Gruppe	Fe/SiO_2[a]	Fe^0/Fe[b]	Fa[c]	SiO_2/MgO[a]
L	$0{,}55 \pm 0{,}05$	$0{,}33 \pm 0{,}07$	24 ± 2	$1{,}59 \pm 0{,}05$
LL	$0{,}49 \pm 0{,}03$	$0{,}08 \pm 0{,}07$	29 ± 2	$1{,}58 \pm 0{,}05$
St. Severin	0,508	0,096	29[13]	1,60

a Gewichtsverhältnisse b Fe^0/Fe = metallisches Fe/Gesamt-Fe c Mol-% Fayalit

Tab. 2. Chemische Merkmale des Meteoriten St. Severin und Vergleich mit der L- und LL-Gruppe der Chondrite Van Schmus und Wood[12]

Severin erhaltenen verglichen. Der Vergleich der chemischen Merkmale zeigt, daß der Meteorit St. Severin zur Gruppe der LL-Chondrite gehört. Die mineralogische Untersuchung (Orcel et al.[1]) ist mit diesem Ergebnis in Übereinstimmung.

[12] W. R. van Schmus u. J. A. Wood, Geochim. Cosmochim. Acta **31**, 747 [1967].
[13] B. Mason, Geochim. Cosmochim. Acta **31**, 1100 [1967].

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Tracer Studies of Atmospheric Exchange Based on Measurements of Cosmic Ray Produced Sodium-22 *

W. Roedel

II. Physikalisches Institut der Universität Heidelberg

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Die Ergebnisse der in Heidelberg während des Jahres 1966 durchgeführten Messungen des durch die kosmische Strahlung in der Atmosphäre erzeugten Na^{22} werden dargestellt. Anhand eines einfachen Modells wird versucht, von den ausschließlich am Boden gewonnenen Meßwerten auf den Austausch zwischen Stratosphäre und Troposphäre zu schließen. Die Modell-Rechnung liefert für das an Aerosole angelagerte Na^{22} eine mittlere Aufenthaltsdauer in der Stratosphäre von 12,3 Monaten; die durch die Tropopause ausgetauschten Luftmengen liegen nach dieser Rechnung zwischen Null (im Spätjahr) und 48 $g/cm^2 \cdot$ Monat (im Frühjahr).

Im Anhang wird die Abnahme des durch die Kernwaffentests von 1961 und 1962 künstlich produzierten Na^{22} diskutiert. Es wird gezeigt, daß die künstliche Aktivität in der Atmosphäre bis auf einen vernachlässigbaren Rest abgefallen ist.

Cosmic rays produce several nuclides by spallation of the constituents of the atmosphere[1]. Because it is possible to estimate the production rate of the spallation products fairly well, and because the production varies strongly with altitude and latitude, the radioactive ones among these nuclides are well suited for tracer studies of transport and exchange of aerosols and air in the atmosphere. Especially sodium-22, an argon spallation product with a half-life of 2.56 yrs, is useful for the study of exchange between stratosphere and troposphere[2, 3, 4].

The aim of the present paper is to discuss the possibility to estimate the exchange between stratosphere and troposphere by using only ground level measurements of radioactivity of medium- or long-lived cosmic ray produced nuclides.

The values of the Na^{22}-concentration in rain and ground-level air at Heidelberg during 1966 as well

* Erweiterte Fassung eines Vortrags, gehalten auf dem CACR-Symposium on Aerosols and Radioactive Tracers in the Atmosphere, Luzern (Schweiz), Oktober 1967.
[1] D. Lal and B. Peters, Cosmic Ray Produced Radioactivity on the Earth; Handbuch der Physik 46/2 [1967].
[2] N. Bhandari and Rama, J. Geophys. Res. **68**, 1959 [1963].
[3] W. Roedel, J. Geophys. Res. **70**, 4447 [1965].
[4] N. Bhandari, D. Lal, and Rama, Tellus **18**, 391 [1966].

as those of the deposition are given; the data are analysed by means of a simple box-model in terms of exchange between stratosphere and troposphere.

It has been shown earlier, however, that artificially produced Na^{22} has been injected into the upper atmosphere during the nuclear weapons tests of 1961 and 1962[3]. 1963 the level of artificial Na^{22} measured at Heidelberg was about one order of magnitude higher than the expected level of cosmic ray produced Na^{22}. The sodium-22 fallout has been measured continuously at Heidelberg ever since that time. The appendix of the present paper gives an analysis of the values of the sodium-22 deposition since 1963. For the mean stratospheric residence time of the artificial sodium-22 a value of 13 months has been found. Furthermore it is shown that the artificial component of sodium-22 in the atmosphere has decreased, till 1966, to a level, which can be neglected for the present discussion of cosmic ray produced radioactivity.

Measurements and Analysis of Cosmic Ray Produced Sodium-22

Fig. 1 shows the specific sodium-22 activity of rain and ground-level air at Heidelberg during 1966 as well as the sodium-22 deposition and the rainfall rate. The experimental procedure is described in[3].

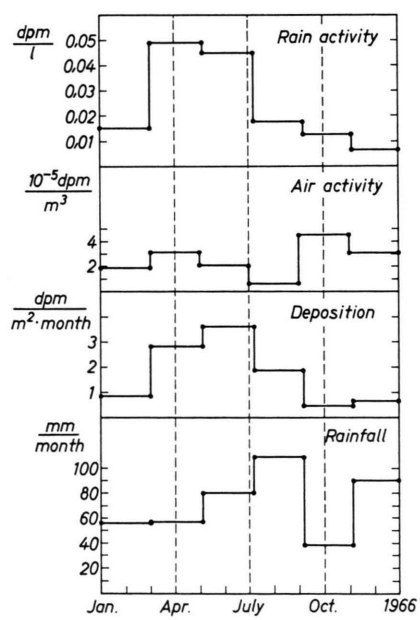

Fig. 1. From top to bottom: 1) Specific Na^{22}-activity in rain; 2) Specific Na^{22}-activity in ground-level air; 3) Total deposition of Na^{22}; 4) Rate of rainfall.

Deposition rate as well as specific rain activity show the expected seasonal variation due to the variation of exchange between stratosphere and troposphere; the specific activity of the air, however, shows a somewhat irreguliar behaviour. There seems to exist an anticorrelation between the specific activity of air and the rainfall rate, but this effect shall not be discussed here.

The measurements are now analysed by a simple model and interpreted in terms of exchange between stratosphere and troposphere.

The measurements to be analysed have been made at one point; the model, therefore, is one-dimensional, only depending on altitude. Because it is not possible, from the ground, to determine continuous transport or diffusion coefficients, a box model with only three distinct boxes is used.

Fig. 2 shows the boxes and the correlated quantities.

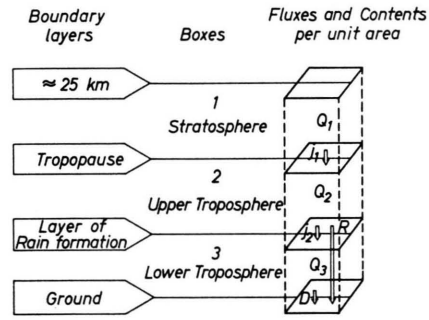

Fig. 2. Illustration of boxes; for explanation of quantities see the text.

Box 1 stands for the stratosphere from the tropopause to an altitude of about 25 km; this upper limit is a little arbitrary; above this altitude practically no production of nuclides by spallation processes occurs, and the exchange in this altitude is weak.

Box 2 represents the upper troposphere between the layer of rain cloud formation and the tropopause.

Box 3 stands for the lower troposphere between the ground and the rain cloud layer. The choice of the altitude of this latter boundary layer is a little arbitrary, too.

The following quantities are used:

$Q_{1,2,3}$ [atoms/cm²] — the Na^{22}-contents, per unit area, of box 1, 2, and 3, respectively,

$P_{1,2,3}$ [atoms/cm² · month] the rate of production by cosmic rays per unit area and unit time,

j_1 [atoms/cm² · month] the net flux of the nuclide from box 1 into box 2,

j_2 [atoms/cm² · month] the net flux from box 2 into box 3,

R [atoms/cm² · month] the rain-out going directly from box 2 to the ground,

D [atoms/cm² · month] the flux from box 3 to the ground (dry fallout),

λ [1/month] the constant of radioactive decay.

Then the following three differential equations hold:

$$\mathrm{d}Q_1/\mathrm{d}t = P_1 - \lambda Q_1 - j_1\,, \tag{1}$$

$$\mathrm{d}Q_2/\mathrm{d}t = P_2 - \lambda Q_2 - j_2 - R + j_1\,, \tag{2}$$

$$\mathrm{d}Q_3/\mathrm{d}t = P_3 - \lambda Q_3 - D + j_2\,. \tag{3}$$

The aim is to calculate j_1 (and furthermore Q_1) from the values of R, D and Q_3, which are measured on ground level.

To find a solution of these equations it is assumed that the total out-flux from a box is proportional to its contents, that means

$$j_1 = \lambda_1 Q_1;\quad j_2 + R = \lambda_2 Q_2;\quad D = \lambda_3 Q_3\,.$$

Furthermore λ_1 is taken as a function of the time (corresponding to the seasonal variation of exchange between stratosphere and troposphere), λ_2 and λ_3 shall be constants.

Then the following time-dependent solution of the differential equations (1), (2) and (3) is obtained:

$$j_1 = \lambda_1 Q_1 = \lambda_1 P_1 e^{-\lambda t} \exp\{-\int_0^t \lambda_1\,\mathrm{d}t'\} \int_0^t e^{\lambda t'} \exp\{\int_0^{t'} \lambda_1\,\mathrm{d}t''\}\,\mathrm{d}t' + \mathrm{const}_1\,\lambda_1 e^{-\lambda t} \exp\{-\int_0^t \lambda_1\,\mathrm{d}t'\}\,, \tag{1a}$$

$$j_2 + R = \lambda_2 Q_2 = \frac{\lambda_2 P_2}{\lambda + \lambda_2} + \lambda_2 \exp\{-(\lambda + \lambda_2)\,t\} \int_0^t j_1 \exp\{(\lambda + \lambda_2)\,t'\}\,\mathrm{d}t' + \mathrm{const}_2\,\lambda_2 \exp\{-(\lambda + \lambda_2)\,t\}, \tag{2a}$$

$$D = \lambda_3 Q_3 = \frac{\lambda_3 P_3}{\lambda + \lambda_3} + \lambda_3 \exp\{-(\lambda + \lambda_3)\,t\} \int_0^t j_2 \exp\{(\lambda + \lambda_3)\,t'\}\,\mathrm{d}t' + \mathrm{const}_3\,\lambda_3 \exp\{-(\lambda + \lambda_3)\,t\}. \tag{3a}$$

These solutions are exact, but they lead, for the purposes discussed here (λ_1 and j_1 are the unknown functions), to integral equations, which are too difficult to be evaluated. Therefore a time-independent steady state solution is used, which follows from the solution given above by setting constant λ_1 in (1a) and j_1 and j_2 in (2a) and (3a) respectively, and by allowing t to go to infinity.

Then

$$j_1 = \lambda_1 Q_1 = \frac{\lambda_1}{\lambda + \lambda_1} P_1\,, \tag{1b}$$

$$j_2 + R = \lambda_2 Q_2 = \frac{\lambda_2}{\lambda + \lambda_2} (P_2 + j_1)\,, \tag{2b}$$

$$D = \lambda_3 Q_3 = \frac{\lambda_3}{\lambda + \lambda_3} (P_3 + j_2) \tag{3b}$$

is obtained.

These solutions, of course, are approximations which are only valid, when averaged values are considered. This holds for (1b) for time intervals of the order of a year, for (2b) and (3b) for intervals of the order of one or two months.

In the steady case the parameters λ_1, λ_2, λ_3 have the meaning of the inverse of mean residence times in the boxes 1, 2, and 3 respectively, that is

$$1/\lambda_1 = \tau_1;\quad 1/\lambda_2 = \tau_2;\quad 1/\lambda_3 = \tau_3\,.$$

j_1 as a function of $T = R + D$ (T = total fallout) and Q_3 follow from (2b) and (3b) being

$$j_1 = (1 + \lambda\tau_2)\,(T + \lambda Q_3 - P_3) - P_2\,. \tag{4}$$

The value of τ_2 in this formula has to be taken from other measurements, for example from measurements of radon daughters of or medium-lived cosmic ray produced nuclides, as Be^7 or S^{35}. τ_2, however, is not a critical quantity, when long-lived nuclides are used; for Na^{22}, an error of 1 month for τ_2 causes for j_1 only an error of about 2.5%. When the calculations are applied, however, to short- or medium-lived nuclides, the value of τ_2 has to be taken carefully.

The next step is to estimate the net exchange of air masses between stratosphere and troposphere by use of the calculated flux of radioactive tracer atoms.

Let be

J_1 [g air/cm² · month] the net flux of air from box 1 into box 2,

$c_{1,3}$ [atoms/g air] the concentration of the considered nuclide in box 1, and box 3 respectively,

ϱ_1 [g air/cm²] the air mass per unit area in box 1.

Then holds

$$J_1 = j_1/c_1; \quad c_1 = Q_1/\varrho_1; \quad J_1 = j_1 \varrho_1/Q_1 .$$

c_1 and Q_1 can easily be calculated from the known flux and the known production rate by means of the solutions of equation (1). In the steady state model (for averaged values)

$$c_1 = (P_1 - j_1)/\lambda \varrho_1 \qquad (5)$$

is obtained.

Results

The model developed above is now applied, in its steady state form, to the measurements shown in Fig. 1

At Heidelberg, in 1966, a total sodium-22 fallout

$$T = 4.1 \cdot 10^3 \text{ atoms/cm}^2 \cdot \text{yr}$$

has been measured; the mean concentration in the ground level air has been found to be

$$c_3 = 6.3 \cdot 10^{-2} \text{ atoms/g air},$$

corresponding to

$$Q_3 = 26 \text{ atoms/cm}^2$$

(the level of the boundary layer between box 2 and box 3 being taken at 4000 m or 610 g/cm²).

The value for the production rates of Na^{22} are taken from [4], τ_2 has been taken to be 1 month. From these values, according to (4), the net sodium-22 flux from the stratosphere to the troposphere has been calculated to be

$$j_1 = 2.9 \cdot 10^3 \text{ atoms/cm}^2 \cdot \text{yr}.$$

The mean stratospheric concentration has been calculated, according to (5), to be

$$c_1 = 13 \text{ atoms/g air}$$

(for ϱ_1 a value of 220 g/cm² has been taken.) This, of course, is only a theoretical figure, but it lies well within the limits of the values given by BHANDARI and RAMA [2], who have found concentrations between 3 and 16 atoms/g air, analysing filters exposed in the stratosphere by planes.

The calculated mean residence time τ_1 in the lower stratosphere amounts to

$$\tau_1 = 12.3 \text{ months}.$$

The time-variation of exchange between stratosphere and troposhere, calculated according to (4) and (5), is shown in Fig. 3. The lower curve shows

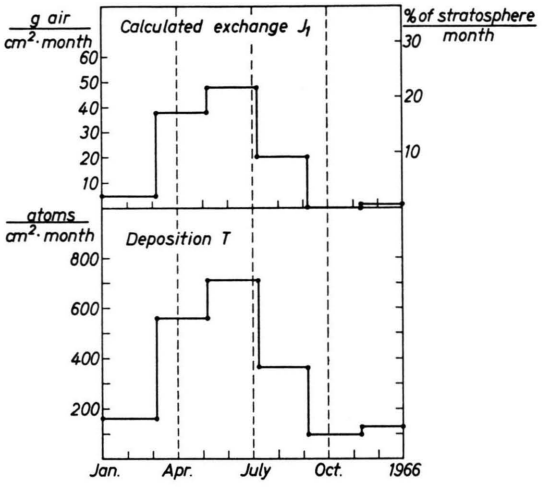

Fig. 3. Measured Na^{22}-fallout (lower curve) and calculated net exchange of air between stratosphere and troposphere (upper curve).

the fallout T, measured on ground, in bimonthly intervals. The upper curve shows the corresponding calculated values of the net air exchange rate J_1 between stratosphere and troposphere. The left scale gives the flux in g air/cm² · month, the right scale in percents/month of a standard stratosphere of 220 g/cm².

The abscissa corresponds to the time of collecting the samples on ground; the calculated exchange J_1 is, compared with the real time of exchange, shifted by about the tropospheric residence time. Time-independent calculations cannot take into account this delay. One has to imagine the upper curve shifted to the left by about one or two months.

Summary

The paper has discussed a method to estimate the exchange of air between the stratosphere and the troposphere only with help of ground-level measurements of cosmic ray produced nuclides.

Of course, the method presented here, a simple box-model calculation, is affected by some inaccuracies. They are caused firstly by the impossibility to treat the dependence on latitude, and by the simplification of the treatment of the dependence on altitude. Secondly they are caused by the difficulties of evaluating the exact time-dependence of the fundamental equations.

On the other hand, the measurements and the evaluation do not require great expenses and can be carried out quite easily. Therefore the method is well suited for a regular and continuous survey.

Appendix

The decay of bomb produced sodium-22.

Table I shows, in the first column, the annual values of the deposition of Na^{22} at Heidelberg from 1963 until 1966. In the last column the net flux from the stratosphere into the troposphere is given (calculated with help of the model developed above).

Year	Na^{22}-Deposition		Na^{22}-Flux from the stratosphere
	atoms/cm² · yr	dpm/cm² · yr	atoms/cm² · yr
1963	50.5×10^7	258	50.6×10^7
1964	26.8×10^7	146	26.8×10^7
1965	8.0×10^7	41	7.2×10^7
1966	4.1×10^7	21	2.9×10^7

Table 1.

The differential equation (1) with the quantities defined above

$$dQ_1/dt = -\lambda Q_1 - j_1 + P_1$$

is used for the analysis of the j_1-values.

With the assumption that the flux j_1 is proportional to the contents

$$j_1 = \lambda_1 Q_1 \quad (\lambda_1 \text{ assumed to be constant})$$

the solution of this equation is:

$$Q_1 = \frac{P_1}{\lambda + \lambda_1} + \text{const} \exp\{-(\lambda + \lambda_1) t\}.$$

Because j_1 is given (instead of Q_1) this solution is multiplied by λ_1 to obtain j_1

$$j_1 = \underbrace{\frac{\lambda_1 P_1}{\lambda + \lambda_1}}_{\text{natural component (stationary)}} + \underbrace{\text{const}\, \lambda_1 \exp\{-(\lambda + \lambda_1) t\}}_{\text{artificial component (decaying)}}.$$

From the latter term it is evident, that $\tau_1 = 1/\lambda_1$ has the meaning of the mean residence time in the stratosphere.

According to the method of least mean squares the parameter λ_1 and the constant of integration are fitted to the j_1-values of Table I.

The best fit is obtained by taking $\tau_1 = 13$ months for the mean stratospheric residence time. Together with the radioactive decay, the mean life-time for the bomb produced sodium-22 in the stratosphere is estimated to be 10 months. From this result and from the fact that the 1966 deposition agrees quite well with the natural production rate, one may infer that the artificial sodium-22-component in the atmosphere has decreased till 1966 to a level which can be neglected for practical purposes.

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