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Solubility and Distribution of Radon in Organic Solvents

S. D. Kal'muk (Bryk), R. G. Makitra, and E. Ya. Pal'chikova

Institute of Geology and Geochemistry of Fossil Fuels,
National Academy of Sciences of Ukraine, ul. Nauchnaya 3a, Lviv, 79053 Ukraine

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Abstract—The influence of physicochemical properties of solvents on the solubility of radon in them can be described by multiparameter linear equations taking into account various solvent characteristics; the decisive (and negative) effect on the radon solubility is exerted by the cohesion energy density and polarity of the solvent. This approach is efficient for summarizing data on the distribution ratios of radon between organic and aqueous phases.

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The solvent effect on the solubility of gases is a problem of not only theoretical but also applied importance. However, to avoid specific solvation effects, the general relationships are usually examined using as examples gases incapable of specific solvation interactions, such as nitrogen, methane, and especially inert gases. Otherwise the specific solvation effects can be very large: for example, the solubility of CO_2 increases by a factor of 10^3 in going from toluene to methanol [1]. However, it should be taken into account that the solubility of gases is influenced not only by physicochemical characteristics of a solvent and a gas, but also by the size of gas molecules.

Extensive data are available on the solubility of inert gases in various liquids. An exception is radon, which is quite natural, taking into account its difficult availability and radioactivity. At the same time, data for radon are of doubtless interest. On the one hand, radon is an inert gas with the highest atomic mass and hence the largest atomic size, and it is known that the solubility of inert gases grows with an increase in their radius. For example, the Henry constant of helium at 273 K in water is 13.1×10^4 atm, and that of xenon, as low as 0.57×10^4 atm [2]; virtually no such data are available for radon. On the other hand, the problem of radon solubility and possible concentration is of practical (in particular, of environmental) importance. Radon is one of major sources of radioactive pollution of the environment, which is due both to its gaseous state and to its continuous generation by the decay of other radioactive elements present in natural rocks and building materials. The problem of radon absorption is also important from the viewpoint of geochemical research, including explorations to reveal deposits of radioactive minerals.

In contrast to inert gases of lower atomic masses, data on the solubility of radon are very limited. The most representative set of data was published as early as 1920 [3] by Schulze, who examined the solubility of radon in water and 13 organic liquids at different temperatures with the aim to check whether the radon solubility obeys the Dolezalek equation. It was found that the radon solubility noticeably varies with temperature and that its experimental values for the majority of solvents differ from the calculated values, especially at low temperatures. Schulze attributed this fact to an increase in the association of liquids and hence to changes in their actual molecular masses (this quantity was used in the calculations). However, this explanation is not convincing, because not only water, aniline, and alcohol, but also acetone and chloroform behave in this case as associated liquids. Data from [3] are cited as the main source in the well-known Kogan's handbook [4]. In later studies, the radon solubility was examined only in one or two solvents and under different conditions, which complicates comparison of the data. The data on the radon solubility (ml ml^{-1} and mole fraction scales), derived from the published papers, are given in Table 1.

If there were no solvation interactions between the solvent and the inserted radon atom, the energy consumption for the radon dissolution ΔG_{sol} would correspond to the energy consumption for the cavity formation in the liquid structure, which is proportoinal to the Hildebrand solubility parameter squared, δ^2. Since $\Delta G_{sol} = -RT \log N$, the proportionality would also be observed between the amount of the dissolved gas ($\log N$) and δ^2 of solvents. However, as far as a decrease in the radon solubility in associated liquids (aniline, alcohol, and especially water) is concerned,

Table 1. Solubility of radon at 18°C[a] in organic solvents (α, ml ml^{-1}, from [3]; scale of mole fractions N) and values of $\log N$ calculated by Eq. (3)

Solvent no.	Solvent	α	$-\log N_{\text{exp}}$	$-\log N_{\text{calc}}$	$\Delta\log N$
1	Carbon disulfide	23.14	1.2588	1.5067	–0.2479
2	Chloroform	15.08	1.3143	1.4487	–0.1344
3	Ethanol	6.17	1.8268	1.9502	–0.1234
4	Ethyl acetate	7.34	1.5346	1.4188	0.1158
5	Benzene	12.82	1.3391	1.2592	0.0799
6	Aniline	3.80	1.8447	1.7641	0.0806
7	Acetone	6.30	1.7167	1.6162	0.1005
8	Diethyl ether	15.08	1.2076	1.1959	0.0117
9	Hexane	16.56	1.0794	0.9162	0.1632
10	Toluene	13.24	1.2534	1.2541	–0.0008
11	Water	0.285	3.6676	3.8232	–0.1556
12	*n*-Pentanol	9.31	1.3925	1.7742	–0.3816
13	Cyclohexane	18.04	1.0462	1.0641	–0.0179
14	Glycerol	0.21	3.1925	2.6825	0.5099
15	*n*-Butanol		1.4559		
16	Isobutanol		1.5003		
17	*n*-Propanol		1.5638		
18	Isopropanol		1.6576		
19	Methanol		2.0269		

[a] The solubility for solvent nos. **15–19** is given for 25°C [5].

we can speak of a qualitative trend rather than a linear correlation: The correlation coefficient r is as low as 0.93 (Fig. 1). Obviously, still worse correlation with δ^2 is observed when the solubility is presented in the nonlogarithmic N units or as Bunsen solubility coefficients α.

Here it is appropriate to mention one more possible effect. The majority of inert gases form hexahydrates in water at low temperature [6]. In particular, the hexahydrate was also identified for radon [7]. It is believed that such compounds are formed owing to van der Waals forces and have a clathrate structure; their stability increases with the atomic mass (and hence atomic size) of the inert gas. Similar clathrates are formed with phenol. The observed noticeable deviation of the point for water from the linear correlation between $\log N$ and δ^2 may be indirectly associated with this phenomenon (Fig. 1).

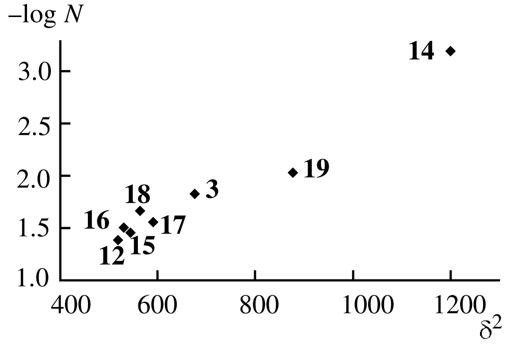

Fig. 1. Correlation between the radon solubility in alcohols (scale of mole fractions N) and their Hildebrand solubility parameter squared, δ^2. Point nos. are solvent nos. in Table 1. Data for solvent nos. **3, 12**, and **14** are given for 18°C, and those for solvent nos. **15–19**, for 25°C.

The lack of a good linear correlation suggests that the solubility of radon in liquids is significantly affected, along with the cohesion energy density of the solvents, also by other factors reflecting physicochemical features of solvents; in particular, nonspecific interactions between the solute and solvent cannot be ruled out. The general consideration of these factors is possible only on the basis of the linear free energy relationship, using linear multicomponent equations. We showed previously that the five-parameter equation reflecting the capability of solvents for specific and nonspecific solvation and their cohesion energy density is suitable for quantitative correlation of data on the solubility of nitrogen [8], oxygen [8], CO_2 [10], and some other gases. In particular, we showed in [11] that data on the solubility of xenon in 31 solvents can be generalized using such an equation with acceptable accuracy (multiple correlation coefficient R 0.952); only two factors, cohesion energy density and polarity of solvents, appear to be significant: As they increase, the gas solubility decreases.

It seemed appropriate to apply this approach to data on the radon solubility (Table 1). It is seen that the radon solubility is the highest in low-polarity solvents containing no hydroxy groups (hydrocarbons, CS_2, and also chloroform) and the lowest in associated hydroxyl-containing solvents (alcohols, water). The pattern becomes particularly clear when the solubility data are presented not on the volume scale, but also on the scale of mole fractions of the absorbed gas. We found in [12] that, to correlate data on the solubility of solids in organic solvents, it is appropriate to consider additionally the factor of molar volume V_M reflecting the structural features of the liquid phase. Therefore, to correlate data from Table 1, we used general six-parameter equation (1):

$$\log N = a_0 + a_1(n^2 - 1)/(n^2 + 2) + a_2(\varepsilon - 1)/(2\varepsilon + 1) + a_3B + a_4E_T + a_5\delta^2 + a_6V_M, \quad (1)$$

where n is the refractive index and ε, dielectric permittivity (parameters determining the polarizability and polarity of solvents), i.e., factors responsible for the nonspecific solvation; B (Palm basicity) and E_T (Reichardt electrophilicity) determine the capability of solvents for specific interactions (with inert radon, however, such interactions are hardly probable); δ (solubility parameter) and V_M (solvent molar volume) characterize the structure of the liquid medium.

The calculation was performed in accordance with the recommendations of the IUPAC Group for Correlation Analysis in Chemistry (CAC IUPAC) [13]. By so doing, we obtained Eq. (2) with a fairly high multiple correlation coefficient R:

$$\log N = 0.630 - (2.38 \pm 1.58)f(n^2) - (2.28 \pm 2.11)f(\varepsilon) - (0.28 \pm 0.88) \times 10^{-3}B + (0.79 \pm 2.70) \times 10^{-2}E_T - (1.40 \pm 0.37) \times 10^{-3}\delta^2 - (4.35 \pm 4.59) \times 10^{-3}V_M. \quad (2)$$

Number of points 14, R 0.967; rms error S ±0.200. Here $f(n^2) = (n^2 - 1)/(n^2 + 2)$; $f(\varepsilon) = (\varepsilon - 1)/(2\varepsilon + 1)$.

As in all the previous calculations, the reliability of the equation and correlation coefficient is confirmed by comparison of the calculated values of their Fisher test with the tabulated values for the corresponding number of points at the confidence level $\alpha = 0.95$. However, large standard deviations of the coefficients at the majority of the terms of the regressions, which are close to, or even exceed the absolute values of these coefficients, suggest that these terms are probably insignificant. Low values of pair correlation coefficients of $\log N$ with separate parameters, equal to 0.189, 0.698, 0.392, 0.896, 0.939, and 0.741, respectively, do not allow unambiguous conclusions about the significance of their contribution to $\log N$. The only exception is the correlation between $\log N$ and the Hildebrand solubility parameter squared, δ^2 (r 0.939), suggesting that the cohesion energy density of the medium exerts a decisive (and negative) effect on the radon solubility. However, the effect of other characteristics of the medium is also noticeable: taking them into account improves the correlation. Therefore, to evaluate the significance of separate terms of Eq. (2), in accordance with the recommendations of [13], we used the leave-one-out procedure. If exclusion of a given term did not lead to a significant decrease in R, this term was considered to be insignificant. By so doing, we proved the insignificance of the factors of specific solvation (without E_T, R 0.967; without E_T and B, R 0.966) and molar volume of the medium and obtained three-parameter equation (3) describing fairly reliably the correlation of the radon solubility with the solvent parameters:

$$\log N = 0.089 - (1.77 \pm 1.38)f(n^2) - (1.84 \pm 0.67)f(\varepsilon) - (1.15 \pm 0.13) \times 10^{-3}\delta^2, \quad (3)$$
$$R\ 0.962,\ S\ \pm 0.213.$$

As can be seen, the capability of solvents for nonspecific solvation also negatively affects the radon solubility, apparently because of the solute effect on the structure-forming ability of the liquid phase. Here it is appropriate to mention a relatively recent study [14] in which it was found that the solubility of nonpolar gases, including Ar, H_2, and Xe, in polar organic solvents can be characterized using the so-called "corrected" solubility parameter δ^x taking into account the polarity and polarizability of liquids. Exclusion of further terms from Eq. (3) impairs the correlation more noticeably; nevertheless, two-parameter equation (4) including only δ^2 and solvent polarity $f(\varepsilon)$ is also adequate:

$$\log N = -0.458 - (1.62 \pm 0.69)f(\varepsilon) - (1.14 \pm 0.14) \times 10^{-3}\delta^2, \quad (4)$$
$$R\ 0.958,\ S\ \pm 0.226.$$

The values of $\log N$ calculated by Eq. (3) and their deviations from the experimental data, $\Delta\log N = \log N_{exp} - \log N_{calc}$, are listed in Table 1, and the correlation between $\log N_{exp}$ and $\log N_{calc}$ is plotted in Fig. 2. As can be seen, the deviations are either smaller than the rms error S ±0.213 or only slightly exceed this value. The only exception is the strongly structured glycerol; however, in this case there also may be an experimental error caused by high viscosity of the medium.

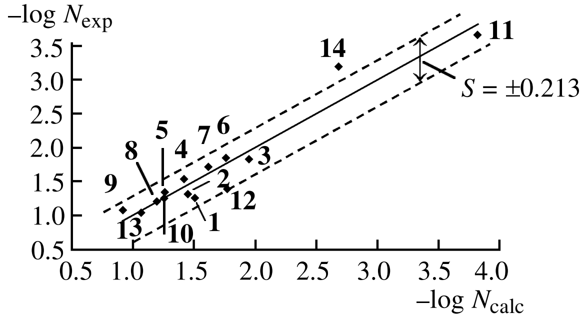

Fig. 2. Correlation between the experimental and calculated [by Eq. (3)] solubilities of radon in solvents. Point nos. are solvent nos. in Table 1.

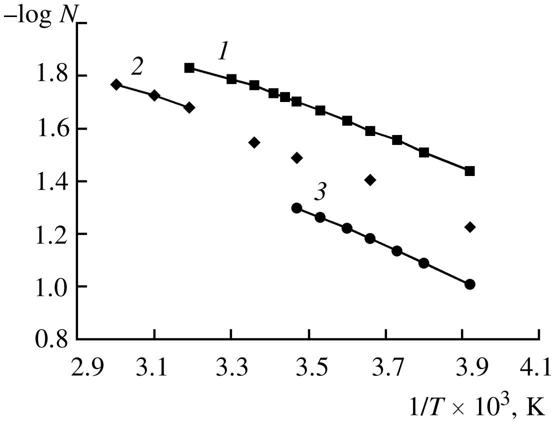

Fig. 3. Temperature dependence of the radon solubility (in mole fractions *N*) in (*1*) acetone, (*2*) chloroform, and (*3*) ethyl acetate.

At the same time, other approaches to estimating the solubility of inert gases in liquids, especially polar, give appreciably less satisfactory results. Here we can only mention an early paper [15] concerning a probable correlation between the solubility of gases and surface tension of solvents. In Linford and Thornhill's study [16], the improved Hildebrand–Lemure theory leads to 50–100% discrepancies between the calculated and experimental solubilities of inert gases, even in *n*-alkanes. Calculations based on data on the liquid–vapor equilibrium for solvents give acceptable accuracy only for nonpolar solvents [17, 18].

It is interesting that, with xenon [11], the same two parameters are significant (and negatively affecting the solubility) as in Eq. (4), and the pair correlation coefficient of $\log N$ with δ^2 for 31 solvents is close (r 0.923).

It should be noted that the coefficient a_0 of Eqs. (2) and (3) should correspond to the ideal solubility of gases without any solvent effects, calculated from the saturated gas pressure at this temperature, $N = 1/P$. However, according to [18], at 25°C this quantity is equal to 0.0752, which is appreciably lower than the values corresponding to a_0 for radon. This inconsistency can be accounted for on the basis of Gerrard's views [5] that the solubility of gases is influenced not only by the energy consumption for the cavity formation in the liquid phase, but also by the enthalpy of transformation of a gas into a hypothetical liquid upon dissolution.

It should also be noted that Gerrard [5] reported data on the radon solubility in 25 solvents but at different temperatures; these data, as a rule, differed from those considered here and obtained at 18°C. Nevertheless, we included in Table 1 data from [5] on the solubility of radon in five alcohols (nos. **14**–**18**). It is noteworthy that the solubility of radon (in mole fractions) regularly increases with an increase in the alkyl chain length in the alcohol, i.e., with an increase in the molecular mass and a decrease in the polarity. However, cyclohexanol deviates from this trend. Similar dependence was also found for argon [19] and other inert gases [20]. The negative effect of the structure-forming ability of a liquid on the solubility of inert gases and an increase in the solubility with an increase in the atomic radius of inert gases were also noted in [21]. We did not use these data (for 25°C) in the main calculation; however, despite different experimental temperature, these data can also be satisfactorily fitted by Eq. (1). In so doing, the correlation is impaired only slightly: For the six-parameter equation, *R* is 0.959, and the simplified three-parameter equation $\log N = f(n^2, \varepsilon, \delta^2)$ does not differ appreciably from the corresponding Eq. (3):

$$\log N = 0.223 - (2.41 \pm 1.44) f(n^2) - (1.42 \pm 0.70) f(\varepsilon) - (1.26 \pm 0.12) \times 10^{-3} \delta^2, \quad (5)$$
$$R\ 0.953,\ S\ \pm 0.209.$$

However, the correlation coefficient of the two-parameter equation $\log N = f(\varepsilon, \delta^2)$ becomes undesirably low: *R* 0.945.

Taking into account successful correlation of data on the radon solubility with multiparameter equation (1) and the fact that similar correlation of data on the enthalpies of solution of xenon was done in [11], it was appropriate to obtain a correlation for the energy parameters of solution of radon. For the majority of solvents studied in [3], the solubility of radon was determined in a relatively wide temperature range, which allows calculation of the enthalpies of solution ΔH_{sol}. Since the relationships $\log N$–$1/T$, as a rule, are satisfactorily linear (Fig. 3) (deviations are observed only for ethyl acetate and diethyl ether, and only at elevated temperatures), determination of ΔH_{sol} (Table 2) is fairly reliable. These data for ten solvents can be correlated by an equation of type (1):

$$\Delta H_{sol} = 7.28 - (7.32\pm0.87)f(n^2) - (2.07\pm1.13)f(\varepsilon) - (0.94\pm0.45)\times10^{-3}B - (36.39\pm12.14)\times10^{-3}E_T + (1.38\pm0.17)\times10^{-3}\delta^2 - (8.97\pm1.06)\times10^{-3}V_M, \quad (6)$$
$$R\ 0.995,\ S\ \pm0.083.$$

As in the case of $\log N$, the pair correlation coefficients are low, except that with δ^2: r 0.868. Since the dissolution of radon in liquids is endothermic, the coefficient at the decisive term of the equation, δ^2, is also positive. The equation can be simplified to a three-paameter equation, but here ΔH_{sol} is significantly affected by the polarizability and basicity of the solvents; these parameters appear in the equation with negative coefficients:

$$\Delta H_{sol} = 4.11 - (5.04\pm1.17)f(n^2) - (2.78\pm0.49)\times10^{-3}B + (1.14\pm0.10)\times10^{-3}\delta^2, \quad (7)$$
$$R\ 0.979,\ S\ \pm0.176.$$

Krestov et al. [22] reported data on ΔG_{sol} and $T\Delta S_{sol}$ of solution of radon in 49 solvents. However, both the ΔH_{sol} values calculated from these quantities and the ΔG_{sol} values themselves are correlated with Eq. (1) unsatisfactorily. To obtain an acceptable value of R 0.956, it is necessary to exclude data for 10 solvents (about 25%) out of 39 ΔH_{sol} values, and none of the terms can be excluded from the correlation without its drastic deterioration. The similar pattern is observed with ΔG_{sol}. This may be due to the fact that the initial values of the Ostwald coefficients were determined in [22] according to [23], by comparative calculation from the solubility of neon in the corresponding solvents. The accuracy of this procedure is moderate. At the same time, it can be mentioned that Abraham [24] reported the values of ΔG_{sol}, based on data from [25], for the radon solution in six solvents (kcal mol^{-1}): hexane 1.55, decane 1.49, toluene 1.81, benzene 1.92, ethanol 2.56, and water 5.15. These values noticeably differ from those given in [22]. The number of solvents examined in [24] is too small to obtain statistically reliable conclusions, but the general trend toward an increase in ΔG_{sol} (i.e., a decrease in the solubility) with an increase in the cohesion energy density and polarity of solvents is clearly seen; moreover, the pair correlation coefficient between ΔG_{sol} and δ^2 is almost equal to unity (!).

Since data on the solubility of radon, i.e., on its distribution between the gas and liquid phases, are satisfactorily correlated with six-parameter equation (1), it was appropriate to check whether this equation is applicable to the distribution of radon between two immiscible phases: aqueous and organic. This correlation would be not only interesting theoretically, but also important for geochemical research. Sekine and Yamasaki [26] reported data on the distribution ratios of radon K_d at 25°C between ten organic solvents and water (Table 3). Correlation of these data with Eq. (1) is characterized by unsatisfactorily low R 0.917. However, exclusion of the most deviating data for bromobenzene gives Eq. (8) with a satisfactory value of R:

Table 2. Experimental and calculated enthalpies of solution of radon in organic solvents ΔH, kcal mol^{-1}

Solvent	ΔH_{exp}	ΔH_{calc}	$\Delta(\Delta H)$
Carbon disulfide	0.3018	0.2986	–0.0032
Chloroform	0.2950	0.2923	–0.0027
Ethanol	0.3014	0.2979	–0.0035
Ethyl acetate	0.3030	0.2868	–0.0162
Aniline	0.1866	0.1870	0.0004
Acetone	0.2717	0.2874	0.0157
Diethyl ether	0.2848	0.2840	–0.0008
Hexane	0.3214	0.3226	0.0013
Toluene	0.2769	0.2856	0.0087
Water	0.5298	0.5302	0.0003

$$\log K_d = -2.75 + (12.01\pm2.20)f(n^2) - (2.11\pm0.34)f(\varepsilon) - (1.42\pm0.39)\times10^{-3}B + (0.12\pm0.02)E_T - (7.39\pm1.24)\times10^{-3}\delta^2 + (3.12\pm1.11)\times10^{-3}V_M, \quad (8)$$
$$R\ 0.976,\ S\ \pm0.04.$$

Simplified equation (9) including only four parameters is also adequate:

$$\log K_d = -1.51 + (9.77\pm2.88)f(n^2) - (0.90\pm0.47)\times10^{-3}B + (0.10\pm0.03)E_T - (6.78\pm1.65)\times10^{-3}\delta^2, \quad (9)$$
$$R\ 0.952,\ S\ \pm0.05.$$

Here the effect of basicity B is relatively insignificant; its exclusion decreases R to 0.930, which is consistent with the incapability of inert radon to participate in donor–acceptor interactions. Thus, the distribution of radon between the aqueous and organic phases, like its solubility, depends on the cohesion energy density of the solvents and on their capability for nonspecific solvation; however, the effect of polarizability in these two cases is opposite.

It should be emphasized that the coefficient at the parameter δ^2 characterizing the self-association of the solvents is negative. Naturally, penetration of a foreign molecule into the structure of a liquid involves the energy consumption for the cavity formation to accommodate the gas molecule. As the tendency of

Table 3. Correlation between the experimental and calculated [Eq. (9)] solubilities of radon

Solvent	$\log K_{\text{exp}}$	$\log K_{\text{calc}}$	$\Delta\log K_{\text{d}}$
Tetrachloromethane	1.7709	1.7967	0.0259
Chloroform	1.7482	1.7810	0.0328
Bromoform	1.5911	1.6061	0.0151
Hexane	1.9031	1.8731	–0.0300
2-Hexanone	1.5682	1.5820	0.0138
Chlorobenzene	1.7482	1.7765	0.0283
Isohexanol	1.5185	1.4946	–0.0239
Nitrobenzene	1.3222	1.3793	0.0571
o-Dichlorobenzene	1.6990	1.5799	–0.1191
Bromobenzene[a]	1.4914	1.6722	0.1808

[a] Data excluded from the calculation.

liquids to self-association increases if their molecules contain groups capable of hydrogen bonding, the minus sign at the polarity parameter is quite understandable, because such substances are usually highly polar. At the same time, increased polarizability of solvents favors the transfer of radon into the organic phase, as indicated by the plus sign at the term with $f(n^2)$, which suggests the possibility of nonspecific solvation of the radon molecule with solvents.

The $\log K_\text{d}$ values calculated by Eq. (9) and their deviations from the experimental values, $\Delta\log K_\text{d} = \log K_{\text{exp}} - \log K_{\text{calc}}$, are given in Table 3. For all the solvents, they are within the error corridor, $S \pm 0.05$, except *o*-dichlorobenzene for which $\Delta\log K_\text{d} = -0.119$ (i.e., 8% of K_d) and, naturally, bromobenzene excluded from the calculations.

Application of Eq. (9) allows not only the choice, based on the known physicochemical characteristics of extractants, of the most reliable K_d values from discrepant data of different authors, but also tentative estimation of K_d for liquids for which experimental data are lacking.

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