Recycling of Bomb Produced ^{36}Cl

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The radioactive nuclide ^{36}Cl ($T_{1/2} = 3 \cdot 10^7$ years) is naturally produced in spallation reactions of high energy cosmic rays on the atmospheric argon. Another important source of ^{36}Cl in nature is its anthropogenic production due to nuclear bomb tests in the 50-60th of the last century. However the time profile of ^{36}Cl obtained in an ice core in Greenland [1] has shown that because of the fast atmospheric wash-out of chlorine no bomb ^{36}Cl should be observable in modern precipitation. So excluding some possible local contamination the ^{36}Cl fluxes on the Earth were assumed to reflect the rate of production of ^{36}Cl and thus to be a cross-check for models of nuclear reactions in the atmosphere due to cosmic rays.

The experiment to measure ^{36}Cl in modern precipitation has been performed and has revealed significantly higher fluxes than it was expected [2]. The observed discrepancy can be explained if part of the bomb produced chlorine was captured by the biosphere and released back into the atmosphere in the form of methyl chloride with a time delay. CH_3Cl has a life time of about 1.1 years in the atmosphere and thus it can be well-mixed over the globe. It decays under reactions with OH radicals, which concentration is higher at low latitudes than in the pole regions and over the continents than over the ocean. Because of that the recycled chlorine should play an important role at equatorial belt and should be hardly noticeable at pole ice caps (e.g. Greenland).

To check the hypothesis:
- The bomb ^{36}Cl input had to be better determined.
- The averaged over a few years latitudinal distribution of ^{36}Cl fluxes had to be measured with better precision.
- The direct measurements of ^{36}Cl in atmospheric CH_3Cl had to be performed.

Lake	Natural	Chernobyl	Bomb	Total	Measured
Constance	1	19		20	38 ± 3
Geneva	1	53	6	60	61 ± 4
Garda	2	44	25	71	69 ± 11
Como	2	26		28	28 ± 4
Lugano North	2	51	9	62	79 ± 7
Lugano South	2	29	5	36	31 ± 6
Maggiore	2	17	1	20	18 ± 4
Onega	3	53	19	75	77 ± 9
Ilmen	43	81		124	121 ± 21
Ladoga	4	52	26	82	78 ± 9
Baikal	8		25	33	24 ± 7
Victoria	2		31	33	30 ± 9
Mweru	0.3			0.3	6 ± 2
Malawi	3		5	8	13 ± 3
Chiemsee	1			1	36 ± 3
Starnberger	3	50	35	88	69 ± 6
Ammersee	2	3		5	41 ± 7
Walchensee	1			1	23 ± 3

Table 1: The ^{36}Cl concentration estimated due to different sources and measured in lakes, in $\frac{at}{cm^3}$

The first two tasks were achieved by ^{36}Cl measuring in lakes with different flushing times. The flushing time (the ratio of volume to outflow rate of a lake) shows the typical time, over which the water is kept and mixed (averaged) in the lake. If this time is of about few years or less there is no bomb ^{36}Cl left in the lake and the measured concentration can be recalculated to the mean ^{36}Cl fluxes over the catchment area. If this time is of about few decades or centuries the most of the ^{36}Cl has the bomb origin.

The stable chlorine concentration was measured by High Performance Liquid Chromatography and the ratio of ^{36}Cl/Cl was obtained using the AMS facility in Garching. The results together with the calculated concentration of ^{36}Cl due to different sources are presented in Table 1.

European lakes contain much ^{36}Cl originated from Chernobyl accident and can be used to study the distribution of this contamination. In the lakes with long flushing time the bomb ^{36}Cl prevails and the data are in a good agreement with the results of our modeling. The lakes with short flushing times contain more ^{36}Cl than it could be expected if recycling didn't take place. However this effect can be also explained by the influence of ground water, which can increase the effective time of water renewal.

The collection of the methyl chloride was performed by pumping air through molecular sieve [3]. A chemical treatment to prepare sample, suitable for AMS measurements was applied to this molecular sieve and for the background determination also to ones through which no air and through which commercial CH_3Cl was pumped. The results of the measurements are presented in Table 2.

Sample	Mass Cl, mg	Ratio of ^{36}Cl/Cl, 10^{-13}
Cl in the air	4.3	29 ± 3
Commercial CH_3Cl	261	0.19 ± 0.09
Molecular sieve only	1.1	1.2 ± 0.4

Table 2: The measurements for ^{36}Cl determination in CH_3Cl

Considering the typical annual chlorine turnover in the form of CH_3Cl to be $2.5 \cdot 10^{12}\ \frac{g}{a}$ we get the minimal estimate for global average flux of the recycled ^{36}Cl 7.5 $\frac{at}{m^2 s}$. This can be compared with the natural global average rate of ^{36}Cl production of about 20 $\frac{at}{m^2 s}$. The contribution of the recycled component to the ^{36}Cl flux should be especially pronounced over the continents and at low latitudes. In fact the role of recycled ^{36}Cl can be much higher. A part of the measured chlorine comes from molecular sieve and has lower ratio ^{36}Cl/Cl. Moreover only methyl chloride with a typical concentration 665 ppt contains much of ^{36}Cl and the used method was sensitive to all organic molecules containing Cl, which concentration is of about 4250 ppt. With the corrections for these effects we get the upper limit for the mean flux of recycled ^{36}Cl 62 $\frac{at}{m^2 s}$.

As a conclusion we can say that the measurements in lakes indicate the existence of bomb ^{36}Cl recycling in the nature and the measurement of CH_3Cl proves it.

References

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