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THE CONTAMINATION OF DUTCH SOILS WITH MERCURY AND A FEW OTHER HEAVY METALS

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ABSTRACT

As a result of his history the average dutchman knows the various soil types in his country quite well, the chemical structure is usually unknown. The paper describes the typical aspects of the soil constituents as there are: clay minerals, sands and organic materials. Specially the properties which control the adsorption of traces of heavy metals and pesticides got attention.

An important part of the paper is devoted to the behaviour of mercury in dutch and other western european soils. Because part of the drinking water companies in the western part of Holland produce their drinking water from the River Rhine by filtration through dunes, the migration of mercury in soils got serious attention. Most mercury compounds (Hg, $HgCl_2$, $HgClCH_3$) do not migrate in soil, the volatile Hg $(CH_3)_2$ migrates fast. The contamination of soils is partly caused by agricultural measures, partly by flooding with water from the Rhine. An analysis of the available data indicated, however, that the main mercury source stems from the wash out of the atmosphere. The contamination of soils in areas with a high population density and much industry is considerably higher than in other areas. A similation model was developed to describe the mercury fall out drain rate.

INTRODUCTION

The average dutchman knows quite well the difference between the various soil types: Clay stands for high production, no recreation possibilities. Sand for forests and heath, although the latter is unfortunately reclaimed for agricultural purposes. Peat provides many possibilities for fishing and boating in the lakes and goes with ditches which are always present in such areas.

But the chemical structure of various soils is, also for many chemists, unknown. Of course, it is true that soil is a most unknown mixture of chemical compounds, but it is certainly possible to give a more exact characterization of a soil, then by a statement as a "typical clay mud", which, not long ago, I noticed in a paper on contamination with heavy metals.

Soils consist out of a mixture of organic and anorganic materials. Fresh organic material in a soil may, during decomposition, slowly release nitrogen, with its beneficient effects on crops. Besides that, fresh organic material has a ion exchange capacity for cations which goes up to 3 milli equivalents per g, the exchange capacity for anions is certainly a factor 10 lower. Older organic material, which hardly decomposes, is called humus, the structure of which is partly known; it contains mainly ring structures. The exchange capacity is much lower than of fresh organic material. Also the capacity to retain water is lower for humus then for fresh organic matter, although the possibility to retain water is one of the most important properties of humus.

The anorganic material consists of particles of various dimensions. The coarse particles consist (in the Netherlands) mainly of quartz (sandy soils, beaches), the smaller particles of clay minerals (clay, loess). The dominant clay mineral in the Netherlands is illite. Illite crystals consist of platelets, each platelet being build up out of two silica and 1 alumina sheet. Due to isomorphic substitution the crystal is negatively charged. The cation exchange capacity is 0.3 meq per g. The edges of the platelets are positively charged (at pH < 6), the anion exchange capacity is, however, verly low. The specific surface area of illite is about 10^6 cm^2 per g. Furthermore all soils contain traces of other minerals; in sandy soils mainly aluminium and iron hydroxides.

CATIONS

It will be clear that the capacity to adsorb all kind of constituents in a soil is extremely high, but that it varies from soil to soil. Cations are always adsorbed, which does not necessarily mean that they can not pass a certain layer of soil. Just as in a chromatographic column, adsorbed cations can be easily replaced by other cations so that the originally adsorbed cations reach again the water phase and thus may be leached downwards. Some of the thump rules, valid in chromatography are valid in soils too, as: divalent ions are adsorbed better than monovalent ions. There exist however many exceptions, for instance potassium ions are, due to

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their dimensions, adsorbed between the silica-alumina layers which form the illite crystal, which results in an extremely high preference of illite for potassium ions. Ammonium and caesium ions, which have similar dimensions as potassium, are also rather strongly adsorbed by illite. In soil science it is customary to call such an adsorption: fixation. From a kinetic point of view fixation can be defined as an adsorption with a very high adsorption rate and a very low desorption rate. Natrium, calcium and magnesium ions show no abnormal behaviour, with strontium ions some fixation exists.

From the heavy metals it is mercury which is fixed very strongly. The adsorption of chromium depends very much on the soil type. Zinc passes a layer of soil without difficulty.

NON-IONIC SUBSTANCES

For non-ionic substances, to which belong a number of heavy metal complexes like heavy metal-fulvic acid and heavy metal-humic acid, there are no general rules. As can be expected, the adsorption on soils rich on humus is much better than on other soils. Desorption from humus hardly occurs. Only in exceptional cases nonionic substances will pass a sandy soil without adsorption. Contrary to the adsorption on humus, the adsorption on sand is rather reversible. Metallic mercury is adsorbed strongly by soils.

ANIONS

Some anions pass a layer of soil rather easily, for instance chloride ions, but also an anion as $Co(CN)_6^{3-}$ is hardly adsorbed. For sulfate ions all kind of references can be found; usually it is adsorbed, but only in small quantities. Phosphate ions do not have much chance to pass a soil; sometimes they are adsorbed as anion, but in general they are precipitated as aluminium or iron phosphate. Nitrate ions are not adsorbed in soil, but nitrogen may be fixed by bacteriological processes so that no leaching occurs. This subject will not be discussed here.

THEORY

I have mentioned the term chromatography a few times. Those familiar with chromatographic theories as of Glueckauf or Hiester and Vermeulen will wonder if these theories can be applied to soils or not. The fact that the soil is heterogenous, that the supply of chemicals and transport medium (water) is irregular and that various processes play at the same time a role allow, of course, no analytical solutions of the relevant differential equations. Modern numerical methods, as for instance mathematical computer simulation can, however, be used with success. Also if very complex situations have to be considered, simulation models can be a great help. One warning must be sounded: from a mathematical point of view, it is not so difficult to include numerous parameters, or to use such parameters as function of time and location, but the difficult point is the enormous quantity of data which is required. As simulation is my personal hobby. it is better not to discuss it on this place, I will only refer to the monograph to be published by PUDOC: Simulation of Leaching and Accumulation in Soils.

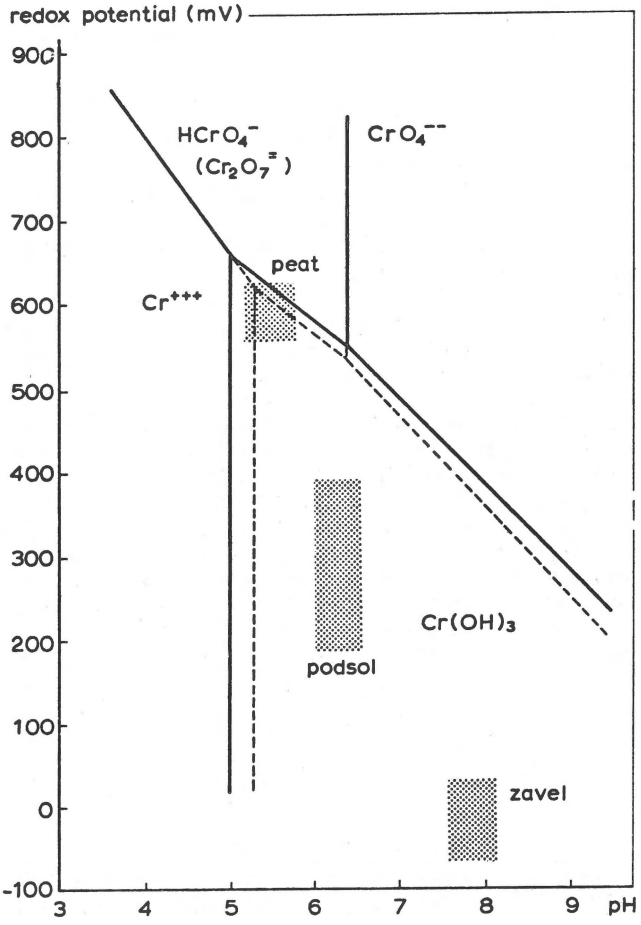

Fig. 1
Redox potential-pH diagram for Cr.
Full lines : boundaries 10^{-3} molar (50 ppm)
Dotted lines: boundaries 10^{-4} molar (5 ppm)
Regions for three soils are indicated, for anaerobic conditions the redox potential may go down to -200 mV.

SOME HEAVY METALS

Chromium may in exceptional cases pass a soil as cation, as chromate anion it is usually adsorbed. Fig. 1 shows the relation between the various forms of chromium as function of redox potential and pH. Values for three representive soils are indicated. The redox potential for water logged soils is usually lower (-130 to -200 mV). It appears that soil

table 1: Description of soils and mercury contents.

Nr.	location	Soil identification		Mean Hg content ppm 0-20 cm	20-100 cm
1	Alkmaar, Neth.	alluvial gley	(clay)	0.09	0.02
2	Hilversum, Neth.	podsol	(sand)	0.09	0.01
3	Amersfoort, Neth.	podsol	(sand)	0.09	0.01
4	Schoonebeek, Neth.	fen	(peat)	0.07	0.04
5	Ahrweiler, FRG.	pseudo gley	(clay)	0.05	–
6	Hannover, FRG.	podsol	(sand)	0.06	–
7	Amiens, France	rendzina	(calc. soil)	0.04	–
8	Amiens, France	para brown earth	(loess)	0.05	–
9	Bari, Italy	terra fusca	(red soil)	0.02	–
10	Ispra, Italy	pseudo gley	(clay)	0.10	–
11	Hillegom, Neth.	dune	(sand)	0.16	0.10
12	Den Helder, Neth.	reclaimed sea soil	(sand)	0.13	0.05
13	Valburg, Neth.	alluvial	(clay)	3.4	0.45
14	Biesbosch, Neth.	alluvial	(peaty clay)	10.4	2.6
15	Dordrecht, Neth.	alluvial	(clay)	0.35	0.26
16	Leiden, Neth.	alluvial, irrigated dune		0.16	0.01
17		alluvial, irrigated dune		0.33	0.07
18		alluvial, non irrigated dune		0.03	0.07

conditions are sometimes such that they are close to the triple point for chromium, thus making the existence of Cr^{+++}, Cro_4^{--} and $Cr(OH)_3$ all possible. Moreover because the adjustment of the equilibrium goes slowly, it will be clear that a prediction of the behaviour of chromium is hardly possible.

Better possibilities exist for the prediction of the fate of zinc. Zinc is adsorbed as cation, but the greater part is desorbed rather easily. That means that zinc, unless it is supplied in small quantities, passes a layer of soil finally. If a continuous supply with water contaminated with zinc occurs – irrigation with sewage water, for instance – first there will appear no zinc, thereafter the concentration goes up till eventually the zinc concentration of the influent is reached. Cadmium is another element which nowadays gets much attention.

Its cation will be adsorbed on both humus and clay minerals. Specially at high pH values, that means for a soil pH $>$ 7.5, the capacity to adsorb cadmium is rather high. An excess of other cations will desorb cadmium.

MERCURY

The heavy metal which got far most attention in the last years is mercurry. At our institute we studied the behaviour of mercury in soils intensively because mercury is used since 50 years as a fungicide. It was therefore already included in the Association's environmental protection programme before the alarming sound of poisoning of seals emerged.

Also for some drinking water companies in The Netherlands mercury is of special concern. Those companies which take their water from beneath soils on which bulbs are cultivated are of course worried by the use of fungicides. But also the "Duinwater" companies which use the River Rhine, directly or indirectly, as source for their water are not at ease because the Rhine is contaminated with mercury. Our investigation on mercury can be divided into three types of approach

- Determination of mercury profiles in the field
- Migration experiments on columns with natural profiles under well defined conditions in the laboratory.
- Development of simulation models which explain the just mentioned lines.

SURVEY OF THE MERCURY CONTENT OF A NUMBER OF SOILS

The mercury content as a function of the depth has been determined for 18 soil profiles. Included are foreland soils from the River Rhine, soils from the bulb growing area, irrigation canals in the dunes and permanent pastures, the latter to act as references.

In Table 1 the sites are listed according to location, soil identification and mercury content. Sampling was done up to a depth of 100 cm in the Netherlands soils, and up to a depth of 25 cm in the soils from other countries. The total mercury content was determined according to Melton et al. (1971), by flameless atomic absorption spectrophotometry, or by neutron activation analysis. The results are presented in Fig. 2 and Table 2.

The soils numbered 1-10 are permanent pastures and may, just as soil 18, be seen as reference soils. In these soils no mercury has been introduced by applying fungicides or by flooding of surface water contaminated with mercury; no

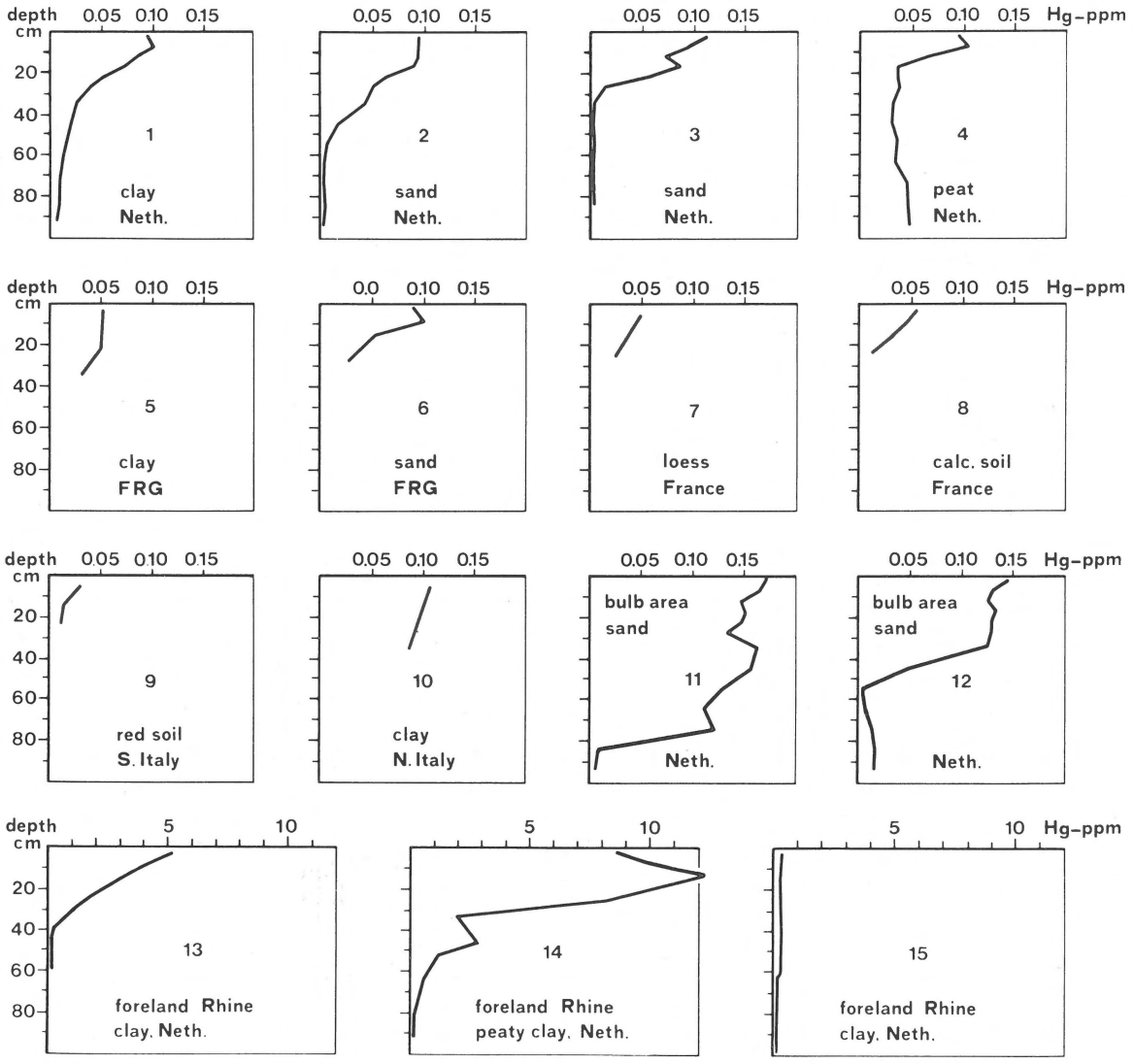

Fig. 2
Mercury concentration in some Western European soils as a function of the depth.

profile disturbances have occurred during the last 20 years. These reference soils differed widely in soil characteristics, and included calcareous and non-calcareous soils. Their clay content ranged from 2 to 20%, their organic matter content from 2 to 12%. A peat soil (soil no. 4) had an organic matter content of more than 95%. The mercury content in the top 20 cm of these soils ranged from 0.02 ppm to 0.10 ppm, and averaged 0.07 ppm. This value is in agreement with values found in soils elsewhere and seems to represent the natural background level. The distribution over the profiles revealed that the mercury content diminished with depth rather quickly; below 20 cm it was never more then 0.04 ppm. The "distribution over Europe' suggests that the mercury content is higher in regions with high industrial activity e.g. at sampling sites nos. 1, 2 and 3 in The Netherlands and no. 10 in Italy. The difference in mercury content of the various locations cannot be explained by differences in soil characteristics as there is no possible relation between mercury content and clay content, organic matter content or pH. The differences can only be explained if we accept the existence of mercury fallout originating from man's activities.

The second group of soils (numbers 11 and 12) consists of two soils from the bulb-growing area in Holland. The application of mercury as a fungicide in these areas has been common practice for 50 years, and the use was intensified about 20 years ago. The soils had a mercury content of about 0.15 ppm in the top 20-cm layer and a distribution pattern over the profile quite different from those of the reference soils (Fig. 2). The mercury content hardly changed with depth but diminished abruptly at a depth of about 80 cm for

Table 2: Mercury content of soil samples of irrigated dunes at Leiduin, Gemeentewaterleiding Amsterdam.

Location				Mercury content in ppm (mg/kg dry soil)
Infiltration canal 7	Dry sludge	0- 2 cm	Infiltration canal was partly dried up	1.14
	Wet sludge	0- 2 cm		1.12
	Grey sand	2- 7 cm		0.13
	Grey sand	7-15 cm		0.018
	Light grey sand	50 cm	Above the sampling sites wet sludge	0.008
	Light yellow sand	75 cm	was present	0.009
	Light yellow sand	100 cm		0.008
	Light yellow sand	150 cm	Contained iron	0.003
Infiltration canal 8	Grey wet sand	0- 5 cm		0.29 ± 0.14
	Grey wet sand	5-15 cm		0.35
	Grey wet sand	15-40 cm		0.12
	Grey wet sand	75 cm		0.039
	Grey wet sand	100 cm		0.038
	Yellow wet sand	150 cm		0.018
Drainage canal	Grey wet sand	0-15 cm		0.014
Non irrigated area (Vogelenveld)	Very dry sand	0-10 cm	Contained humus	0.043
	Very dry sand	20-30 cm	Contained iron	0.011

The standard deviation of the chemical determination varies from 0.020 to 0.002 ppm. The standard deviation presented is caused by the unequal distribution of mercury in the soil.

soil no. 11 and at a depth of about 40 cm for soil no. 12. It is common practise in the bulb area to plough occasionally to a depth of 40 cm, but sometimes even to 100 cm depth. These ploughing depths were found back in the distribution pattern of the mercury.

It is obvious that mercury accumulation did occur in these soils, the accumulated amount is, however, not excessive. Leaching seems to be absent.

The soils number 13-15 are foreland soils from the River Rhine. The Rhine is heavily polluted and discharges into the North Sea about 70 ton of mercury annually. The foreland soils are frequently flooded, usually in winter. The soils sampled in these areas showed mercury contents of over 10 ppm. Obviously the mercury stems from the Rhine and is strongly adsorbed by clay. Profile no. 15 is situated in an area reclaimed 50 years ago, which till then had been frequently flooded with Rhine water. It shows not only that 50 years ago the Rhine was already polluted with mercury, but also that even after 50 years the mercury is still present in the soil. Neither leaching, nor evaporation seem to play an important role. It looks that mercury once being in the soil, sticks to that soil for a long time.

The results of the soils number 16-18 are presented in table 2. Accumulation of mercury took place in the top soil of the irrigation canals. Also the sludge held back a considerable amount of the mercury. At a depth of 1 meter the mercury level was not strongly different from that of other soils at comparable depth. Also a sample taken from the drainage canal showed a very low mercury concentration. So it seems that accumulation only takes place in the top soil layer of the irrigation canals. The possibility that drainage water is contaminated with mercury is therefore very low. Mercury compounds which do not adsorb at all are, of course, not held back by soils; we are, however, not aware of the existence of such compounds.

TRANSPORT EXPERIMENTS IN THE LABORATORY

Transport experiments were carried out in the laboratory under controlled conditions with radioactive γ-emitting mercury compounds in columns with undisturbed soil profiles. The columns were 100 cm long with an inner diameter of 12 cm and were provided with rain heads so that solutions, in the form of uniformly distributed small droplets (0.05) ml), could be added to the column. In this way the soils were not saturated with water. Automatic control units maintained a proper supply of the influent. A scanner has been developed to scan a column for γ-radiation and thus record the distribution pattern of the compound without destroying the column. Transport experiments were carried out with this technique on soils from the bulb-growing area in Holland, for the investigation of ^{203}Hg labelled dimethyl mercury, monomethyl mercury chloride, mercury chloride and metallic mercury.

After labelling the column was scanned to check the distribution of the label. Then leaching was started with a flux of 24 mm per day with an artifical soil solution (0.03 N $CaCl_2$, 0.001 N KCl, and 0.001 N NaCl). The use of distelled water should at such a high flux, completely leach the soil and thus disorientate the soil particles). Air was sucked continuously over the top of each column to remove volatile

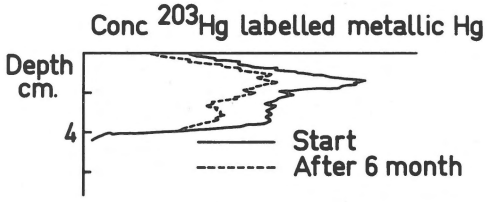

Fig. 3
Result of leaching experiments in a column with ^{203}Hg labelled metallic Hg.

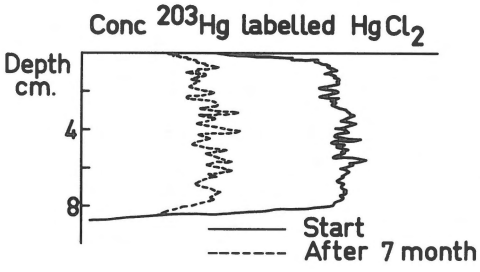

Fig. 4
Result of leaching experiments in a column with ^{203}Hg labelled $HgCl_2$.

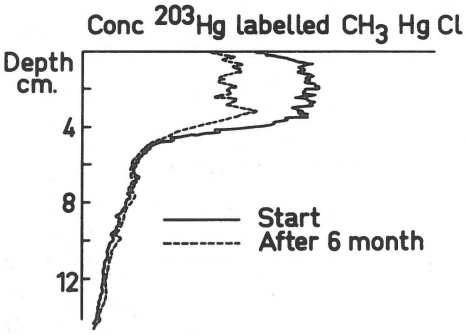

Fig. 5
Result of leaching experiments in a column with ^{203}Hg labelled CH_3HgCl.

Table 3: Volatilization of mercury.

Mercury compound investigated	Volatilized Hg as percentage of Hg added during the experiment		Duration of the experiment (days)
	continuous leaching	interrupted leaching	
$HgCl_2$	0.05	0.03	302
ibid	0.08	0.03	
CH_3HgCl	0.91	0.73	199
ibid	0.78	0.58	
Hg (metal)	0.18	0.53	139
ibid	0.62	0.98	

compounds which might escape from the soil. These compounds were then trapped. The columns were scanned monthly. Four columns were used for each mercury compound mentioned. Two of the four columns were leached continuously, the other two periodically, usually 16 hours a week, keeping the net flux equal for all colimns. The temperature was kept at 20°C. The results of these experiments are recorded in figs. 3-5 and table 3.

From the figures it is obvious that migration of metallic mercury and $HgCl_2$ does not occur, CH_3HgCl was leached downwards a little between the moment that the compound was added to the soil and the first measurement, thereafter the migration was almost negligible. Table 3 shows that evaporation was slow for all the compunds, whether they were leached continuously or periodically. Differences do exist, for example the rate of evaporotion for $HgCl_2$ was much slower than for CH_3HgCl. In general, however, these experiments confirm our statement that mercury, once it is present in the soil, sticks to that soil for a long time.

As dimethyl mercury is highly adsorbed on perspex, PVC, rubber, grease and even Teflon, the installation could not be used for transport studies of this mercury compound. An all glass installation had to be used, therefore the transport studies had to be limited to disturbed soils.

A 20 cm glass tube was filled with soil. ^{203}Hg-marked dimethylmercury was injected at the bottom of the tube, while a continuous stream of nitrogen gas (saturated 80% with water) was blown through the top of the tube, keeping the dimethylmercury concentration there close to zero. The volatilization was determined by measuring the decrease of the ^{203}Hg activity at the injection point as a function of time.

In consecutive experiments the tube was filled with various soils.

In fig. 6 the results of these measurements are shown as the percentage of the injected dimethylmercury present at the injection point as a function of time.

It appears from this figure that without soil all dimethylmercury applied has evaporated and diffused over a distance of 20 cm in about 150 hours. A soil layer of approximately 20 cm more than doubles this time.

ORIGIN OF THE CONTAMINATION OF PASTURES WITH MERCURY

A quick glance on the data shows that the mercury concentration from a depth of 60 cm is on average close to 0.01 ppm, while the concentration in the upper layers is much higher. This indicates that the mercury content of the upper layers must be caused by an addition of mercury to the soil. Because the pastures have never been treated with mercury their contamination must stem from elsewhere. As many mercury compounds are rather volatile it seems probable that evaporated mercury is precipitated by rain and then sticks to the soil. The key question in such a reasoning

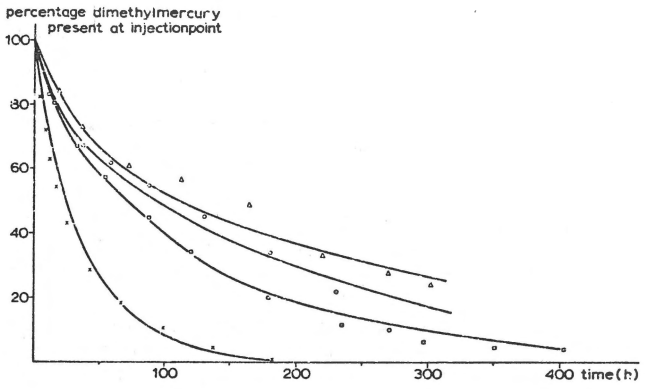

Fig. 6
Transport of dimethylmercury towards a soil surface. Injection point about 20 cm below the surface. □ sandy soil (podsol, moisture 6%); o loess (moisture 6%; Δ loess (moisture 12%); x evaporation without soil.

is of course: 'What is the source from which the mercury has evaporated?'

Anderson and Anderson (1972) worked out a computer simulation model for the behaviour of mercury in the ecosystem. The amount of mercury in the atmosphere is, according to Anderson and co-workers, thought to stem from three sources: Hg in oil, coal and ores. The oil production since 1800, multiplied by the mean mercury content of oil, yields the contribution of mercury to the atmosphere caused by the combustion of the oil. For coal the same reasoning can be followed. From the gross mercury production 3% is lost to the air in mixing and melting. Of this production, less the mercury which is conserved by recycling, 37% is discharged into the air as metallic or inorganic compounds and 6% as organic compounds. As the conversion of organic mercury into inorganic mercury occurs rather easily (Spangler et al, 1972), we did, contrary to Anderson, not separate the various mercury compounds.

The equation for the rate with which this man-made mercury is brought into the atmosphere, $Hg_{air,t}$, is then $Hg_{air,t} = C_t \cdot CFR + O_t \cdot OFR + 0.03 \cdot P_t + 0.43 \cdot 0.97 \cdot P_t (1 - RCF)$ in which

C_t = coal production as f(time) CFR = Hg conc. in coal
O_t = oil production as f(time) OFR = Hg conc. in oil
P_t = Hg production as f(time) RCF = Hg recycling fraction

In addition to Anderson's data we have the natural mercury background in rain, called Hg_{degas}, calculated from the mercury content of the 2000 year old ice sheet at Greenland (Weiss et al, 1971).

Considering a residence time of 0.25 year and no evaporation from the soil, we have calculated the Hg drain rate from $Hg_{drain,\ t} = a \cdot Hg_{air,t} + b.Hg_{degas}$ in which a and b are conversion factors to obtain the proper units (assuming a uniform distribution of the mercury over the earth's surface).

Calculations carried out along these lines result in a mercury contamination of about 0.01 ppm Hg above the "natural background" of 0.01 ppm Hg. In fact this situation is only found in a soil near Bari, indicating that extra contamination plays no significant role in that area.

It means that in the other areas the contribution of the man-made mercury, is higher than expected with a uniform distribution; so a regional multiplication factor (R) must be introduced.

$Hg_{drain,t} = a \cdot R \cdot Hg_{air,t} + b \cdot H_{degas}$

For the soils near Ahrweiler and Amiens, the regional multiplication factor equals about 5, for the Netherlands soils near Alkmaar, Hilversum and Amersfoort and the soils near Ispra and Hannover the regional mutiplication factor R equals 10.

Indeed both the population density and industrial activity in the western part of the Netherlands are very high so that a "more than average" contamination with man-made mercury may be expected. The same can be said for Ispra which is situated close to the industrial area round Milan. Also for Hannover, situated about 200 km north-east of the Ruhrgebiet a high value for R is understandable. The areas near Amiens and Ahrweiler have a less dense population and less industrial activity. This is reflected by a lower mercury level.

According to this model the total drain rate for the Netherlands territories was 3 ton/year in 1900, 5 ton/year in 1952 and 9 ton/year in 1972. Compared to a mercury consumption of 100 ton/year these figures are relatively low (ref. 6). It may indicate that the mentioned total drain rates only represent net fluxes and that, in fact, both the drain rate and evaporation must be raised by the same amount.

The contribution of the mercury applied in agriculture, which is only 2 ton/year, is, in relation with these figures, very small. It is difficult to compare the atmospheric contamination with the contamination of the Rhine. It is well known that this river carries annually about 70 tons of mercury to The Netherlands, the greater part, however, eventually reaches the North Sea.

We realize that both the model and the input data which are used, are only approximations. For the time being they must be seen as a means to test concepts and to deepen one's understanding of the problem. Thus we think that the trend expressed by the just mentioned regional multiplication factor, R, is right. But not too much attention should be given to the evaluated values of R. The data for the net flux can hardly be wrong because they are in agreement with the observed mercury contamination of the soil.

CONCLUSION

Both the column experiments and measurements in the field indicate a strong adsorption of mercury and its compounds in the top layer of soil. Leaching is hardly involved.

Evaporation takes place; for dimenthyl mercury the evaporation rate is high, for the other compounds probably

low. There is no indication that organic mercury applied in agriculture behaves differently in soils than mercury stemming from atmospheric fallout, so special concern for the use of fungicides in tulip fields seems not necessary. In dunes irrigated with mercury polluted water, mercury is held back in the top layer of the soil. Although this proces reduces the mercury contamination of the water very strongly, it does not exclude the possibility that solved mercury compounds remain in solution.

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