed and calculated Kr-85 profiles on November 16th, 1986.

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