Tracer Studies of Atmospheric Exchange

Based on Measurements of Cosmic Ray Produced Sodium-22 *

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(Z. Naturforsch. 23 a, 51—55 [1968]; received 2 October 1967)

Die Ergebnisse der in Heidelberg während des Jahres 1966 durchgeführten Messungen des durch die kosmische Strahlung in der Atmosphäre erzeugten Na\textsubscript{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\textsubscript{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\textsuperscript{2} • Monat (im Frühjahr).

Im Anhang wird die Abnahme des durch die Kernwaffentests von 1961 und 1962 künstlich produzierten Na\textsubscript{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\textsuperscript{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\textsuperscript{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\textsubscript{22}-concentration in rain and ground-level air at Heidelberg during 1966 as well as

\begin{itemize}
  \item [3] W. ROEDEL, J. Geophys. Res. 70, 4447 [1965].
\end{itemize}

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\textsuperscript{1} D. LAL and B. PETERS, Cosmic Ray Produced Radioactivity on the Earth; Handbuch der Physik 46/2 [1967].

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\textsuperscript{2} N. BHANDARI and RAMA, J. Geophys. Res. 68, 1959 [1963].

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\textsuperscript{3} W. ROEDEL, J. Geophys. Res. 70, 4447 [1965].

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\textsuperscript{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 artifi-
cially produced Na$^{22}$ has been injected into the
upper atmosphere during the nuclear weapons tests
of 1961 and 1962\textsuperscript{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 deposi-
tion 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\textsuperscript{3}.

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, how-
ever, shows a somewhat irregular behaviour. There
seems to exist an anticorrelation between the spe-
cific 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-dimen-
sional, 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 quan-
tities.

![Diagram of boxes](image)

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

Box 1 stands for the stratosphere from the tropo-
pause to an altitude of about 25 km; this upper
limit is a little arbitrary; above this altitude prac-
tically no production of nuclides by spallation pro-
cesses 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:

\[ Q_{1,2,3} [\text{atoms/cm}^2] \]

the Na$^{22}$-contents, per
unit area, of box 1, 2, and 3, respectively.
The rate of production by cosmic rays per unit area and unit time,

\[ j_1 \text{ [atoms/cm}^2 \cdot \text{month]} \] the net flux of the nuclide from box 1 into box 2,

\[ j_2 \text{ [atoms/cm}^2 \cdot \text{month]} \] the net flux from box 2 into box 3,

\[ R \text{ [atoms/cm}^2 \cdot \text{month]} \] the rain-out directly from box 2 to the ground,

\[ D \text{ [atoms/cm}^2 \cdot \text{month]} \] the flux from box 3 to the ground (dry fallout),

\[ \lambda \text{ [1/month]} \] the constant of radioactive decay.

Then the following three differential equations hold:

\[ dQ_1/dt = P_1 - \lambda Q_1 - j_1, \quad (1) \]

\[ dQ_2/dt = P_2 - \lambda Q_2 - j_2 - R + j_1, \quad (2) \]

\[ dQ_3/dt = P_3 - \lambda Q_3 - D + j_2. \quad (3) \]

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 \( \lambda_1 \) is taken as a function of the time (corresponding to the seasonal variation of exchange between stratosphere and troposphere), \( \lambda_2 \) and \( \lambda_3 \) shall be constants.

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

\[ H = \lambda_1 Q_1 = \lambda_1 P_1 e^{-\lambda t} \exp \left\{ - \int_0^t \lambda_1 dt' \right\} \int_0^t e^{\lambda_1 t'} \exp \left\{ \int_0^{t'} \lambda_1 dt'' \right\} dt' + \text{const}_1 \lambda_1 e^{-\lambda t} \exp \left\{ - \int_0^t \lambda_1 dt' \right\}, \quad (1a) \]

\[ j_2 + R = \lambda_2 Q_2 = \frac{\lambda_2 P_2}{\lambda + \lambda_2} + \lambda_2 e^{-(\lambda + \lambda_2) t} \int_0^t \lambda_2 \exp \left\{ (\lambda + \lambda_2) t' \right\} dt' + \text{const}_2 \lambda_2 e^{-(\lambda + \lambda_2) t}. \quad (2a) \]

\[ D = \lambda_3 Q_3 = \frac{\lambda_3 P_3}{\lambda + \lambda_3} + \lambda_3 e^{-(\lambda + \lambda_3) t} \int_0^t \lambda_3 \exp \left\{ (\lambda + \lambda_3) t' \right\} dt' + \text{const}_3 \lambda_3 e^{-(\lambda + \lambda_3) t}. \quad (3a) \]

These solutions are exact, but they lead, for the purposes discussed here (\( \lambda_1 \) 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 \( \lambda_1 \) in (1a) and \( j_1 \) and \( j_2 \) in (2a) and (3a) respectively, and by allowing \( t \) to go to infinity.

Then

\[ j_1 = \lambda_1 Q_1 = \frac{\lambda_1}{\lambda + \lambda_1} P_1, \quad (1b) \]

\[ j_2 + R = \lambda_2 Q_2 = \frac{\lambda_2}{\lambda + \lambda_2} (P_2 + j_1), \quad (2b) \]

\[ D = \lambda_3 Q_3 = \frac{\lambda_3}{\lambda + \lambda_3} (P_3 + j_2) \quad (3b) \]

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.

For a steady case the parameters \( \lambda_1, \lambda_2, \lambda_3 \) have the meaning of the inverse of mean residence times in the boxes 1, 2, and 3 respectively, that is

\[ \frac{1}{\lambda_1} = \tau_1; \quad \frac{1}{\lambda_2} = \tau_2; \quad \frac{1}{\lambda_3} = \tau_3. \]

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

\[ j_1 = (1 + \lambda \tau_2)(T + \lambda Q_3 - P_3) - P_2. \quad (4) \]

The value of \( \tau_2 \) 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 Be⁷ or S³⁵. \( \tau_2 \), however, is not a critical quantity, when long-lived nuclides are used; for Na²², an error of 1 month for \( \tau_2 \) 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 \( \tau_2 \) 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
\[ J_1 \text{ [g air/cm}^2 \text{ • month]} \] the net flux of air from box 1 into box 2,
\[ c_{1,3} \text{ [atoms/g air]} \] the concentration of the considered nuclide in box 1, and
\[ q_1 \text{ [g air/cm}^2 \text{]} \] the air mass per unit area in box 1.

Then holds
\[ J_1 = j_1/c_1; \quad c_1 = Q_1/q_1; \quad J_1 = j_1 q_1/Q_1. \]

\(c_1\) 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)
\[ c_1 = (P_1 - j_1)/\lambda q_1 \quad (5) \]
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
\[ T = 4.1 \times 10^3 \text{ atoms/cm}^2 \cdot \text{yr} \]
has been measured; the mean concentration in the ground level air has been found to be
\[ c_3 = 6.3 \times 10^{-2} \text{ atoms/g air}, \]
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 \(^4\), \(\tau_2\) 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
\[ j_1 = 2.9 \times 10^3 \text{ atoms/cm}^2 \cdot \text{yr}. \]
The mean stratospheric concentration has been calculated, according to (5), to be
\[ c_1 = 13 \text{ atoms/g air} \]
(for \(q_1\) 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, who have found concentra-
tions between 3 and 16 atoms/g air, analysing filters exposed in the stratosphere by planes.

The calculated mean residence time \(\tau_1\) in the lower stratosphere amounts to
\[ \tau_1 = 12.3 \text{ months.} \]

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

<table>
<thead>
<tr>
<th>Year</th>
<th>Na$^{22}$-Deposition</th>
<th>Na$^{22}$-Flux from the stratosphere</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>atoms/cm$^2$ \cdot yr</td>
<td>dpm/cm$^2$ \cdot yr</td>
</tr>
<tr>
<td>1963</td>
<td>$5.0.5 \times 10^7$</td>
<td>258</td>
</tr>
<tr>
<td>1964</td>
<td>$2.6.8 \times 10^7$</td>
<td>146</td>
</tr>
<tr>
<td>1965</td>
<td>$8.0 \times 10^7$</td>
<td>41</td>
</tr>
<tr>
<td>1966</td>
<td>$4.1 \times 10^7$</td>
<td>21</td>
</tr>
</tbody>
</table>

Table I.

The differential equation (1) with the quantities defined above

$$\frac{dQ_1}{dt} = -\lambda Q_1 - j_1 + P_1$$

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 $\lambda_1$ to obtain $j_1$

$$j_1 = \frac{\lambda_1 P_1}{\lambda + \lambda_1} + \text{const} \lambda_1 \exp\{-(\lambda + \lambda_1)t\}.$$ 

natural component artificial component
(stationary) (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 $\lambda_1$ 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.

**Acknowledgement**

I am very indebted to the Bundesministerium für Wissenschaftliche Forschung for financial support.