coincidences observed [9] between the 381 and 620 keV γ rays. The log ft value of the β branch feeding the 1142 keV level is 7.5, which indicates that a spin assignment of $1/2-$ or $3/2-$ is likely. The K-conversion coefficient of the 1002 keV transition determined gives an M 2 character for that transition, while the 381 keV transition is found to have M 1 + E 2 character, see the table. According to these results, the 1142 keV level is assigned as $3/2-$. A level at 1170 keV is introduced, since the 989 keV γ ray was found to be in coincidence with the 181 keV transition. The existence of this level fits the replacement of the weak 409 keV transition which is coincident with the 620 keV transition, in the decay scheme. The log ft value of the β branch feeding the 1170 keV level suggesting a spin $1/2-$, $3/2-$ or $5/2-$. A spin of $1/2-$ can be excluded in view of the feeding to the $5/2+$, 181 keV level. Of the remaining spin values, $5/2-$ is somewhat less probable [9] since the log $1/24\,(W_0^2-1)\,ft$ value is rather low for a unique first-forbidden β transition. The M 1 character for the 409 keV transition may support the $3/2-$ assignment for the 1170 keV level. A new level at energy of 1198 keV proposed [9] due to the coincidence relation of the 1017 keV γ ray with the 181 keV transition. The appearance of the K-conversion line of the 1017 keV transition in the conversion spectrum confirms the existence of a level at 1198 keV. The study [9] of β transition feeding the 1198 keV level leads to a spin assignment of $5/2-$.

On the Behavior of Short-lived Cosmic Ray Produced Nuclides in the Lower Atmosphere

WALTER ROEDEL

II. Physikalisches Institut der Universität Heidelberg

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Some aspects of short-lived cosmic ray produced nuclides in the lower atmosphere, especially of Na^{24}, and their qualities as tracers for atmospheric motion are studied. The Na^{24} production rate has been estimated. The distribution of this nuclide as a function of altitude in the lower atmosphere has been described by a steady state eddy diffusion model. Measurements of Na^{24} in ground-level air have been carried out. The measured activities varied from $0.07 \cdot 10^{-3}$ to $0.28 \cdot 10^{-3}$ dpm/m³. These figures are in good accordance with the calculated values for reasonable diffusion parameters.

Calculations suggest that CRP-nuclides with life-times in the order of a day are useful as tracers for atmospheric motions in the range of eddy diffusion coefficients of about $5 \cdot 10^4$ to several 10^6 cm² sec^{-1}, and for altitudes below four of five kilometers.

Furthermore some relations between the specific radioactivity of rainwater and the specific activity of cloud-level air, with special respect to short-living CRP-nuclides are found. The evaluation of some measurements of Na^{24} activity in rain water shows good consistency between real atmospheric conditions and calculated values.

General Remarks

In the last ten or fifteen years many papers have been published on the behavior of long- and medium-lived cosmic ray produced (CRP-) nuclides in the atmosphere. A survey of the production mechanisms of CRP-nuclides as well as of the investigations about transport and distribution of medium- and long-lived isotopes in the atmosphere and the hydrosphere is given by LAL and PETERS [1].

On the other hand, only a few studies have been carried out on the behavior of short-lived CRP-nuclides in the atmosphere. The first detection of a short-living argon spallation product has been reported in 1956 by WINSBERG [2], who has measured the activity of Cl^{39} (55 min half-life) in rain water. Several years later, in 1963, Na^{24} with 15 hrs half-life has been detected in rain water by ROEDEL [3]. Hereafter, several other short-lived argon spallation products have been found: Mg^{28} (21 hrs), Si^{31} (2.6 hrs), S^{38} (2.9 hrs),

Reprint requests to Dr. W. ROEDEL, II. Physikalisches Institut der Universität Heidelberg, *D-6900 Heidelberg*, Philosophenweg 12.

[1] D. LAL and B. PETERS, Cosmic Ray Produced Radioactivity on the Earth, Handbuch der Physik 46/2 [1967].
[2] L. WINSBERG, Geochim. Cosmochim. Acta **9**, 183 [1956].
[3] W. ROEDEL, Nature **200**, 999 [1963].

Cl^{38} (37 min) and Cl^{34} (32 min) have been detected by BHANDARI et al.[4] (1966); independently PERKINS et al.[5] (1965) reported the detection of Cl^{38} and S^{38}, and HUSAIN and KURODA[6] (1966) the detection of Mg^{28}.

As far as known to the author, only BHANDARI et al.[4] and a group of the Battelle Memorial Institue, Pacific Northwest Lab. (C. W. THOMAS, private communication, 1967), have made use of short-lived CRP-nuclides as tracers for meteorological studies.

All the above mentioned isotopes were measured in rain water, no measurements of short-lived CRP-nuclides in air samples were reported till today.

Now it was possible to carry out the first measurements of Na^{24} in ground-level air; the results are reported in the present paper. Attempts have been made to calculate the production rate of Na^{24} as well as its distribution by turbulent mixing in the lower atmosphere. Calculated and measured concentrations are compared. Measurements of Na^{24} in rain water samples are also discussed.

Problems involved in the use of short-living nuclides as tracers for atmospheric motions are different from the problems connected with medium- and long-lived natural or artificial nuclides: The lifetime of radioactivity is shorter than the mean lifetime of aerosols in the atmosphere; thus deposition rates, mean residence times or similar quantities are no more significant. To draw conclusions, specific activities (or ratios of specific activities) have to be considered. Attempts have therefore been made in the last section of the present paper to estimate the ratio of the concentration in cloud-level air to concentration in rain water, with special respect to short-lived CRP-nuclides.

The Production of Na^{24} in the Troposphere

The missing of cross section data for the production of nuclides with atomic number $A<32$ by argon spallation makes it impossible to calculate the production rate of Na^{24} in the atmosphere directly. The only way out of this difficulty is to extrapolate from the well known source function of CRP-Na^{22}, calculated by BHANDARI et al.[7].

Laboratory investigations with radiochemical determination of yields of Na^{22} and Na^{24} have been carried out by BATZEL et al.[8] (copper target, 340 MeV protons), by RUDSTAM[9] (vanadium target, 187 MeV protons), and by FRIEDLANDER et al.[10] (aluminium target, protons from some hundreds to some thousands MeV). The results of these works show that the relative yields of Na^{22} and Na^{24} do almost not vary (from 1.3 to 2.3, energy dependence included) for the very different target materials, even when the absolute yields vary by several orders of magnitude from about 0.05 mbn (vanadium target) to about 15 mbn (aluminium target).

In the present paper the data of FRIEDLANDER et al.[10] are adopted for estimating the Na^{24} production by argon spallation: These data systematically cover a wide range of energy, and the atomic number of aluminium differs not too much from the atomic number of argon.

All measurements quoted above have been performed with protons; it was not possible to find data on yields from neutron induced spallation. According to Monte-Carlo-calculations on the processes during the intranuclear cascade (METROPOLIS et al.[11]), the production probability for nuclei with lower neutron numbers may be a little higher, when the target is bombarded with neutrons, in comparison with a bombardement by protons. This effect, however, is small; the second step of the spallation, the evaporation of nucleons from the excited nucleus and the following decay processes further equalize the yields (for the two stage theory of spallation see SERBER[12]). For the want of better knowledge, no difference between neutron and proton cross sections is made in the present paper.

The energy spectrum of the atmospheric nucleons has been taken from SCHOPPER et al.[13].

[4] N. BHANDARI, S. G. BHAT, D. P. KHARKAR, S. KRISHNA SWAMY, D. LAL, and A. S. TAMHANE, Tellus **18**, 505 [1966].
[5] R. W. PERKINS, C. W. THOMAS, M. W. HILL, and J. M. NIELSEN, Nature **205**, 790 [1965].
[6] L. HUSAIN and P. K. KURODA, Science **154**, 1180 [1966].
[7] N. BHANDARI, D. LAL, and RAMA, Tellus **18**, 391 [1966].
[8] E. R. BATZEL, D. R. MILLER, and G. T. SEABORG, Phys. Rev. **84**, 671 [1951].
[9] S. G. RUDSTAM, Phil. Mag. **44**, 1131 [1953].
[10] G. FRIEDLANDER, J. HUDIS, and R. L. WOLFGANG, Phys. Rev. **99**, 263 [1955].
[11] N. METROPOLIS, R. BIVINS, H. STORM, A. TURKEVICH, J. M. MILLER, and G. FRIEDLANDER, Phys. Rev. **110**, 185 [1958].
[12] R. SERBER, Phys. Rev. **72**, 1114 [1947].
[13] E. SCHOPPER, E. LOHRMANN, and G. MAUCH, Nukleonen in der Atmosphäre, Handbuch der Physik 46/2 [1967].

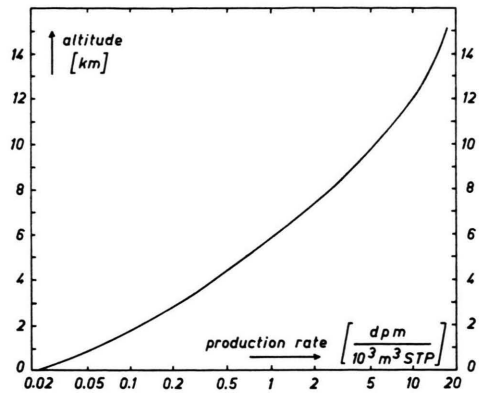

Fig. 1. Production rate of Na^{24} at 50° N.

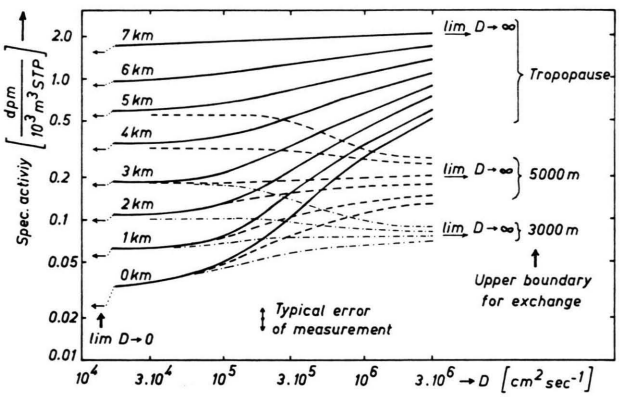

Fig. 2. Calculated specific Na^{24}-activity as a function of diffusion coefficient D at different altitudes, for three upper exchange boundaries. The arrows indicate at the left the specific activities for $D=0$ (equilibrium with production rate) and at the right the specific activities for $D \to \infty$ (perfect mixing), respectively.

The production rate of Na^{24} has been calculated for 50° geomagnetic latitude, and is shown in Fig. 1 as a function of altitude.

Distribution of Na^{24} in the Lower Troposphere

In this section the distribution of cosmic ray produced sodium-24 in an idealized atmosphere has been calculated.

It is assumed that the distribution of the nuclide in the atmosphere can be described by equilibrium between production, radioactive decay, and time-independent eddy diffusion in the direction of the z-(altitude-) axis.

To find an analytical solution of the diffusion equation, calculations have been carried out with diffusion coefficients uniform in the z-axis. The error due to the decrease of exchange length towards the ground (and therefore of the diffusion coefficient) becomes very small, when the condition that the diffusive flux is zero for $z=0$ is introduced afterwards.

The vertical diffusion coefficient D has been varied from $2 \cdot 10^4$ cm^2 sec^{-1} to $3 \cdot 10^6$ cm^2 sec^{-1}. D-values below or beyond these figures seem to be very improbable with respect to a stationary theory. Furthermore these figures cover the range, which JACOBI [14] has found to be suitable to explain the concentrations of Rn- and Tn-daughters in the troposphere.

Another parameter involved in the present calculation is the altitude of the upper boundary of exchange. A natural boundary, of course, is the tropopause, for which the calculation has been carried out at first. It might be possible, however, that inversion layers at lower altitudes are barriers for the exchange. This is suggested by observations of REITER [15] at the Zugspitz-Massiv (Bavaria), who reports strong and long-lasting boundaries of this kind for fission products (at an altitude of about 2.5 km). For two such barrier altitudes, 3000 m and 5000 m, the Na^{24} concentration has been calculated, too.

Figure 2 shows the calculated specific activity of Na^{24} as a function of the (uniform) diffusion coefficients for different altitudes. The full lines give the values for exchange up to the tropopause, the dotted lines the values for the boundaries at 3000 m and 5000 m respectively.

The plotted results show that the concentrations of Na^{24} in the lower troposphere are very sensitive to the variation of eddy diffusion coefficients in the range of $5 \cdot 10^4$ to about 2 or $3 \cdot 10^6$ cm^2 sec^{-1}. Below these values the specific activity is nearly in equilibrium with the production. For diffusion coefficients beyond several 10^6 cm^2 sec^{-1} the troposphere can be considered to be well mixed even with respect to a nuclide with 15 hrs half-life; a further increase of eddy diffusion strength does not change the concentrations very much. The calculations show also that beyond altitudes of four or five kilometers

[14] W. JACOBI, Forschungsbericht des Hahn-Meitner-Instituts, Berlin, HMI-B 21 [1962].

[15] R. REITER, Nukleonik 7, 79 [1965].

(exchange up to the tropopause assumed) the sensitivity of specific activity to eddy diffusion strength decreases.

At this point some remarks shall be made on the behavior of other cosmic ray produced nuclides: In the solution of the diffusion equation the diffusion coefficient D as well as the radioactive decay constant λ appear only in the term D/λ (or λ/D). As a consequence of this the calculations indicate that a corresponding behavior is to be expected for equal ratios of diffusion coefficient to decay constant, if nuclides with similar relative dependence of production rate on altitude are considered. This means that the concentration of Cl^{39} (55 min half-life), for instance, is almost in equilibrium with production for diffusion coefficients up to some times 10^6 cm^2 sec^{-1}. Be^7 can be considered to be fairly well mixed in the troposphere for diffusion coefficients down to $5 \cdot 10^4$ cm^2 sec^{-1}; the Be^7-activity will be in equilibrium with production for diffusion coefficients below about $5 \cdot 10^2$ cm^2 sec^{-1}. The relative Mg^{28}-distribution is very similar to that of Na^{24}.

Na^{24}-Measurements in Ground-Level Air and in Rain

Measurements of the specific Na^{24}-activity in ground-level air have been carried out for the first time. The results are shown in Table 1.

Each sample corresponds to about 8000 m^3 of air collected within about 30 hours (two half-times of Na^{24}). The chemical treatment is almost the same as the procedure described in Ref. [3]. The measurements of decay rates were performed by a very sensitive β-γ-coincidence spectrometer (ROEDEL [16]).

The comparison of the measurements (Table 1) with the calculated values (Fig. 2) shows that there is good agreement between measured and calculated activities, with reasonable turbulence strength, but only if exchange up to the tropopause is assumed. If at all, it might be possible to explain L 4 and L 5 also by the assumption of a low lying boundary. But this would be compatible only with very high exchange intensity below the assumed boundary ($D > 10^6$ cm^2 sec^{-1}). Such a strong turbulence, however, is very improbable for the given atmospheric conditions.

In the last column the calculated diffusion coefficients corresponding to the measured activities are given, exchange being assumed up to the tropopause.

The evaluation of measurements of short-living CRP-nuclides in rain water theoretically needs the knowledge of the ratio of concentration in rain water to the concentration in air to be scavenged by rain. The knowledge in this field, however, is still poor. To gain a working aid, it is tried, in the next section of this paper, to evaluate the available literature data and to estimate the rain-out efficiency and the ratio of specific activity of air and rain water.

Table 2 shows the results of Na^{24} measurements in rain water, carried out some years ago. The altitude of clouds is given by the Mannheim Station of the Deutscher Wetterdienst (some kilometers from Heidelberg). The concentration in cloud-level air is estimated by help of the considerations of the next section. The liquid water content of clouds is, for this estimate, taken to be $1-2$ g/m^3 STP for R 1

Sample	Collection date	Weather Conditions	Na^{24}-Activity	Calcul. diffusion coeff.
			dpm/10^3m^{3}STP	cm^{2}sec^{-1}
L 1	11. 7. 1968	Cyclone with center over Spain, high turbulence, passing of a warm front	0.28 ± 0.05	9.5×10^5
L 2	27. 7. 1968	Eastern border of an anticyclone with center over the British Islands, arctic air masses, very windy	0.18 ± 0.04	5.5×10^5
L 3	22. 8. 1968	Center of an anticyclone over Germany, fine weather, but somewhat windy	0.16 ± 0.05	4.9×10^5
L 4	6. 9. 1968	Weak anticyclone between two cyclones, warm air masses, stable air layers	0.07 ± 0.02	1.7×10^5
L 5	20. 12. 1968	Southern border of a cyclone with center over the British Islands, maritime air, stable layers	0.09 ± 0.01	2.5×10^5

Table 1.

[16] W. ROEDEL, Nucl. Instr. Methods **61**, 44 [1968].

Sample	Collection date	Intensity of rain mm	Weather Conditions	Altitude of clouds m	Na^{24}-activity in rain dpm/l	Estimated Na^{24}-activity in cloud-level air dpm/10^3 m^3 STP
R 1	7. 6. 1963	29	Weak cyclone, maritime air masses, weak turbulence	700—3000	0.18 ± 0.02	0.30—0.40
R 2	3. 7. 1963	24	Strong local thunderstorm, very warm matirime air masses, weak anticyclone	1200—3000*	0.35 ± 0.03	($\leqq$) 0.70—0.90
R 3	7. 10. 1963	38	Passing of a cold front, maritime air masses	100—4000	0.19 ± 0.03	0.30—0.40

* It may be possible that the altitude of the local thunderstorm cumuli at Heidelberg was higher than the cloud altitude given by the Mannheim station.

Table 2.

and R 3 (nimbostrati), and to be $2-3$ g/m^3 STP for R 2 (thunderstorm cumuli). R 1 and R 3 correspond to D-values of $2-4\cdot10^5$ cm^2 sec^{-1}, R 2 corresponds to $1-2\cdot10^6$ cm^2 sec^{-1} (mixing up to the tropopause is assumed). These results seem to be in a good agreement with the atmospheric conditions.

It should be pointed out that the estimate of the concentration ratio in rain and air (of the next section) does not take into consideration that an exchange can take place between air masses already scavenged and air masses still bearing radioactivity (at the border of a cloud, for instance, especially in cumuli). This effect increases, of course, the specific activity of air. Therefore the $\leqq$-symbol has been put before the R 2 air value.

A further remark has to be made at this point: Each steady state theory (as the eddy diffusion calculations presented above) averages implicitely over the life-time of a nuclide. On the other hand, it is always possible that single air motions cause a distribution which differs much from the steady state and cannot be described by a stationary calculation.

Activity Ratio of Rain Water and Cloud-level Air

Conveniently two stages are distinguished, when the activation of rain water by radioactivity is considered: The activation of droplets in the cloud, the "rain-out", and the attachment of radioactivity on the falling drops below the cloud, the "wash-out". Rain-out as the most important process shall be dealed with at first.

According to JUNGE [17] proportionality can be assumed between the concentration of substances in cloud water, C_{rain}, and the concentration in cloud level air, C_{air}; furthermore C_{rain} will be, in a good approximation, proportional to the inverse of the liquid water content per unit volume of cloud air, L:

$$C_{\text{rain}} = (\varepsilon/L)\, C_{\text{air}}\,.$$

The proportionality constant ε is often called "rain-out-efficiency".

L of a raining cloud is about 2 g/cm^3 STP within fairly narrow limits (MASON [18]). Nevertheless, L is always an uncertain quantity in the estimate of C_{air}, but it does not differ for different nuclides and can thus be eliminated, if only the ratio of two or more nuclides is of interest. The following considerations are primarily concerned with ε, which can differ for different nuclides.

It must be emphasized once again that this estimate cannot take account of the dynamical behavior of a cloud, e. g. condensation-evaporation cycles or the exchange between air already scavenged and air still bearing radioactivity.

Considering the activation of cloud water by CRP-nuclides one has to distinguish two mechanisms: The charging of water with radioactivity borne by aerosols which is described by the rain-out-efficiency for aerosols, $\varepsilon_{\text{aerosol}}$, and the direct attachment of radioactive atoms, produced in situ, on cloud droplets, described by the corresponding constant $\varepsilon_{\text{atom}}$. The latter process is important for short-lived CRP-nuclides.

A general remark has to be made at this point: The distribution of radioactivity borne by aerosols

[17] CHR. E. JUNGE, Air Chemistry and Radioactivity, Academic Press, New York and London 1963.

[18] B. J. MASON, The Physics of Clouds, Oxford University Press, London 1957.

depends on the history of the air and is the result of turbulent mixing, circulations and similar processes which mostly take place before cloud formation. The distribution of free radioactive atoms is only given by the production rate and does not depend on the history of the air.

Let τ be the mean life-time of a cloud, P the production rate, and λ the radioactive decay constant; then we have

$$C_{\text{rain}} = (1/L)\,[C_{\text{air}} \cdot \varepsilon_{\text{aerosol}} \cdot e^{-\lambda\tau} + (P/\lambda) \cdot \varepsilon_{\text{atom}} \cdot (1 - e^{-\lambda\tau})\,].$$

The probability for the attachment of atoms with atomic numbers of 20 to 40 is about $0.3 - 1.0\ \text{sec}^{-1}$ (JACOBI [14]); thus one can set $\varepsilon_{\text{atom}} = 1$.

For long-lived nuclides, $1/\lambda \gg \tau$,

$$C_{\text{rain}} = (\varepsilon_{\text{aerosol}}/L) \cdot C_{\text{air}}$$

is obtained. For short-living CRP-nuclides and fairly long-living clouds, $1/\lambda \ll \tau$, the relation is reduced to

$$C_{\text{rain}} = P/(\lambda \cdot L)$$

that means that the specific activity in rain and the production rate are in equilibrium.

Now the rain-out-efficiency for aerosols, $\varepsilon_{\text{aerosol}}$, has to be considered. As JUNGE [17] points out, $\varepsilon_{\text{aerosol}}$ can be split into three terms

$$\varepsilon_{\text{aerosol}} = \varepsilon_{\text{c}} + \varepsilon_{\text{d}} + \varepsilon_{\text{f}}\,.$$

ε_{c} is given by the "consumption" of aerosols as condensation nuclei, ε_{d} by the attachment of the remaining aerosols on cloud droplets by Brownian diffusion, and ε_{f} by the motion of aerosols towards the droplets, against the vapor pressure gradient.

The last effect has been detected by FACY [19]. Measurements and calculations by GOLDSMITH [20], however, indicate that ε_{d} is in the order of 10^{-3}. It can thus be neglected.

ε_{c} depends upon the particle size and the chemistry of the aerosol. Large and hygroscopic particles are preferred as condensation nuclei. For the attachment of mass on cloud water (that means mass of material/unit volume of water) JUNGE [17] estimates ε_{c} to be about 0.5 for air of continental origin and high aerosol concentration, about 0.8 for continental air and low aerosol concentration, and about 0.9 to 1.0 for maritime air.

ε_{c} for the consumption of radioactivity is lower than ε_{c} for the consumption of mass. This is due to the fact that the radioactive aerosol spectrum (radioactivity versus particle size) is, in comparison with the mass spectrum (mass versus particle size), shifted towards smaller particles, while the consumption probability increases for larger particles.

The mass of an aerosol particle is proportional to r^3, r being the particle radius. The combination of this factor r^3 with the natural size spectrum given by JUNGE [17] shows that particles with $r < 0.05\ \mu$ (or already $< 0.1\ \mu$) contribute almost nothing to the total mass. The coefficient of the attachment of radioactive atoms on aerosols, however, decreases more slowly than r^3 for decreasing r (BAUST [21]). Starting with Junge's aerosol spectrum Baust has calculated for the attachment of Th B (10.6 hrs half-life) that particles $r > 0.5\ \mu$ contribute almost nothing to the total radioactivity, whereas they contribute the greatest part to the total mass; according to Baust's calculations one third of the total radioactivity is attached on particles with $r < 0.1\ \mu$. ε_{c} becomes the smaller the shorter the half-life of the nuclide in question, because the size spectrum of aerosols shifts, by coagulation, towards larger particles for older aerosols.

For numerical values of ε_{c} only a rough and somewhat arbitrary estimate is possible. By help of the data given by JUNGE [17] and of the results of BAUST [21] ε_{c} may be estimated in the case of maritime air to be in the order of 0.7 for Cl^{39}, 0.8 for Na^{24} and 0.9 for long-lived nuclides, and in the case of continental air to be about $0.35 - 0.45$ for Cl^{39}, $0.5 - 0.6$ for Na^{24} and 0.7 for long-lived nuclides.

The efficiency of aerosol attachment on cloud droplets by Brownian diffusion, ε_{d}, is small compared with ε_{c}. Because of the higher mobility of smaller aerosol particles ε_{d} increases with decreasing particle size. ε_{d} shows therefore a behavior inverse to that of ε_{c}, and thus equalizes $\varepsilon_{\text{aerosol}}$ a little for different nuclides. According to measurements and considerations made by JACOBI [14], ε_{d} is for short-living nuclides (as Cl^{39}) in the order of 0.1 to 0.15, and decreases for nuclides with longer life-times. JUNGE [17] has estimated ε_{d} to be about 0.01 for the attachment of mass. This value can probably also

[19] L. FACY, Geofis. Pura Appl. **40**, 217 [1958].
[20] P. GOLDSMITH, H. J. DELAFIELD, and L. C. COX, Geofis. Pura Appl. **50**, 278 [1961].
[21] E. BAUST, Z. Phys. **199**, 187 [1968].

be adopted for the attachment of long-lived nuclides on cloud droplets.

The second mechanism for the activation of rain water, the attachment of radioactivity on falling rain drops below the cloud ("wash-out"), is a very small (and in most cases negligible) effect as far as CRP-nuclides are considered: According to calculations of LANGMUIR and of LANGMUIR and BLODGETT [22] the cross section of a falling rain drop for the attachment of aerosols is in a wide range of drop sizes given by the geometrical cross section multiplicated with a factor $\eta \leqq 1$, which depends only on the aerosol radius. η varies from $\eta = 1$ for $r \geqq 10\ \mu$ to about $\eta = 0.5$ for $r = 5\ \mu$, $\eta = 0.2$ for $r = 2\ \mu$ and finally to $\eta = 0$ for $r < 1\ \mu$. On the other hand, BAUST [21] has calculated that the contribution of particles with $r \geqq 0.8\ \mu$ to the total activity (Th B) is almost zero.

Langmuir's calculations are corroborated by observations made by REITER [23]: Reiter reports an increase of fission product activity in rain water below the cloud of 20% (maximum); simultaneously he finds a decrease of the concentration of aerosol particles with $r > 3\ \mu$, whereas he does not observe a decrease of concentration of smaller particles (Aerosol size distribution of fission products is shifted towards larger particles in comparison with the natural aerosol, see for instance SCHUMANN and BAUST [24]).

In any case an attachment of atoms and small ions on the falling rain drops by molecular diffusion could be possible; but experiments carried out by JACOBI [14] have shown that the increase of specific activity in rain below the cloud by molecular diffusion is below 5%, even for short-lived radon daughters, and can thus be neglected in the frame of the estimate presented here.

The very complex problem of increase of the specific activity by partial evaporation of rain drops below the cloud base shall be left out here. The evaporation effect is equal for all nuclides and can formally be allowed for by a correction of the quantity L. Furthermore, investigations of KEGELMANN [25] have shown that evaporation is no more important for drop sizes usually ocurring in single rains with intensities beyond three to four mm. Useful rains for tracer studies by CRP-nuclides are mostly beyond this limit.

It should be emphasized that the considerations presented here shall only be a preliminary aid for the evaluation of measurements of short-living CRP-nuclides in rain water.

Notes added in proof: Some parts of this paper have been presented at the CACR-Symposium, Heidelberg, September 8—13, 1969. — During this conference J. A. YOUNG, C. W. THOMAS, N. A. WOGMAN, and R. W. PERKINS (Battelle Memorial Inst., Richland, Wash.) have also reported measurements of the Na^{24}-activity in the air (to be published in J. Geophys. Res., in 1970).

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[22] J. LANGMUIR, General Electric Res. Lab. Rep. RL-224 [1945]. — J. LANGMUIR and K. B. BLODGETT, General Electric Res. Lab. Rep. RL-225 [1945].

[23] R. REITER, Atomkernenergie **5**, 68 [1961].

[24] G. SCHUMANN and E. BAUST, Proc. Coll. Intern. Pollution Radioactive des Milieux Gazeux (Saclay 1963), Vol. I. 11. Presse Univ. de France, Paris 1965.

[25] G. KEGELMANN, Forschungsbericht des Bundesministeriums für Wissenschaftliche Forschung K 65-05 [1965].