

Monthly Deposition of Cadmium in Rural and Industrial Areas of Germany (Bayern, Pfalz, Ruhr District) and Its Influences upon an Agricultural Model System¹

K. H. RUNKEL AND H. D. PAYER

Gesellschaft für Strahlen- und Umweltforschung, mbH, Institut für Toxikologie und Biochemie, Abt. Toxikologie, Ingolstädter Landstrasse 1, D-8042 Neuherberg, Federal Republic of Germany

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Monthly depositions of cadmium were collected by a modified Bergerhoff method and measured by AAS during a 3-year period in rural areas of the Pfalz and in an industrial area of the Ruhr district. Another one year period included measurements in rural areas of southern Bavaria and on a Dutch island. The log-normally distributed deposition rates of cadmium at the rural areas in southern Germany amounted to only 20% of those of the industrial district. The depositions on the Dutch island were twice as high as the depositions on the rural areas of southern Germany. The monthly cadmium deposition rates show only little periodical fluctuation during the year and scatter around more or less constant median values of 25 and 120 $\mu\text{g} \cdot \text{m}^{-2} \cdot \text{month}^{-1}$ at the rural and industrial areas, respectively. When open air mass cultures of algae were taken as an agricultural model, the organisms, depending on their growth rate, accumulated 0.4–4.0 ppm of cadmium (dry matter based). The course of the cadmium accumulation reflects the deposition rate of the area where the algae were grown. No growth depression of the algae due to cadmium can be observed under the given deposition rates.

I. INTRODUCTION

Among the pollutants of our environment, cadmium (Cd) has become increasingly hazardous to human health. In addition to inhalation of contaminated air (2%), man receives more than 95% of his Cd burden by ingesting contaminated food (LAUB, 1980; Käferstein *et al.*; 1979, UBA, 1977). This Cd burden reaches man either directly by consumption of agricultural products (60%) or via meat and other animal products (40%, Wosing-Narr and Musche, 1977), which depend mainly on feed of agricultural origin. Hence, the sources by which agricultural products are contaminated by Cd are of great importance if the Cd uptake by man is to be controlled. According to a recent study by UBA (1980), 90% of soil Cd is due to atmospheric fallout and input by fertilizers, each contributing approximately equal amounts. The significance of this Cd input into soil depends on its availability to the agricultural plants (Kloke, 1980).

In order to study the possible significance of Cd input into agricultural systems, we measured monthly deposition rates of Cd at different locations and the uptake of this Cd by plants. The latter was composed of an intensive hydroponic culture of microalgae exposed to the open air. Such a model culture permits frequent and homogenous sampling during the vegetation period and reproducible conditions for the control of inputs and outputs. Therefore, conclusions are possible concerning the Cd transfer within agricultural production systems.

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II. MATERIALS AND METHODS

A. Sampling

Monthly samples of dust deposition were collected according to a modified Bergerhoff method (VDI, 1972, Herpertz, 1969). Beakers of glass or Teflon were initially tested for Cd release, which was found to range from 0.5 to 1.5 μg before cleaning; glass containers of higher Cd release (up to 10 μg per beaker) were eliminated. Beakers of both glass and Teflon, precleaned in hot 10% HNO_3 showed Cd release of 0.01 μg . When exposed at the sampling station, beakers of 62- to 68-cm² opening area and of 1200- to 1500-ml volume were partly filled with 3% HNO_3 (100–500 ml depending on the expected climatic conditions) in order to reduce evaporation and losses by wind.

B. Sample Preparation

The deposition samples were not passed through a screen, but samples containing large numbers of insects were eliminated, if this was indicated by the Nalimov outsider test of the measured values (Kaiser and Gottschalk, 1975). The samples were then transferred to quartz glass containers and concentrated to near dryness; 4 ml acid (65% HNO_3 analytical grade + 35% HCl analytical grade, 1:3) was added and heated for 10 min at 100°C. The acid was then evaporated and the residue was dissolved and made up to 10 ml volume by adding 0.5 N HCl . After centrifugation for 6 min at 3000g in Teflon tubes, the supernatant was used for analysis by flameless atomic absorption spectrometry (AAS). Since ashing in an oven at 450°C (Runkel and Baak, 1972) and treatment by hydrofluoric acid did not alter the results, the simpler treatment by HNO_3/HCl was used throughout the investigation.

C. Analytical Precision and Accuracy

The analyses for Cd of 326 deposition samples were performed by the standard addition method of flameless AAS, with an instrument detection limit of 0.2 $\mu\text{g}/\text{liter}$ of Cd. The overall precision of sample treatment and analysis was found to be $\pm 10\%$ and $\pm 20\%$ at deposition rates of 4 and 1 $\mu\text{g} \cdot \text{m}^{-2} \cdot \text{day}^{-1}$, respectively. Twelve blank samples resulted in a value of $0.06 \pm 0.04 \mu\text{g} \cdot \text{m}^{-2} \cdot \text{day}^{-1}$ corresponding to a detection limit of 0.18 $\mu\text{g} \cdot \text{m}^{-2} \cdot \text{day}^{-1}$ and a final sample concentration of 4 $\mu\text{g}/\text{liter}$.

The analysis for Cd in the microalgae followed the method of dry ashing as described before (Runkel, 1975; Runkel and Baak, 1972). The technique used had a precision of $\pm 1.5\%$ at an algae Cd content of 5 $\mu\text{g}/\text{g}$ (flame technique) and $\pm 12\%$ at 0.05 $\mu\text{g}/\text{g}$ (flameless AAS).

The accuracy of the analysis was verified by using standard reference material (Spinach NBS 1570, indicated Cd content: 1.5 $\mu\text{g}/\text{g}$), the Cd content of which was determined as $1.44 \pm 0.06 \mu\text{g}/\text{g}$.

D. Collection Sites and Period

The deposition samples were collected at 16 locations of four regions (Fig. 1). Three stations in Bayern, which were located inside of the GSF area, and in the rural areas of Grünbach (Erding) and Altenstadt (Schongau), were operated during 1977. Seven stations were established in 1975 in the Pfalz at Grünstadt-Ponderosa, Sie-

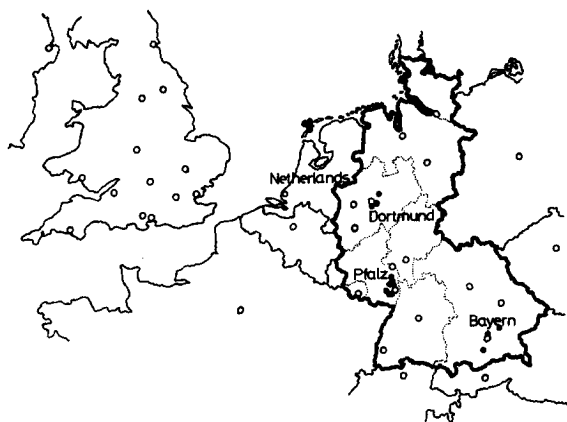


FIG. 1. Sites where the atmospheric depositions were collected (black dots).

beldingen-BFA, Hanhofen, Obersülzen, Heidesheim, Landau-Queichheim, and at Ilbesheim. The former three stations were observed for 3 years; the latter four stations were cancelled after a 2-year collection period when it was obvious that all seven stations gave practically the same results. Three stations were established in 1974 at Dortmund within the industrial area of the Ruhr district of NRW; after the first year we reduced the number of stations to 2, which were observed until 1977. Another three stations were observed during 1975 on Vlieland, a Dutch island which erroneously was considered free of industrial influence. However, as can be seen from Fig. 1, these islands are located between the two industrial areas of SE Great Britain and of the Ruhr district, and hence may receive emissions from either direction.

E. Algae Cultures

Mass cultures of the unicellular algae *Scenedesmus obliquus* (strain 276-3a of the Culture Collection, Göttingen, Germany) were maintained in several basins of 80-m² culture area exposed to the open air. They were harvested and treated as described by Stengel (1970). Minerals were supplied to these cultures as inorganic chemicals (analytical grade) or as phosphate fertilizer prepared for commercial use. All inputs including rainfall and supply water were carefully recorded. The samples for analysis were aliquots of 2 to 5 g of 158 algae samples harvested at different times during a 2-year period.

F. Evaluation of Data

For the purpose of comparison with data contained in the literature, the monthly deposition rates were transformed to daily values. Since all data populations investigated here proved to have a log-normal distribution (Figs. 3, 11), the usual statistical tests for outsiders (Nalimov) or for significance (Student's *t* test) were performed by using the normally distributed logarithms of the data. Statistics were calculated according to usual procedures (Kaiser and Gottschalk, 1975). For the reason of practical relevance the deposition rates were compared on the basis of arithmetic mean values, whereas the Cd contents of the algae were compared on the basis of median values as described earlier (Payer *et al.*, 1975).

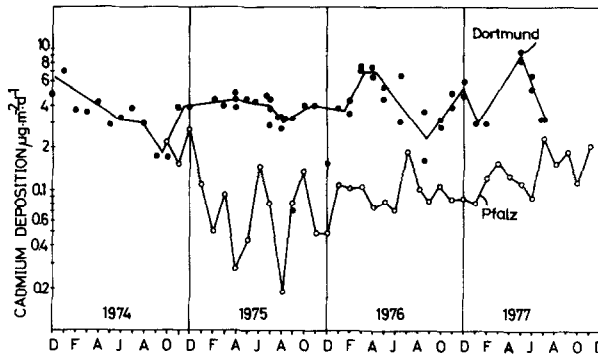


FIG. 2. Course of the monthly cadmium deposition rates at Dortmund (single samples) and in the Pfalz (median values of 3–7 samples).

III. RESULTS

A. Deposition Rates of Cd

Depositions of Cd in Dortmund and in the Pfalz are compared in Fig. 2 during a 4-year period. The data from Dortmund show individual samples and therefore give an impression of the variability of the data. The data measured at different locations of the Pfalz were pooled to median values since there was no statistically significant difference between the data of these stations. At neither the rural nor the industrial area a tendency of the Cd depositions could be established during the observation period. The analysis of the data as shown in Fig. 3 results in clear log-normally distributed deposition rates which, in the rural area, were only one quarter of those of the industrial area.

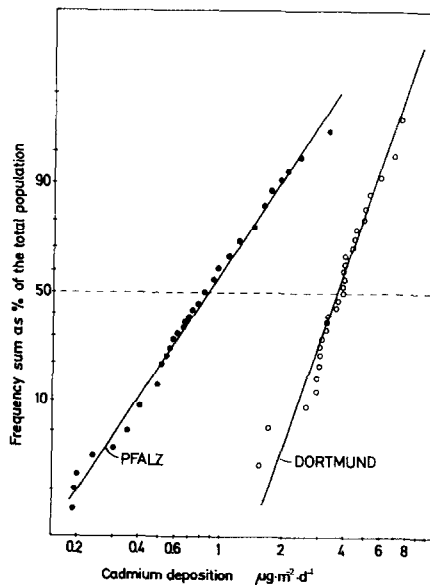


FIG. 3. The log-normal distributions of the atmospheric cadmium fallout in two different regions of Germany.

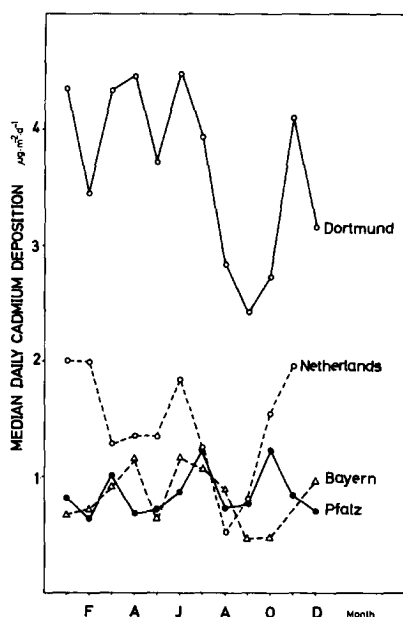


FIG. 4. Seasonal variations of the atmospheric cadmium fallout in four different regions.

In order to check for seasonal variations, the annual course of Cd deposition rates of four areas were compared by their median values in Fig. 4. Seasonal variations were not very strong except a depression of Cd deposition from August to October in Dortmund and in the island of The Netherlands. Regional differences are quite clear and statistically of high significance as far as the data of Dortmund and of The Netherlands differ from the rural areas of southern and western Germany. Bayern and the Pfalz however, have practically the same low deposition rates.

The annual mean deposition rates are summarized in Table 1. The results are complemented by additional data from a few deposition measurements made in Thailand. Beside the regional mean, deposition rates for comparison are also given on an annual basis per hectare, and the limits of our analytical method are indicated at the lower part of the table. The latter data show clearly that all investigations were well above these limits. The rural areas of Bayern seemed to have less Cd deposition than the near urban area of the GSF. Since the number of samples is not sufficient to prove the difference statistically significant, it might be useful to take all results from Bayern as equal. It is quite clear from Table 1 that rural areas of southern Germany show only half the deposition rates of "clean" areas of The Netherlands and only one-fifth of the deposition rates of the industrial area. As shown in Fig. 5 where Cd concentrations in air ($\mu\text{g}/\text{m}^3$) and deposition rates ($\mu\text{g} \cdot \text{m}^{-2} \cdot \text{day}^{-1}$) of rural, urban, and industrial areas were correlated over a wide range, our results compare well with data in the literature (UBA, 1977; UBA, 1980).

B. Accumulation of Cd by Algae Cultures

1. *Growth versus Cd content of algae.* As shown in Fig. 6, there is a good negative correlation between the growth rate and the Cd content of the algae. The relation does not mean a growth depression of the algae by Cd, but, depending on the climatic

TABLE 1
DEPOSITION RATES OF CADMIUM CALCULATED FROM MONTHLY COLLECTIONS OF ATMOSPHERIC FALLOUT

	Results (mean values)		Statistics ^a		Collection data		
	$\mu\text{g} \cdot \text{m}^{-2} \cdot \text{day}^{-1}$	$\text{g} \cdot \text{ha}^{-1} \cdot \text{year}^{-1}$	Median $\times s^{\pm 1}$ ($\mu\text{g} \cdot \text{m}^{-2} \cdot \text{day}^{-1}$)	n^d	$P\%^b$	Collection period (years)	Number of stations
Altenstadt	0.7	2.8	0.68×1.7049	8	<0.5	1977	1
Erding	0.8	3.0	0.71×1.7799	11	<0.5	1977	1
GSF München	1.1	4.0	0.87×2.165	11	<10	1977	1
Bayern (combined)	(0.9)	(3.3)	(0.76×1.8836)	(30)	<0.1	1977	(3)
Pfalz	1.1	3.9	0.85×1.8651	180	<0.1	1975-1977	7
Netherlands	1.5	5.5	1.33×1.6761	30	—	1975	3
Dortmund NRW	3.9	14.2	3.68×1.3877	59	<0.1	1974-1977	2
Thailand (rural) ^c	1.3	4.7		3		1977	2
Blank	0.06 ± 0.04^d	0.21		12			
Detection limit	0.18	0.66					

^a The scattering factor s has to be multiplied with the median value in order to get the upper ($\text{m} \cdot \text{s}$) and the lower limit ($\text{m} \cdot \text{s}^{-1}$) of the 68% of the population; n is the number of samples analyzed.

^b P indicates the probability of error of the calculation by which the population of data is different from the data of The Netherlands. The differences between data from the Pfalz and from Bayern are statistically not different ($P > 10\%$).

^c Data included for comparison.

^d $\pm$ Standard error.

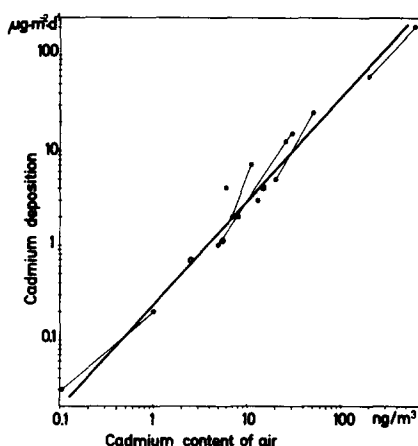


FIG. 5. Correlation between cadmium deposition rates and cadmium contents of air at different regions: black dots = data from the literature; connected dots indicate ranges reported in the literature (UBA, 1977, 1980); open circles = our own results combined with related data from the literature.

conditions, fast-growing algae accumulate less Cd than slowly growing algae. At the growth rate of $1 \text{ g} \cdot \text{m}^{-2} \cdot \text{day}^{-1}$ the Cd content of the algae expresses the daily input per square meter which is found to be approximately $4.5 \mu\text{g} \cdot \text{m}^{-2} \cdot \text{day}^{-1} = 16 \text{ g} \cdot \text{ha}^{-1} \cdot \text{year}^{-1}$. This input is derived almost entirely from atmospheric depositions since inputs by water and fertilizers are negligible at such a low growth rate.

2. *Influence of fertilizer.* Plant cultures accumulate Cd not only from atmospheric depositions but also from nutrients which are contaminated by Cd. Algae raised with pure chemicals (Fig. 7) contain 1.3 ppm less Cd than algae grown on commercial fertilizers (difference of the median values). This contribution of Cd by fertilizers is also evident from Fig. 8; different doses of fertilizer were added to our cultures. The increasing Cd content of the plants indicates a Cd content of 4.5 ppm Cd in the fertilizer. Direct analysis of the fertilizer resulted in 5.3 ppm Cd. The applied average

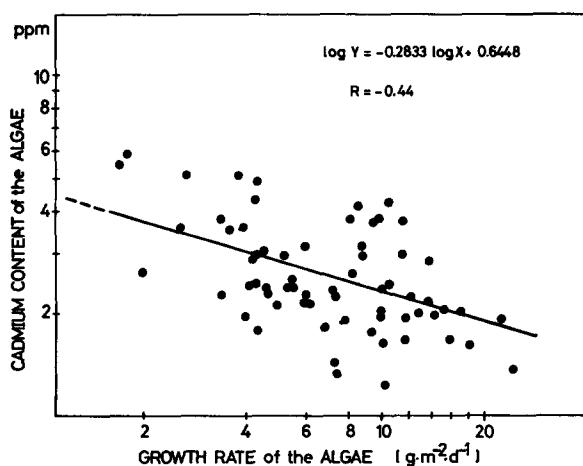


FIG. 6. Correlation between the growth rate of the algae and the accumulation of cadmium by the algae grown outdoors at Dortmund.

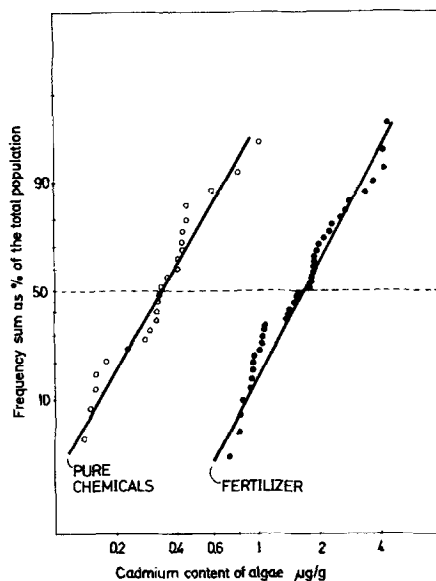


FIG. 7. The log-normal distributions of the cadmium contents of the algae grown with or without fertilizer.

fertilizer dose of $2.6 \text{ g} \cdot \text{m}^{-2} \cdot \text{day}^{-1}$ ($\text{P}_2\text{O}_5 = 0.67 \text{ g} \cdot \text{m}^{-2} \cdot \text{day}^{-1}$ equal to $250 \text{ kg} \cdot \text{ha}^{-1} \cdot \text{year}^{-1}$; usual application in agriculture: $50 \text{ kg} \cdot \text{ha}^{-1} \cdot \text{year}^{-1}$) corresponds to a Cd input of $14 \mu\text{g} \cdot \text{m}^{-2} \cdot \text{day}^{-1} = 51 \text{ g} \cdot \text{ha}^{-1} \cdot \text{year}^{-1}$ which is now the major source of Cd for this type of plant culture.

3. *Influence of irrigation.* In order to trace the influence of supply water upon the Cd contamination of our cultures, we divided the samples according to a two-factorial experimental design in which rain water and tap water supply were separated as the two factors. The four populations are shown in Fig. 9. The insert indicates the median Cd content of these four populations. The statistical evaluation results in a significant contribution of 0.7 ppm of Cd by tap water. The small contribution of Cd by rain

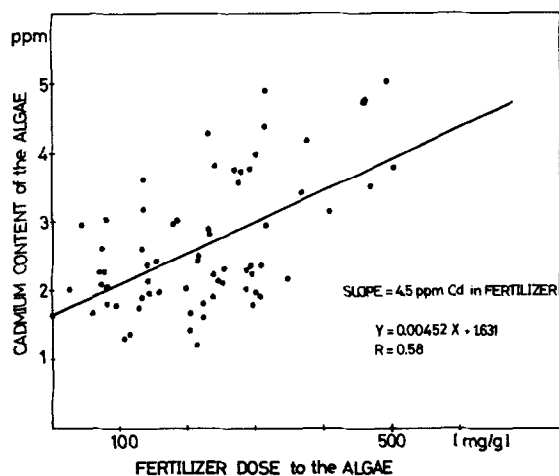


FIG. 8. The influence of the phosphate fertilizer on the cadmium contents of algae grown outdoors at Dortmund.

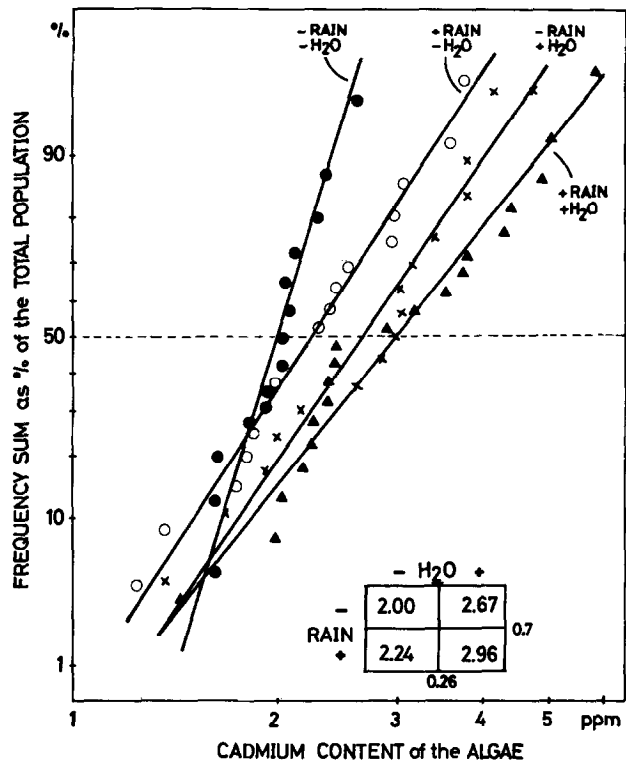


FIG. 9. Four different populations of microalgae receiving cadmium by rain, by tap water, by both rain and tap water, or by neither one (= cadmium from air and fertilizer only). The insert summarizes median values and mean differences of the four populations analogous to a two-factorial analysis.

water is statistically not significant. The dose-dependent Cd accumulation by algae, which is shown in Fig. 10, indicates a Cd content of tap water of 0.7 $\mu\text{g/liter}$, comparing well with a median Cd content of 0.6 $\mu\text{g/liter}$ obtained by direct analysis of 17 water samples of Dortmund tap water (Fig. 11). If the mean water supply to the

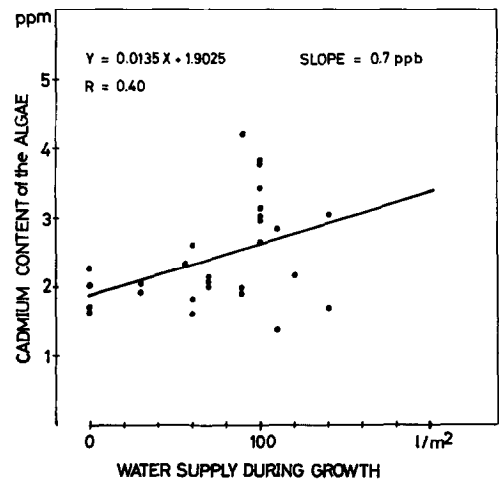


FIG. 10. The influence of supply water on the cadmium content of algae grown outdoors at Dortmund.

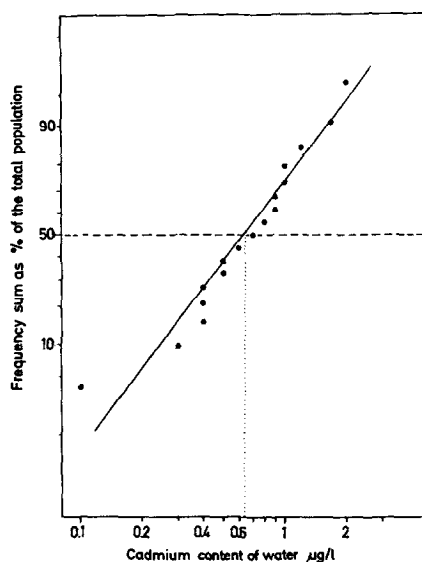


FIG. 11. The log-normal distribution of cadmium contents of tap water sampled at Dortmund.

algae cultures of $10 \text{ liter} \cdot \text{m}^{-2} \cdot \text{day}^{-1}$ is taken into account, this corresponds to a Cd input of $6 \mu\text{g} \cdot \text{m}^{-2} \cdot \text{day}^{-1} = 22 \text{ g} \cdot \text{ha}^{-1} \cdot \text{year}^{-1}$.

IV. DISCUSSION

A. Deposition Rates of Cd

As already indicated by Fig. 5 our measurements of Cd depositions are well in agreement with data of other authors. The four to five times higher depositions in the industrial Ruhr district of NRW compared to rural areas of southwest Germany were already indicated by our preliminary investigations concerning the atmospheric fallout of Cd, Pb, and Fe (Payer and Runkel, 1978). Further analyses of the depositions of Mn, Zn, and Cu confirm this tendency of heavy metal precipitation in Germany (Payer *et al.*, in preparation).

Since Cd in air is bound mainly to very small particles (Müller, 1981), it will be subject to long distance transportation. This may explain the relatively high deposition rates found at the Solling, 140 km east of the Ruhr district (Ulrich *et al.*, 1976) and at the Dutch island included in our investigation. The latter may get part of its depositions from industrial Great Britain, 300 km south west (Mc Innes, 1978) as well as from the Ruhr district 200 km to the southeast. At least some relation between the Cd depositions of The Netherlands and of the Ruhr district is indicated by the common seasonal depression of Cd depositions from August to October; this variation is not significantly observed in the rural areas of southern Germany.

As mentioned by Müller (1981) and confirmed by measurements of Nürnberg (1981), about 80% of such fine particles are precipitated by rain and found as Cd deposition in the washout. The difference between total Cd deposition and the washout by rainfall seems to be not only a numerical percentage; it also influences the course of seasonal variations of the Cd deposition, and therefore, it cannot be neglected.

B. Accumulation of Cd by Microalgae

As shown earlier (Payer *et al.*, 1976), microalgae cultures are useful for monitoring heavy metals from the environment. If the actual measured inputs and the mean accumulations of Cd by algae are compared (Table 2) the conclusion is that all depositions of Cd are quantitatively absorbed by these plants. The comparison is based on a mean growth rate of $10 \text{ g} \cdot \text{m}^{-2} \cdot \text{day}^{-1} = 36 \text{ tons} \cdot \text{ha}^{-1} \cdot \text{year}^{-1}$. This high productivity requires high input from fertilizer which is approximately five times that used in conventional agricultural practices.

C. Impact of Cd Depositions on Soil and Grain

If we calculate the total input of Cd to soil by combining our measured deposition data with inputs from fertilizer and other data as reported in the literature (UBA, 1980; Stenström and Vahter, 1974), agricultural soils in rural Bayern as well as in the Pfalz receive so-called "normal" amounts of Cd (Table 3). The total input of 10 to $12 \text{ g} \cdot \text{ha}^{-1} \cdot \text{year}^{-1}$ compares well with the actual increase of $12 \text{ g} \cdot \text{ha}^{-1} \cdot \text{year}^{-1}$ calculated from soil Cd contents for 1930–1970 in the European Community (UBA, 1980). The total Cd input to soils of industrial areas is about double that of rural areas and compares approximately with the $20 \text{ g} \cdot \text{ha}^{-1} \cdot \text{year}^{-1}$ Cd input measured by Ulrich and co-workers in the Solling (1976). Whereas industrial areas are contaminated mainly by atmospheric Cd fallout, rural areas receive most of their Cd by fertilizers. Either source may contribute quite an amount of Cd to ground water from where it is again available to lakes, rivers, tap water, and irrigation systems.

Provided all inputs to soil are absorbed by agricultural plants as was found for our model plant, one may find 0.2–0.4 $\mu\text{g/g}$ of Cd in grain if the transfer factor of 10% from cereal plant to grain is realistic. Although complete absorption of Cd by higher plants is not the rule, such high Cd contents were actually found in several grain samples especially from NRW (Ocker, 1977). The mean Cd contents of grain analyzed in the same study also reflect the regional differences of deposition rates. The estimated possible maximum values are still not yet reached (Table 3) but according to analyses of wheat of NRW (Markard 1981) and of cattle kidney of even rural Bayern (Kreuzer *et al.*, 1976), 40 and 17%, respectively, of the samples already exceed the set limit of 0.1 and 0.5 $\mu\text{g/g}$, respectively. These examples indicate the significance of the actual

TABLE 2
BALANCE OF DEPOSITIONS AND CONTAMINATIONS BY CADMIUM

Source	Deposition rate ($\mu\text{g} \cdot \text{m}^{-2} \cdot \text{day}^{-1}$)			Accumulation by algae ^a ($\mu\text{g/g}$)		
	Dortmund	Thailand		Dortmund	Thailand ^b	
Air + rain	4	1.3	1.3	0.5	not determined	
Tap water	6	2.2	2.2	0.7	not determined	
P-fertilizer	14	14	0	1.3	1.3	0
Total	24	17.5	3.5	2.5	1.7	0.35

^a Mean growth rate of the algae: $10 \text{ g} \cdot \text{m}^{-2} \cdot \text{day}^{-1}$.

^b Data partly taken from Payer *et al.* 1976, partly unpublished.

TABLE 3
IMPACT OF CADMIUM DEPOSITIONS ON SOIL AND GRAIN

	Inputs ($\text{g} \cdot \text{ha}^{-1} \cdot \text{year}^{-1}$)				
	Normal (UBA, 1980)	Bayern (rural)	NRW (Dortmund) (industrial)	Maximum/limits (UBA, 1980)	
Air	5	3 ^a	14 ^a	27	9
Fertilizer	5	5	5	13 ^c	(11)
Sludge	1	1	1	75	75
Other	1	1	1	—	
Total input	12	10	21	115	95

	Sorption by grain ($\mu\text{g/g}$)				
	Calculated ^b	Bayern	NRW	Maximum	Limits
Calculated ^b	0.25	0.2	0.4	2.3	1.9
Measured ^d					
Mean	0.06	0.05	0.10		
Maximum	0.4	0.4	0.4	0.4	0.1

^a Our own data from this paper.

^b Calculated for a yield of $5 \text{ tons} \cdot \text{ha}^{-1} \cdot \text{year}^{-1}$, 10% of Cd in grain.

^c Stenström and Vahter, 1974.

^d Ocker, 1977.

Cd inputs by both fertilizer and atmospheric fallout which need more control in future.

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