



An excess of ^{36}Cl in modern atmospheric precipitation

A. Blinov ^{a,*}, S. Massonet ^b, H. Sachsenhauser ^b, C. Stan-Sion ^c, V. Lazarev ^{a,b}, J. Beer ^d,
H.-A. Synal ^e, M. Kaba ^f, J. Masarik ^g, E. Nolte ^b

^a Department of Cosmic Research, State Technical University St. Petersburg, Polytechnicheskaya 29, St. Petersburg 195251, Russia

^b Technical University of Munich, Munich, Germany

^c Institute of Nuclear Physics and Engineering, Bucharest, Romania

^d Federal Institute for Environmental Science and Technology (EAWAG), Switzerland

^e Institute for Intermediate Physics, ETH-Hoenggerberg, Switzerland

^f University of Ivory Coast, Abidjan, Ivory Coast

^g Komensky University, Bratislava, Slovakia

Abstract

Measurements of ^{36}Cl were performed in modern annual precipitation from many world-wide distributed stations. The measured deposition fluxes were on the average half an order of magnitude larger than fluxes calculated from the cosmic-ray-induced atmospheric production. These discrepancies were tentatively explained by recycling of bomb ^{36}Cl from the biosphere as methyl chloride (CH_3Cl) into the troposphere, destruction of CH_3Cl by OH reactions and subsequent tropospheric deposition of ^{36}Cl . Simulations were performed for natural atmospheric production, bomb fall-out and recycling of ^{36}Cl . The simulations including recycling were qualitatively able to explain the excess in deposition fluxes, especially for tropical regions. © 2000 Elsevier Science B.V. All rights reserved.

1. ^{36}Cl in the modern environment

There are two dominant sources of the long-lived radionuclide ^{36}Cl ($T_{1/2} = 301,000$ yr) in the modern environment. ^{36}Cl is naturally produced in the atmosphere by cosmic-ray-induced nuclear reactions mainly on atmospheric argon [1]. The important reactions are $^{40}\text{Ar}(n,p4n)^{36}\text{Cl}$ and $^{40}\text{Ar}(p,2p3n)^{36}\text{Cl}$. The production cross-sections for the reactions $^{40}\text{Ar}(p,2p3n)^{36}\text{Cl}$ and $^{40}\text{Ar}(p,\alpha n)^{36}\text{Cl}$

were measured in the energy range 17–1000 MeV [2,3]. These results were used to calculate the mean global atmospheric production rate of ^{36}Cl to be 24 atoms $\text{m}^{-2} \text{s}^{-1}$ [2], which is close to 19 atoms $\text{m}^{-2} \text{s}^{-1}$ obtained earlier in [4] and more recently in [5]. ^{36}Cl produced in the stratosphere is transported through the tropopause into the troposphere essentially at mid-latitudes. From the troposphere, ^{36}Cl is removed as wet or dry deposition. Removed ^{36}Cl follows the geochemical chlorine cycle and is incorporated into natural archives such as polar ice or groundwater [6].

An additional natural source of ^{36}Cl is in situ production in the lithosphere. The nuclear reactions are induced by cosmic-ray neutrons and

* Corresponding author. Tel.: +7-812-5526480; fax: +7-812-5527954.

E-mail address: blinov@phtf.stu.neva.ru (A. Blinov).

muons. This source could potentially contribute to the tropospheric ^{36}Cl flux by surface erosion processes. In rock material on the Earth's surface, ^{36}Cl is mainly produced by neutron-induced spallation reactions on potassium and calcium and neutron capture reactions on ^{35}Cl . For vanishing erosion rates, the surface saturation ratios were obtained to be 3.6×10^{-15} for $^{36}\text{Cl}/\text{Ca}$ [7] and 4.8×10^{-15} for $^{36}\text{Cl}/\text{K}$ [8]. For a mean global erosion rate of $60 \mu\text{m}/\text{yr}$ [9], the surface saturation concentrations are about two orders of magnitude lower [10]. With lithospheric abundances of 3.65% for Ca and 2.58% for K, the ^{36}Cl flux due to eroded land is $0.3 \text{ m}^{-2} \text{ s}^{-1}$. This estimate shows that erosion in situ produced ^{36}Cl cannot give a globally significant contribution to the atmospheric ^{36}Cl flux.

The human nuclear activities established an anthropogenic source of ^{36}Cl in the environment. Bomb tests on barges in the fifties added a fluence to the atmosphere which was equivalent to more than thousands of years of natural atmospheric production. In polar ice cores of Dye 3 (Greenland, 65°N), the ^{36}Cl bomb pulse [11,12] was measured to have a fluence (time integrated flux) of $2.4 \times 10^{12} \text{ atoms m}^{-2}$. The fact that the troposphere is effectively cleaned of anthropogenic ^{36}Cl can be seen from the measured time profile of the ^{36}Cl bomb pulse in ice. However, if part of the bomb ^{36}Cl is stored in the biosphere and re-enters the troposphere after a mean residence time of several tens of years, the tropospheric fall-out would be increased. The storage of bomb ^{36}Cl in the biosphere was first proposed by [13] and later qualitatively applied in [14]. The compound to be considered re-entering the troposphere is methyl chloride (CH_3Cl), the most abundant natural organic chlorine-containing gas in the atmosphere [2,15–17]. It is naturally produced by biogenic processes on land and in the ocean, and anthropogenically by biomass burning. The main sink of CH_3Cl is its oxidation by free OH radicals in the troposphere [17]. Tropospheric OH concentrations are highest in tropical regions, higher in the northern than in the southern hemisphere and higher on land than on sea [18].

For the global ^{36}Cl fall-out, the Chernobyl accident in 1986 was a local event since the radioactivity did not reach the stratosphere for world-

wide distribution and was washed out from the troposphere within weeks. In the Munich area, concentrations of 6400 Bq of ^{137}Cs [19] and of $1.6 \times 10^{11} \text{ }^{36}\text{Cl}$ atoms/l were measured in rainwater collected on 30 April 1986. Using the total ^{137}Cs release from this accident [20], the total ^{36}Cl release can be estimated to be about $2 \times 10^{24} \text{ }^{36}\text{Cl}$ atoms, corresponding to approximately five years of global natural atmospheric production.

In Section 2, the sampling procedure, the sample preparation, measurements and experimental ^{36}Cl concentrations are presented. In Section 3, atmospheric transport models for ^{14}C , ^{90}Sr and ^{36}Cl are discussed. The experimental data are compared with modelled ^{36}Cl fall-out fluxes and fluences and observed discrepancies are explained by recycling of bomb-produced ^{36}Cl from the biosphere. In the conclusion, possible experiments to test the proposed hypothesis are considered.

2. Experimental procedure and results

Annual atmospheric precipitation samples were collected at 28 meteorological stations with geographical latitudes between 45°S and 80°N . The collecting procedure did not distinguish between wet and dry deposition. The samples were transported in liquid form to Technical University of Munich, State Technical University St. Petersburg and EAWAG Duebendorf for chemical preparation. The chloride concentration in water was measured by column ion chromatography and partly by neutron activation analyses. In two cases only, Kankan (Guinea) and Fargernes (Norway), chlorine carrier had to be added to the initial water. Each sample was filtered and evaporated down to a volume of about 15 ml. Chlorine was extracted by a standard chemical procedure in the form of AgCl with a typical yield of about 80%. Seven samples were subdivided into two parts and independently analysed at TUM and ETH Zurich.

The $^{36}\text{Cl}/\text{Cl}$ ratios were measured by AMS technique at the accelerator laboratories of Munich and ETH/PSI. Descriptions of the experimental facilities and details of measuring procedures can be found elsewhere [12,21]. The measured $^{36}\text{Cl}/\text{Cl}$ ratios were in the range between

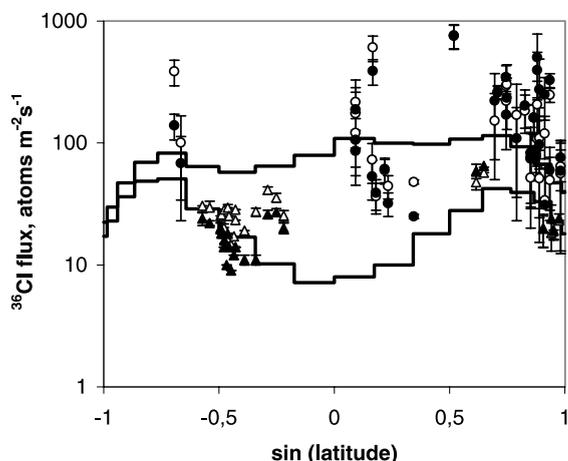


Fig. 1. Measured ^{36}Cl deposition fluxes (full circles), local precipitation corrected fluxes (open circles) together with measured (full triangles) and precipitation corrected ^{36}Cl fluxes (open triangles) of other groups [31–35] in comparison to deposition fluxes of cosmic-ray-produced ^{36}Cl (lower histogram) calculated with the model described in Sections 3.1 and 3.2. The upper histogram includes recycling of bomb ^{36}Cl .

1.2×10^{-14} and 1.8×10^{-12} . Blank samples prepared identically had $^{36}\text{Cl}/\text{Cl}$ ratios below 2×10^{-14} and typically in the range of 10^{-15} . The blank values were subtracted from the ratios measured for precipitation samples. The measured ^{36}Cl concentrations lie in a wide range between 1 and 31 million atoms/l. Neither a clear latitudinal pattern nor a dependence on the precipitation rate is observed in the present data. The deduced experimental ^{36}Cl deposition fluxes are shown in Fig. 1 together with data from other groups. Experimental details on the stations, locations, precipitation rates, chloride concentrations and ^{36}Cl fluxes are given in [14]. From Fig. 1 it is seen that there is an excess of ^{36}Cl in the measured fluxes over the natural fall-out predictions.

3. Comparison with simulation calculations

3.1. Atmospheric transport model and rate constants

In order to describe intensity and latitudinal dependence of the measured ^{36}Cl deposition fluxes, a model for atmospheric input, transport and

fall-out was developed. The atmosphere was divided into 18 latitudinal belts of 10° each. Every belt was subdivided into four stratospheric layers of equal air mass and one tropospheric layer. Most of the rate constants describing the exchange between the boxes are determined by steady-state conditions. A few basic rate constants were fixed by simulating time, latitudinal and altitude dependencies of fall-outs and atmospheric concentrations of the ^{14}C , ^{36}Cl and ^{90}Sr bomb pulses [22]. The division of the stratosphere into four layers was done in order to describe the different stratospheric residence times for gaseous compounds as e.g. $^{14}\text{CO}_2$ and H^{36}Cl and aerosol bound radionuclides as e.g. ^{90}Sr [22] which are subjected to gravitational settling.

Details of the model for an ordinary latitudinal belt are shown in Fig. 2 for ^{36}Cl and ^{90}Sr (left) and for ^{14}C (centre). In addition to exchange between the atmospheric layers, the aerosol bound ^{90}Sr is subjected to gravitational settling so that the downward transport of ^{90}Sr is faster than that of ^{14}C and ^{36}Cl . For $^{14}\text{CO}_2$, exchange with the mixed and deep ocean layers has to be taken into account. Figs. 3 and 4 show the results of simulations for the time dependence of the ^{14}C and the latitudinal dependence of the ^{90}Sr bomb pulses, respectively, in comparison to measured data [23,24].

Time constants (inverse rate constants) were obtained around 2 weeks (4 weeks) for the exchange between neighbouring tropospheric (stratospheric) latitudinal belts, and around 4 weeks (8 weeks) across the equator (see Fig. 2). The exchange between the four stratospheric layers was adjusted with time constants of 4.5 yr (layer 1 \rightarrow layer 2), 1.5 yr (layer 2 \rightarrow layer 3) and 0.5 yr (layer 3 \rightarrow layer 4). A mean time constant of 0.3 yr was used for the transport from the stratospheric layer 4 into the troposphere. The stratospheric gravitational settling of ^{90}Sr was described with a mean aerosol radius of $0.3 \mu\text{m}$ and stratospheric distances (see Fig. 2). The transport from the troposphere into the stratosphere takes place in tropical regions between 30°N and 30°S with seasonal latitudinal variation [6]. The latitudinal ^{90}Sr fall-out distribution was obtained from the latitudinal dependence of the transport from the stratosphere into the troposphere and from the

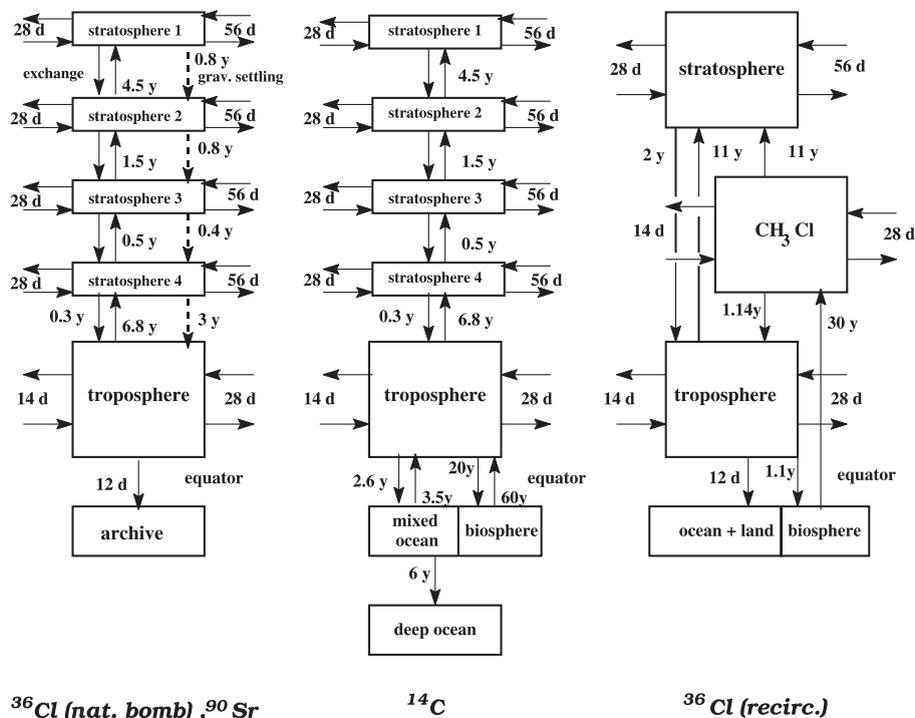


Fig. 2. Atmospheric transport model for natural and bomb ^{36}Cl and for bomb ^{90}Sr (left), for bomb ^{14}C (centre) and for recycling of bomb ^{36}Cl via the biosphere into the troposphere (right). Gravitational settling in the stratosphere is only used for ^{90}Sr . Shown are the details for one latitudinal belt of 10° width. The shown time constants are average values. The time constants are latitude dependent. The transport through the tropopause has a seasonal dependence.

mean precipitation rates in the latitudinal belts. The transport into the troposphere was found to be strongest in the regions of the polar and subtropical jet streams, i.e., of the troposphere folds. This downward transport was obtained to be at maximum in summer and vanished in winter with a sinusoidal time dependence.

For ^{90}Sr and ^{36}Cl , tropospheric fall-out was described by wet and dry deposition. Using tropospheric residence times from [25] and estimates for dry deposition [6], the rate constant for tropospheric deposition was presented in a form $P(\lambda)/(19\text{d} + 1/34\text{d})$ (m/yr)/19d + 1/34d, with the mean precipitation rate $P(\lambda)$ in m/yr in the considered latitudinal belt λ and a mean global precipitation rate of 1.0 m/yr.

For $^{14}\text{CO}_2$, the following time constants were used: troposphere \rightarrow mixed ocean layer 2.6 yr; mixed ocean layer \rightarrow troposphere 3.5 yr; mixed ocean layer \rightarrow deep ocean 6 yr; troposphere \rightarrow biosphere 20 yr; biosphere \rightarrow troposphere 60 yr.

3.2. Production, transport and deposition of natural ^{36}Cl

In [2], the mean cosmic-ray-induced production rate of ^{36}Cl was calculated to be about 24 atoms/cm² s. The latitude and altitude dependence of the production rate was taken from [5]. The ^{36}Cl fall-out fluxes obtained with the model of Section 3.1 are plotted as lower histogram in Fig. 1.

The fluxes derived from the ^{36}Cl measurements have to be corrected for the local precipitation rates and then compared with the model predictions. Assuming that the longitudinal mixing is fast against tropospheric deposition, this correction is given by

$$\frac{P(\lambda) \text{ (m/yr)/19d} + 1/34\text{d}}{(P_{\text{local}}(\lambda) \text{ (m/yr)/19d} + 1/34\text{d})}, \quad (1)$$

where $P_{\text{local}}(\lambda)$ (m/yr) is the local precipitation rate.

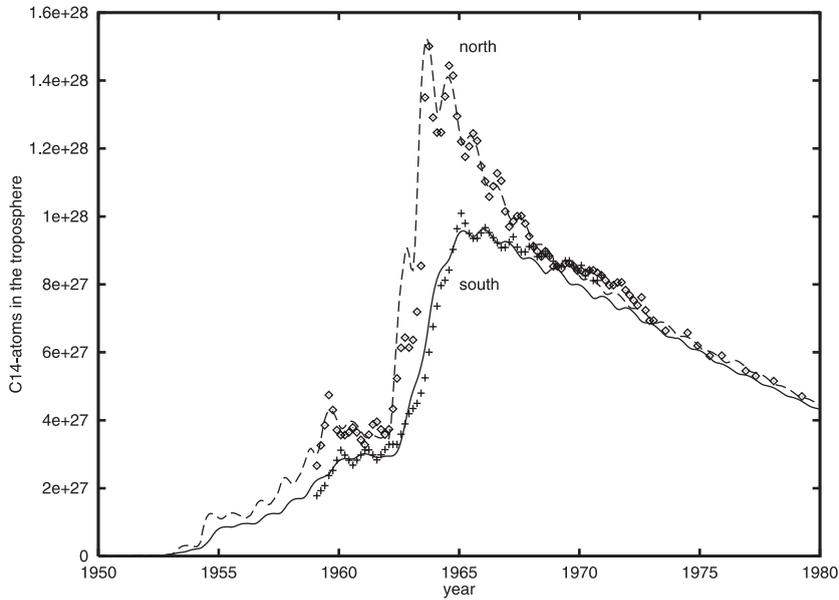


Fig. 3. Time dependence of the ^{14}C bomb pulse (northern hemisphere: full line; southern hemisphere: broken line) simulated with the model of Fig. 2 (centre) in comparison to measured data [23].

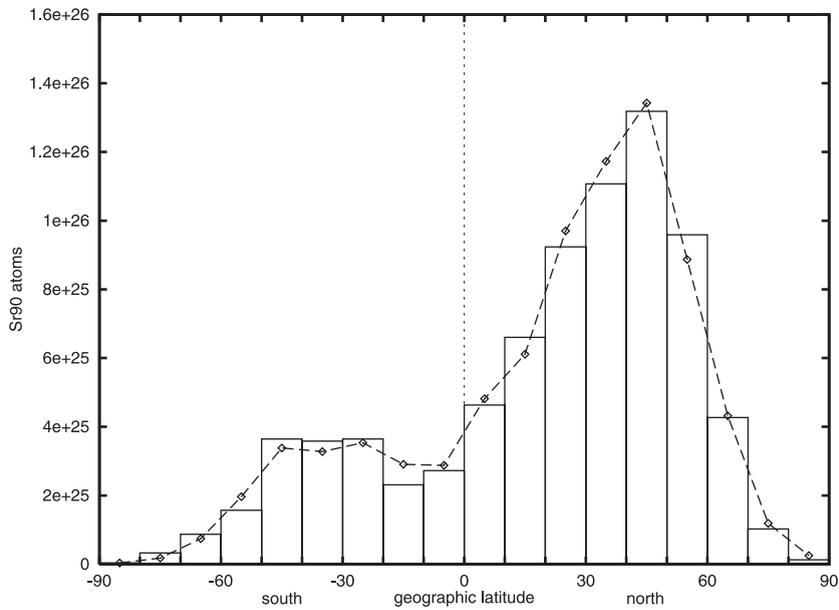


Fig. 4. Latitudinal dependence of the ^{90}Sr bomb pulse (circles) simulated with the model of Fig. 2 (left) including gravitational settling in the stratosphere compared to measured data [24] (histogram).

For high latitudes, the agreement between measurements and simulation becomes better after the precipitation correction is applied. However, a

discrepancy remains, especially for tropical regions, where the measured values are up to one order of magnitude larger than the simulation results.

A mechanism which could explain this discrepancy is storage of bomb ^{36}Cl in the biosphere, re-entering into the troposphere as biogenic CH_3Cl , destruction by OH reactions and subsequent tropospheric deposition.

3.3. Simulations of the ^{36}Cl bomb pulse

As described in [22], the explosions on barges at Bikini and Eniwetok atolls in the years 1954, 1956 and 1958 were the main sources of bomb ^{36}Cl . The stratospheric input distribution was chosen to be the same as for ^{90}Sr : layer 1 (10%); layer 2 (70%); layer 3 (20%). With these input data and with a normalisation of a total deposit of 5×10^{26} ^{36}Cl atoms corresponding to a mean fluence of 0.98×10^{12} ^{36}Cl atoms/ m^2 , the latitudinal ^{36}Cl fall-out distribution was calculated (see Fig. 5). According to this simulation, 61% are deposited in the Northern and 39% in the Southern hemisphere.

These simulations were compared with measured fluences of the ^{36}Cl bomb pulse at Dye 3 [12], Fiescherhorn, Guliya and Huascarán [26] and with

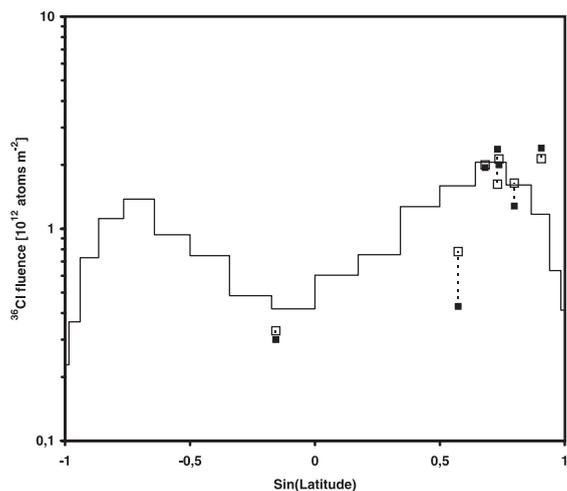


Fig. 5. Latitudinal dependence of the ^{36}Cl bomb pulse fluence (histogram) simulated with the model of Fig. 2 (left) together with measured fluences (full squares) of Dye 3 (65°) [12], Fiescherhorn (47°) [26], Guliya (35°) [26] and Huascarán (-9°) [26] and fluences derived from ^{36}Cl concentrations in Lake Baikal (around 53°) [27], Lake Superior (around 48°) [28] and Lake Michigan (around 43°) [28]. Precipitation corrected fluences are plotted as open squares.

those we derived from measured ^{36}Cl concentrations in Lake Baikal [27], Lake Superior and Lake Michigan [28] after subtraction of natural ^{36}Cl contributions. The fluences corrected for local precipitation rates with (1) are reasonably well described by the simulations. The low fluence at Guliya could be due to losses of snow and the large Dye 3 fluence could be caused by local tropospheric transport processes different from those used in the model of Section 3.1.

3.4. Recycling of bomb ^{36}Cl via the biosphere

The model used to describe the recycling of bomb ^{36}Cl via the biosphere including its injection into the troposphere as methylchloride, its destruction by OH radicals and tropospheric deposition is shown in Fig. 2 (right). In the simulation, 1/8 of the bomb ^{36}Cl fall-out over land between 60°S and 60°N was put into the biosphere. The mean biospherical residence time was assumed to be 30 yr. CH_3Cl is an organic chlorine compound produced in biological processes as e.g. biomass burning [15,16]. It is destroyed by OH reactions with a mean lifetime of 1.14 yr [29]. In the simulation, this lifetime was weighted with the latitudinal dependence of tropospheric OH concentrations which were taken from [30]. OH concentrations are largest in tropical regions and larger in the northern than in the southern hemi-

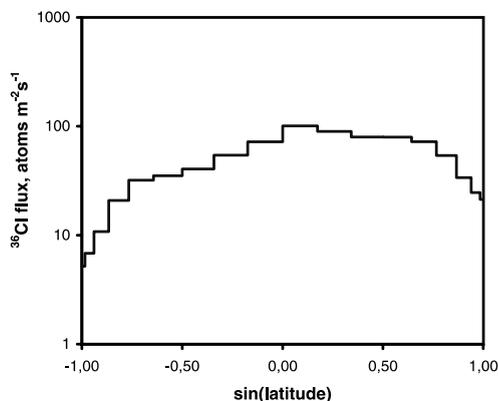


Fig. 6. Latitudinal dependence of the ^{36}Cl deposition fluxes due to recycling of bomb ^{36}Cl from the biosphere as CH_3Cl into the troposphere, simulated with the model of Fig. 2 (right).

sphere [18]. After destruction of CH_3Cl , ^{36}Cl is removed by tropospheric deposition. The mean time constant for the transport from the stratosphere into the troposphere was taken to be 2 yr.

Fig. 6 shows the additional ^{36}Cl deposition fluxes obtained in this simulation. These fluxes are highest for tropical regions and larger for the northern than for the southern hemisphere. For the belt $0\text{--}10^\circ\text{N}$, this additional flux is as large as $100\text{ atoms/m}^2\text{ s}$. When this flux is added to the natural fall-out (upper diagram in Fig. 1), it is seen that this mechanism would be able to describe the observed discrepancies shown in Fig. 1.

4. Conclusion

Measured ^{36}Cl deposition fluxes show considerable discrepancies compared to natural fluxes due to cosmic-ray-induced atmospheric production. These discrepancies are especially pronounced in tropical regions. These ^{36}Cl deposition fluxes in excess to natural atmospheric production are tentatively explained by storage of part of bomb ^{36}Cl in the biosphere, reemission into the troposphere in the form of CH_3Cl and subsequent tropospheric deposition. Simulations with ^{36}Cl bomb fall-out including this recycling process yield highest deposition fluxes for low northern latitudes and are able to explain qualitatively the measured fluxes.

Experiments to support the above hypothesis as e.g. measurements of ^{36}Cl in atmospheric CH_3Cl and in water of lakes with short residence times from tropical, mid-latitude, and high-latitude regions are in progress.

Acknowledgements

This work was supported by DFG, BMBF and the scientific-technical cooperation WTZ. We would like to thank Dr. M. Lawrence (Max Planck Institute for Chemistry, Mainz, Germany) for sending the OH data before publication. Three of us (A. B., C. St. and V. L.) would like to thank the German WE Heraeus foundation for supporting

their participation in the AMS-8 conference (Vienna, 1999).

References

- [1] D. Lal, B. Peters, *Handbuch der Physik*, Vol. 46/2, Springer, Berlin, 1967, p. 551.
- [2] D. Huggle, A. Blinov, C. Stan-Sion, G. Korschinek, C. Scheffel, S. Massonet, L. Zerle, J. Beer, Y. Parrat, H. Gaeggeler, W. Hajdas, E. Nolte, *Planet. Space Sci.* 44 (1996) 147.
- [3] C. Stan-Sion, D. Huggle, E. Nolte, A. Blinov, M. Dumitru, *Nucl. Instr. and Meth. B* 117 (1996) 26.
- [4] A. Blinov, *Secular Solar and Geomagnetic Variations in the Last 10,000 years*, Reidel, Dordrecht, 1988, p. 329.
- [5] J. Masarik, J. Beer, *J. Geophys. Res.* 104 (D10) (1999) 12099.
- [6] T.E. Graedel, P.J. Crutzen, *Atmospheric Change. An Earth System Perspective*, Freeman, New York, 1993.
- [7] B. Dockhorn, S. Neumaier, F.J. Hartmann, C. Petitjean, H. Feastermann, E. Nolte, *Z. Phys. A* 341 (1991) 117.
- [8] J.M. Evans, J.O.H. Stone, L.K. Fifield, R.G. Cresswell, *Nucl. Instr. and Meth. B* 123 (1997) 334.
- [9] C.-F. You, T. Lee, L. Brown, J.J. Shen, J.-C. Chen, *Geochim. Cosmochim. Acta* 52 (1988) 2687.
- [10] B. Heisinger, M. Niedermayer, F.J. Hartmann, G. Korschinek, E. Nolte, G. Morteani, S. Neumaier, C. Petitjean, P. Kubik, A. Synal, S. Ivy-Ochs, *Nucl. Instr. and Meth. B* 123 (1997) 341.
- [11] D. Elmore, L.E. Tubbs, D. Newman, X.Z. Ma, R. Finkel, K. Nishiizumi, J. Beer, H. Oeschger, M. Andree, *Nature* 300 (1982) 735.
- [12] H.-A. Synal, J. Beer, H.J. Bonani, M. Suter, W. Woelfli, *Nucl. Instr. and Meth. B* 52 (1990) 483.
- [13] J.C.D. Milton, H.R. Andrews, L.A. Chant, R.J.J. Cornett, W.G. Davies, B.F. Greiner, Y. Imahori, V.T. Koslowsky, J.W. McKay, G.M. Milton, *Nucl. Instr. and Meth. B* 92 (1994) 440.
- [14] C. Scheffel, A. Blinov, S. Massonet, C. Stan-Sion, H. Sachsenhauser, J. Beer, H.-A. Synal, P.W. Kubik, M. Kaba, E. Nolte, *Geophys. Res. Lett.* 26 (10) (1999) 1401.
- [15] M.A.K. Khalil, R.M. Moore, D.B. Harper, J.M. Lobert, D.J. Erickson, V. Koropalov, W.T. Sturges, W.C. Keene, *J. Geophys. Res.* 104 (D7) (1999) 8333.
- [16] J.M. Lobert, W.C. Keene, J.A. Logan, R. Yevich, *J. Geophys. Res.* 104 (D7) (1999) 8373.
- [17] W.C. Keene, M.A.K. Khalil, D.J. Erickson, A. McCulloch, T.E. Graedel, J.M. Lobert, M.L. Aucott, S.L. Gong, D.B. Harper, G. Kleiman, P. Midgley, R.M. Moore, Ch Seuzaret, W.T. Sturges, C.M. Benkovitz, V. Koropalov, L.A. Barrie, Y.F. Li, *J. Geophys. Res.* 104 (D7) (1999) 8429.
- [18] M.O. Andreae, P.J. Crutzen, *Science* 276 (1996) 1052.
- [19] J. Heinzl, G. Korschinek, E. Nolte, *Phys. Scr.* 37 (1986) 314.
- [20] OECD/NEA, *Chernobyl Ten Years on Radiological and Health Impact*, Paris, 1996.

- [21] P.W. Kubik, G. Korschinek, E. Nolte, *Nucl. Instr. and Meth. B* 1 (1984) 51.
- [22] L. Zerle, T. Faestermann, K. Knie, G. Korschinek, E. Nolte, J. Beer, U. Schotterer, *J. Geophys. Res. Atmos.* 102 (1997) 19517.
- [23] I. Levin, K.O. Münnich, W. Weiss, *Radiocarbon* 22 (1980) 379.
- [24] UNSCEAR Rep, To the General Assembly, UN Science Committee on the Effects of Atomic Radiation, New York, 1982.
- [25] G.M. Raisbeck, F. Yiou, M. Fruneau, J.M. Loiseaux, M. Lieuvin, J.C. Ravel, *Geophys. Res. Lett.* 8 (1981) 1015.
- [26] H.A. Synal, Workshop Glaciers from the Alps: Climate and Environmental Archives, Wengen, Switzerland, 1996.
- [27] T.M. Beasley, L.W. Cooper, J.M. Grebmeier, L.R. Kilius, H.-A. Synal, *Environ. Sci. Technol.* 31 (1997) 1834.
- [28] J.C.D. Milton, G.M. Milton, H.R. Andrews, L.A. Chant, R.J. Cornett, W.G. Davies, B.F. Grainer, Y. Imahori, V.I. Koslowsky, T. Kotzer, S.J. Kramer, J.W. McKay, *Nucl. Instr. and Meth.* 123 (1997) 382.
- [29] R. Koppmann, F.J. Johnen, C. Plass-Dulmer, J. Rudolph, *J. Geophys. Res.* 98 (1993) 20517.
- [30] M.G. Lawrence, P.J. Crutzen, P.J. Rasch, B.E. Eaton, N.M. Mahowald, *J. Geophys. Res.* 104 (D21) (1999) 26245.
- [31] D.L. Knies, D. Elmore, P. Sharma, S. Vogt, R. Li, M.E. Lipschutz, G. Petty, J. Farrell, M.C. Monaghan, S. Fritz, E. Agee, *Nucl. Instr. and Meth. B* 92 (1994) 340.
- [32] L.J. Hainsworth, A.C. Mignerey, G.R. Helz, P. Sharma, P.W. Kubik, *Nucl. Instr. and Meth. B* 92 (1994) 345.
- [33] M.D. Keywood, L.K. Fifield, A.R. Chivas, R.W. Creswell, *J. Geophys. Res.* 103 (D7) (1997) 8281.
- [34] C. Lukaszcyk, Doctors Thesis, ETH – Zurich, Zurich, 1994.
- [35] R.C. Finkel, K. Nishiizumi, D. Elmore, R.D. Ferraro, H.E. Gove, *Geophys. Res. Lett.* 7 (1980) 983.