COMMISSION OF THE EUROPEAN COMMUNITIES

environment and quality of life

On the future average mercury content of air, soil and river sediments in the EEC and in the world's oceans



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INTRODUCTION

Metallic mercury and mercury compounds occur naturally in the environment, but under normal conditions they are present only at low levels. In addition to the natural sources, to which belong volcanic activity, the heat streams of geysers and the degassing process of the earth's crust (metal deposits), increasing amounts are being released by the use of mercury and the consumption of mercury-containing primary materials. With the increased use of mercury and the increased consumption of coal and oil, the flow of mercury through the environment has increased and has often resulted, at least locally, in increased levels of mercury. The problem of mercury pollution has therefore given rise to growing concern.

In the EEC at present about 1700 tons of mercury per year are being discharged into the environment by the use of mercury, while a further quantity of about 600 tons per year is released into the environment by the consumption of coal, oil, natural gas and the refinement of metals.

Since the slight decrease in demand for mercury in the EEC, initiated after 1969, will have to continue in the future, the Environment and Consumers Protection Service of the Commission in Brussels has requested that a study be devoted to the impact of reductions in certain mercury consumption sectors on the future mercury concentration levels in the EEC.

For this study use has been made of a time-dependent computer model set up earlier (EUR 5683), which simulates the pathway of mercury through the environment, giving average mercury concentration levels in the atmosphere, soil and river sediments in the EEC as a function of time.

In addition, in an extended model, the impact of the total mercury released throughout the world on the average mercury content of ocean water as a function of time, has been investigated. . х

<u>PART I</u> - MODEL FOR THE SIMULATION OF THE PATHWAY OF MERCURY THROUGH THE ENVIRONMENT



1. The Pathway of Mercury through the Environment

Natural mercury and mercury emitted from human activity enter the biosphere via air, soil and surface waters, in which it then moves in a rather complex manner.

From the soil it can evaporate into the air or be transported by erosion to rivers and lakes.

Mercury in the air can precipitate once more by dry fallout or be washed out by rainfall, or, during the time that it remains in the air, be transported rapidly elsewhere. When mercury is washed out by rain or snowstorms and deposited onto the soil it can evaporate again or be carried by erosion via the rivers to the ocean, where it is deposited on the bottom of the ocean. Another path by which mercury reaches this sink is in rain falling over the oceans.

Figs. 1 and 2 schematically represent the pathway of mercury through the environment, respectively in the natural state and in that in which mercury is also emitted into the environment from human activity.

For the European region under consideration, the mercury in air in the natural model (Fig. 1) can be assumed to have originated mainly from natural sources outside the EEC, such as volcanos and Icelandic geysers, represented by AMI. In the industrial model (Fig. 2), however, the discharge of man-made mercury into the air for the EEC is relatively higher than that of the surrounding regions, so that it may be assumed to be partially transported outside the considered region (ocean and surrounding countries), represented by AMO.

1.1 Basic Considerations

- For the simulation of the pathway of mercury no distinctions will be made for the various mercury compounds involved, particularly where the conversion of organic mercury compounds into inorganic mercury, or the reverse, can occur rather easily /17,35,367. Therefore, the time-constants to be used in the simulation model, which determine the transport of mercury through the different compartments of the environment, have to be seen as averages for all the mercury and mercury compounds involved.

The mercury concentration levels in air, soil and river sediments will be assumed to be constant with time in the natural state (considered to be before the year 1750), since the system existed for thousands of years, much longer than the time-constants which determine the flow of mercury through the different compartments of the biosphere. Use will therefore be made of this equilibrium situation in the natural model to determine these time-constants for the use in the industrial model. The following mercury concentration levels in the natural state will be assumed:

			2	
in	air	2	ng/cm ³	[2]
in	soil	50	ppb	[5,6,22]
in	river sediments	100	ppb	[6]
in	river water *	0.05	ppb	[7, 38, 39]
in	rain water	0.06	ppb	[4,5]

- The contribution of the weathering of mercury-containing rocks to the mercury emissions into air is assumed to be negligible for the considered region in Europe.

Airborne mercury is considered to be homogeneously distributed and concentrated up to an altitude of 5000 m, whereas mercury precipitation from the air by rainfall or snow is considered to occur every 12 days 28, 4, 23, 28 which assumes that all the mercury present in the air up to an average altitude of 500 m will be completely washed out.

Mercury precipitated from the air onto the soil is considered to be firmly retained mainly in the upper 10 cm of the soil layer [9], whereas mercury evaporation from the soil into the air (for a normal background concentration in soil) is considered to occur mainly from the upper 10 cm of the soil layer [17]. Since the soil layer involved in these processes may be somewhat more or less than 10 cm, depending on the type of soil, the type of management of all and the automatical sector.

mercury compounds involved and most of all on the extent to which the soil might have been disturbed by man, and where experimental soil concentrations given in the literature are mostly expressed as averages over a depth of 20 cm, the results for the average mercury content will be expressed as an average over a 20 cm deep soil layer, taking into account the natural background level over the second 10 cm layer.

- The mercury concentration of the streams in the natural state will be considered to be maintained by the erosion of mercury-containing soil and rocks, which, together with the mercury precipitated into the st reams and lakes by rainfall, is considered to form the soil to streams run-off.

Since most of the mercury in the streams is absorbed by sediment particles and a dynamic equilibrium will exist between the mercury concentration in river water and that in sediments 10/, only the mercury concentration in sediments will be considered in both models.

^{*} including both dissolved Hg and Hg absorbed by suspended particles.

The mercury in the sediments up to an average depth of 10 cm / 36 / will be considered to be involved in the pathway of the mercury to the ocean. The part below 10 cm is regarded as a sink. Mercury evaporation from streams and lakes is considered to be negligible.

As the mercury precipitation into the streams and lakes by rain fall is considered to be included in the mercury run-off "soil-streams", consequently the mercury run off "streams-ocean" equals in the natural state to the mercury run-off soil-streams.

2. The Natural Model

2.1 Numerical Assumptions

Considered Region (EEC):

Soil surface = $1.5 \cdot 10^6 \text{ km}^2 / 1/$ Surface of rivers and lakes (5% of soil surface) = $7.5 \cdot 10^4 \text{ km}^2 / 1/$

Air

Altitude of mercury-containing air Average mercury concentration,	CAM	-	5000 m 2 ng/m ³	[2]
Mass of mercury in air,	\underline{AM}	=	15 tons	
Average altitude over which Hg is conside to be washed out by rain/snow fall	red	Ξ	500 m	
Rain				
Average precipitation rate		Ξ	1000 mm/m^2	year [3,28]
Average mercury content of rain/snow		=	0.06 ppb	[4, 5]
Average mercury precipitation rate \underline{P}	RSM	=	90 tons/year	
Soil				
Thickness of the soil layer		=	10 cm	[9,17]
Soil density		Ξ	1.5 kg/dm ³	
Average mercury content of soil,	<u>CSM</u>	=	50 ppb	[5,6,22]
Mass of mercury in soil,	<u>SM</u>	=	11,250 tons	
Mud				
Thickness of the mud-layer		=	10 cm	
Mud density		=	1 kg/dm^3	
Average mercury content of mud,	CMM	=	100 ppb	[6,10]
Mass of mercury in mud,	MM	=	750 tons	-

River water

River water to ocean run-off	$= 3.8 \cdot 10^{11}$	tons/year /4.267
Average mercury content of river water Mercury run-off streams to ocean BSOM	= 0.05 ppb = 19 tons/ve	[7,39]

2.2 The Mass-Transport of Mercury between the Different Compartments

Taking the steady state condition for the pathway of mercury up to the year 1750 as the condition of dynamic equilibrium for each of the compartments involved, the mass-transports of mercury can be calculated from a mass-balance, which turns out to be (see also Fig. 1):

 $\begin{array}{rcl} \underline{Air} & AMI & = 19 \ tons/year & \underline{Soil} & PRSM & = 90 \ tons/year & EVM & = 71 \ tons/year & RSSM & = 19 \ tons/year & RSSM & = 19 \ tons/year & \underline{Mud} & RSSM & = 19 \ tons/year & RSOM & =$

2.3 The Time-Constants which Determine the Mercury Transport from one Compartment to Another

The respective time-constants can be defined as:

 $T = \frac{A}{dA/dt}$, in which A = the total amount of mercury in the compartment concerned, dA/dt = the rate at which mercury is added or removed from the compartment in question, T = the time-constant for the mercury input or output, so that, with $PT = \frac{AM}{PRSM}$, $EMT = \frac{AM}{EVM}$, $RSSMT = \frac{SM}{RSSM}$, $RSOMT = \frac{MM}{RSOM}$: time-constant Hg precipitation air-soil PT = 0.166 years time-constant Hg evaporation soil-air EMT = 158.511 time-constant Hg run-off soil-streams RSSMT = 59211 time-constant Hg run-off streams-ocean RSOMT= 39.5

The time-constants will be used in the industrial model.

3. The Industrial Model

The main mercury emissions from human activity can be divided into:

- 1) the release of mercury due to the use of mercury,
- 2) the release of mercury due to the use of primary materials.
- 3.1 Mercury Emissions due to the Use of Mercury
- 3.1.1 Loss of Mercury in Mining and Smelting

Fig. 3 represents the estimates for the yearly mercury production, PM /8/, in Italy, the only mercury producing country of the EEC. The estimates for production after 1975 have been adapted to the estimates for the future mercury demand in the EEC (see § 3.1.2), considering the hypothetical case that the future demand of mercury in the EEC will be approximately covered by mercury production in Italy. Since about 3% of the gross mercury production can be considered as being lost into the air during mining and smelting /10/, the mercury emissions into air amount to:

0.03 ***** PM tons/year.

3.1.2 The Consumption of Mercury

The estimates for the use of mercury, UM, in the EEC as a function of time is represented by Fig. 3 and are based on estimates given in the literature [8, 9]. For the model it is presumed that consumption will remain constant [10] from the year 2000 onwards.

Since for the period up to the year 1970 a certain percentage of the used mercury, UM, may be considered to have been recycled estimated at RCF = 0.18 / 10, 12/, from a technical viewpoint this recycling percentage will be applied for the whole time-interval in question.

Consequently, the net consumption of mercury, CM, amounts to:

CM = UM (1 - RCF) tons/year.

This amount of mercury finally ends up in the environment, partially in air, partially in soil and the rest in sediments. Tables I to III represent estimates for the mercury discharge into air, soil and sediments due to the mercury consumption for the years 1972, 1980 and 1990, giving the discharge percentages in air, soil and sediments of the total mercury consumption, being respectively PPAM, PPSM and PPMM, which for simplicity will be represented by one average value and assumed to be representative for the whole time-interval in question. Consequently, the yearly mercury discharge will be respectively:

Air	PPAM 🛣 CM	tons/year
Soil	PPSM * CM	tons/year
Sediments	PPMM ≵CM	tons/year.

The discharge percentages turn out to be approximately:

$$PPAM = 0.60 PPSM = 0.11 PPMM = 0.29.$$

The total release of mercury due to the use of mercury thus amounts to:

Air	PPAM * CM + 0.03 * PM	tons/year
Soil	PPSM \star CM	tons/year
Sediments	PPMM * CM	tons/year.







Figure 4 : Mercury emissions from metal refineries (PCZ) and the consumption of natural gas (G) as a function of time



Figure 5 : COAL (C) and OIL (O) Consumption as a function of time

-15-

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Common]	
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throughout	
charge	
Disc	
rcury	

	F	able I	- 1972		Т	able II	- 1980		•	Table]	п - 19	06
DISSIPATIVE, FUNCTIONAL	Air	Soil [*]	Sedi- ments	Total	Air	Soil [‡]	Sedi- ments	Total	Air	Soil [‡]	Sedi- ments	Total
Electro-technical, including dry batteries ^o	330	100	1	430	235	75	1	310	180 -	60	8	240
Dentistry	60	9	54	120	67	2	61	135	20	~	63	140
Paint Industry	100	ı	0	100	30	I	1	30	10	1	I	10
Paper Industry	2	ı	9	8	1	ı	ı	ſ	ł	١	ı	I
Agriculture	3.5	63	1	66.5	2	38	•	40	1	19	I	20
Pha rmaceutics	1	ŝ	37	40	1	1	19	20	1	ł	10	10
Chlor-alkali Industry (mercury in caustic soda, Cl ₂ , H ₂)	35	6	56	100 /8/	18	4	32	54 [8]	13	4	21	38 <i>[</i> 8]
Laboratory and Chemical Products	1	1	1	1	30	1	40	20	20	I	20	40
Present and Future Applications	1	•	1	1	43	2	20	70	43	2	20	70
Others	10	ŝ	17	30	2	1	9	ß	ı	ł	1	1
NON-DISSIPATIVE, FUNCTIONAL						<u> </u>						
Chlor-alkali Industry	482	1	238	720 [8]	265	1	131	396 <i>[</i> 8]	189	ſ	93	282 [8]
Catalystics	•	1	60	60	1	t	40	40	1	1	30	30
	1022.5	184	468	1674.5	692	132	346	1170	526	26	257	880
TOTAL tons/year	(61%)	(11%)	(28%)	(100%)	(26%)	(11%)	(30%)	(100%)	(%09)	(11%)	(%62)	(100%)

-16-

★ 20 cm top soil

o mercury-containing solid waste dumped onto waste deposits, is considered to contribute to emissions of mercury into the air, because these emissions are often enhanced by burning or self-ignition of the waste deposits.

- 3.2 Mercury Emissions due to the Use of Mercury-Containing Primary Materials
- 3.2.1 Mercury Emissions due to the Refinement of Lead, Copper, Zinc etc.

Estimates for the annual emission of mercury due to the refinement of lead, copper, zinc, etc., PCZ, considered to be completely released into the air and represented by Fig. 4, are based both on estimates for the EEC for 1970 and on the assumption that the use of the metals in question followed the same pattern of increase as mercury.

From 1975 onwards a decrease in mercury emission is expected [8] due to recovery and in the model the emission level reached by the year 2000 will be taken as remaining subsequently constant [10].

3.2.2 The Consumption of Coal, Oil and Natural Gas

Figs. 4 and 5 represent the estimates for the consumption of coal (including lignite), C, crude oil, O, and natural gas, G, in the EEC.

For the period from 1975 to the year 2000 consumption is considered to increase yearly by 4%, whereas in the model from 2000 onwards consumption is considered as remaining constant /10/. The mercury content of hard coal (including lignite), crude oil and natural gas are estimated respectively to be:

CFR = 0.30 ppm
$$(5, 13, 14)$$

OFR = 0.02 ppm $(13, 15)$
GFR = 5 µg/Nm³ (13) .

If we assume the mercury released by the consumption of coal, oil and natural gas to be completely emitted into air, the yearly amount of mercury released thus amounts to:

<u>Air</u> CFR $\mathbf{*}$ CFR $\mathbf{*}$ OFR $\mathbf{*}$ O + GFR $\mathbf{*}$ G tons/year.

3.3 The Total Mercury Discharge from Human Activity

The total mercury discharge in air, soil and sediments, obtained from the foregoing, amounts respectively to (see Fig. 2):

Air : IAM = PPAM * UM *(1-RCF)+0.03 * PM + PCZ + CFR * C + OFR * O + GFR * G Soil : ISM = PPSM * UM * (1-RCF) Sediments : IMM = PPMM * UM * (1-RCF).

For a survey of the estimates of the total mercury discharge in the EEC in the years 1972, 1980 and 1990, see the Appendix.

3.4 The Time-Constant TO

The time-constant still to be defined is TO, being the time-constant for outgoing mercury AMO in the industrial model and as previously explained, inherent to the strong increase of the mercury release from human activity. TO, being in the order of a few days /16/, will be evaluated by satisfying the condition that the average mercury content of rain water approximately increased by a factor of 1.5 /5, 24, 21/ for the period 1930 - 1935, and 2.0 /21, 24/ for 1970 - 1975, with respect to that in the year 1750. It is found that TO should be in the order of 0.011 year.

3.5 The Computer-Program - Industrial Model

```
VWE KL=VWEN
WF WG
A.M. AMA, J+OT*(VWE.JK+EVM.JK-PRSH.JK+IAH.JK+AMI.JK-AMO.JK)
A.M. AMA
T.M. KL=PPAN.K*CO.K+CFR*C.K+OFR*O.K+PCZ.K+0.03*PM.K+G.K*GFR
OFF.0.23
GFR.0.23
GFR.
LCD
LCD
LCD
LCD
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<u>PART II</u> - VALIDITY AND CHARACTERISTICS OF THE MODEL

1. <u>The Mercury Concentration in Air</u>, Soil and Sediments in the EEC as a Function of Time (Time-Interval 1750 - 1975)

The results of the average mercury concentration levels in air, soil and sediments for the time-interval 1750 - 1975 given by the computer model are represented by Fig. 6.

According to these results the average mercury concentrations would have been increased in the EEC as follows:

		1750	1930-1935	<u>1970-1975</u>
Air	in ng/m^3	2	3	4
Soil	in ppb	50	62	73
Sediments	in ppb	100	780	1470

a) Air and Rainwater

Taking into consideration the assumption that mercury in the air is completely washed out every 12 days (see § 1.1), the average mercury concentration would have varied between zero, just after a rain fall, and a value just before rain fall of:

 $\frac{1750}{4} \frac{1930-1935}{6} \frac{1970-1975}{8}$ Air in ng/m³ (just before rainfall)

Consequently, the average mercury content of rain water would have been increased, as can be calculated, as follows:

	1750	<u>1930-1935</u>	<u>1970-1975</u>
Rainwater in ppb	0.06	0.09	0.12

In addition, where the mercury content of air over oceans, averaging approximately 0.5 ng/m³, may influence the air over coastal regions by sea winds (30), the following two extremities in the distribution pattern of mercury in the air and rain water can be deduced from the computer results:

		1750	<u>1930-1935</u>	<u>1970-1975</u>
Air	in ng/m^3			
	max.	8	12	15
	average	2	3	4
	min.	0	0	0



Fig. 6 : The mercury concentration levels as a function of time Hg in air = $A(ng/m^3)$, soil =S(ppb), mud = M(ppb)

		1750	1930-1935	1970-1975
Rain water	in ppb			
	max.	0.105	0.174	0.225
	average	0.06	0.09	0.12
	min.	0	0	0

It is rather difficult to find comparable data for the average mercury concentration in air in literature, due to the inhomogeneous distribution of mercury in the atmosphere, attributed to a dependence upon the type of region (coastal, rural or industrial), wind direction, wind speed, temperature, pressure, the frequency of precipitation, daytime, season and altitude. Moreover, mercury concentrations in the air can be reduced to zero level by rain storms $\sqrt{32}$.

The only information available on mercury concentration levels in the air and rain water in the past are those published by Stock and Cucuel indicating an average mercury concentration of 8 ng/m³ for "unpolluted air", and of 0.2 ppb for rainwater, measured in Germany in 1934 [5, 6].

Measurements of the mercury content of the Greenland Ice-cap /4/show an increase of the mercury concentration in snow from 0.06 ppb in 1750 up to an average of 0.125 ppb (fluctuation between 0.087 and 0.230 ppb, due to inhomogeneous distribution in the air) for the present /4,21,31/. In addition, the mercury content of rain water in Sweden is reported (1965) as averaging 0.12 ppb /24/.

The mercury concentrations in air over the continents at present are generally falling between less than 1 and 10 ng/m³ (31, 32). Higher values of up to 14-16 ng/m³ (6, 23), are found over industrial areas, lower ones, 0.6-0.7 ng/m³, over oceans (30). Excluding the influences of strong natural or industrial local pollution which cannot be predicted by the computer model, the results for the mercury concentrations in air and rain water for the time-interval 1750-1975 as demonstrated before, conform to the experimental data given in the literature, especially where a variation of the precipitation frequency may influence the deduced maximum values for the mercury content both in the atmosphere and rain water.

b) Soil

The average values for the mercury content in soil for the whole timeinterval in question, as given by the computer model, agree rather well with the experimental averages given in the literature.

Amongst the scarce information available, concerning the mercury content of soil in the past, belong that published by Stock and Cucuel for certain regions in Germany in 1934 $\lfloor 5 \rfloor$:

<u>Soil</u>	<u>Average Hg</u> content in ppb	Depth	Location
forest soils	30 - 81	not indicat	ed
11 11	100 - 290	tt tt	(near Karlsruhe)
arable soils	30 - 70	tt it	•
clay	30	tt II	
tt -	30	it tt	
sand	29	tt it	

Amongst the more recent experimental values for the average mercury concentration of undisturbed soils in the EEC, measured over a depth of 20 cm, some have been published by Frissel, et al., in 1974 /17/, i.e.:

Location	Soil	Hg-content	Depth
		in ppb	<u>in cm</u>
Alkmaar, NL	clay	90	0 - 20
Hilversum, NL	sand	90	0 - 20
Amersfoort, NL	tt	90	0 - 20
Schoonebeek, NL	peat	70	0 - 20
Leiden, NL	dune	30	0 - 20
Ahrweiler, FRG	clay	50	0 - 20
Hanover, FRG	sand	60	0 - 20
Amiens, F	calc. soil	40	0 - 20
Amiens, F	loess	50	0 - 20
Bari, I	red soil	20	0 - 20
Ispra, I	clay	100	0 - 20
Hillegom, NL *	dune	160	0 - 20

* Bulb-growing area, where mercury has been applied as a fungicide for 50 years with an intensified use over the last 20 years.

Frissel et al. indicated an average value of 70 ppb /17 for the above mentioned results, which conforms with the average computer result for 1972.

In addition, Martin (1963) indicated the mercury content of some English soils as being between 10 and 60 ppb 25, whereas Anderson reported in 1967 a variation in the mercury content for Swedish soils of between 10 and 90 ppb 6, with an average of 70 ppb 18.

c) Sediments

Comparable values for the average mercury content in the past and present are published by J. Bouquiaux /19/.

Location:	Windermere	Lake, GB (ru	ral zone) [33]	
Period:	520 - 1400	1400 - 1870	1870 - 1915	1915 - 1972
Average Hg-content in ppb :	122	286	608	1026

The results given by the computer model average for approximately the same periods:

Period:	up to 1750	1750 - 1870	1870 - 1915	1915 - 1972
Average				
Hg-content				
in ppb:	100	260	560	1080

In addition, a comparison can be made between the average mercury concentration of sediments in the EEC for the period 1969-1972, given by the computer model as being 1470 ppb, and the results of measurements on several samples of river and lake sediments in the EEC for the same period as listed in the following table $\sqrt{19}$:

Period:	1969 - 1972	<u>Average Hg-</u>
	Location	content in ppb
Belgium	Meuse - North of Liège	270
	" - South of Liège	1230
	Sambre	2560
	Schelde	390
	Vesdre	1200
	Yser	50
	Other tributaries into the North Sea	300
Denmark	River Grinstedt - North of Grinstedt	26
	" - South of Grinstedt	3620
<u>Great Brit.</u>	Lake Windermere (rural zone)	1026
FRG	Danube - Isar	120
	- Regen	40
	Elbe	7600
	Ems	4400
	Rhine – Bodensee (Eastern part)	300
	- Main	5000
	- Neckar	1100
	Weser	2 300
Luxemburg	Esch-sur-Sûre	20
		1080

Location	<u>Average Hg-</u> content in ppb
The Netherlands Rhine - Biesbosch	23300
- Noorderleeg	2700
- Julianapolder	1300
- Uithuizerwadpolder	1600

From the computer model it is also possible to deduce the maximum possible mercury concentration in sediments in 1972, considering the average computer result for 1972, which is 1420 ppb, to be a logarithmic average of the two extremes, for maximum polluted and unpolluted sediments. Since the average value for unpolluted sediments in the computer model amounts to 100 ppb, which is the initial average value for 1750, the maximum mercury concentration in sediments for 1972 turns out to be 21, 500 ppb, which is of the same order of magnitude as that found for the Biesbosch (Rhine) in 1972.

- 2. <u>Some Aspects of Industrial Mercury Emissions on Environmental</u> Mercury Pollution, Demonstrated by the Computer Model
- 2.1 The Impact of Mercury Consumption on the Environmental Mercury Pollution

The impact of industrial mercury consumption on the mercury pollution of air, soil and sediments is demonstrated by comparing the results for a zero mercury consumption after 1750, represented in Fig. 7, with those including the consumption of mercury given in Fig. 6. As can be concluded, the contribution of the consumption of mercury to the mercury pollution of the atmosphere is approximately equal to that of mercurycontaining primary materials.

As the mercury release due to the consumption of mercury-containing primary materials is considered to end completely in air, a zero consumption of mercury after 1750 would theoretically have contributed to unpolluted rivers and lake sediments.

2.2 The Impact of Direct Industrial Mercury Discharge into Streams

The impact of direct industrial mercury discharge into streams as demonstrated in Fig. 6, is shown up by a comparison with a zero industrial mercury discharge into streams for the same time interval (1750 - 1975)represented in Fig. 8.

Fig. 8 demonstrates that the mercury pollution of river sediments would have remained approximately equal to the initial level in the natural state, despite the increase of the mercury concentration in soil for the same time-interval. This is in agreement with the results of the experiments performed by Frissel et al., which showed that the mercury content in soil due to direct industrial mercury discharge, does not influence measurably the mercury content of the ground water /17/. The negligible increase of the mercury content of river sediments shown by the computer results, may be partially attributed to the increase of the mercury concentration of eroded soils and partially to that of rain water ending up directly or indirectly in streams.

2.3 The Impact of Direct Mercury Discharge on the Mercury Content of Soil (Including the Application of Mercury in Agriculture)

Fig. 9 shows the influence of the increased mercury concentration in air on the mercury content of the top soil as given by the computer results for a zero direct mercury discharge onto the soil. On the other hand, by comparing the results given in Fig. 9 with those of Fig. 6, it is possible to see what the approximate contribution of the direct mercury discharge onto the soil must have been to the ultimate mercury content of soil during the time-interval 1750-1975.



Fig. 7 : Zero mercury consumption after 1750. Hg in air = A(ng/m³), soil = S(ppb), mud = M(ppb)

407 200. 1500. 1000. . دون د CAMEA.CSMES.CMMEM 1 1 ••• 1650. 1900-1800. 1550.1 1 150.

Fig. 8 : Zero industrial Hg discharge into streams after 1750. Hg in air = A(ng/m³), soil = S(ppb), mud = M(ppb)



Fig. 9 : Zero direct mercury discharge onto soil after 1750. Hg in air = A(ng/m³), soil = S(ppb), mud = M(ppb)

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PART III - THE FUTURE AVERAGE MERCURY CONCENTRATION LEVELS IN AIR, SOIL, RIVER SEDIMENTS AND OCEAN WATER

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1. The Future Average Mercury Concentration Levels in Air, Soil and Sediments in the EEC

On the basis of the estimates for the future demand/consumption of mercury in the EEC for the years between 1980 and 2000 $\lfloor 9 \rfloor$, and on the assumption in the model of a constant consumption after that date $\lfloor 10 \rfloor$, the future average concentration levels would change as shown in Fig. 10.

- Air: A decrease may be expected to a level, approximately equal to that of 1930 (3 ng/m³) by the year 2000, and due to the assumption of a constant consumption of mercury and mercury-containing primary materials after that year, this level will remain constant from then onwards.
- Soil: The average mercury content will increase only negligibly, due to a reduced direct mercury discharge after 1980 (see Tables I to III), and as indicated by the computer results, as follows:

1975	2000	2100
74 ppb	76 ppb	80 ppb

Sediments: The strong increase of the average mercury content of river sediments as shown by Fig. 10, will be arrested in 1980 due to an expected decrease of direct mercury discharge into streams, whereas only after 1990, due to the long mercury run-off streams-ocean, (time-constant is 40 years), a decrease can be expected of almost 25% by the year 2100.

2. <u>The Impact of Reduced Mercury Emissions from the Chlor-Alkali</u> <u>Industry after 1980</u>

Upon request 20/, an investigation has been made as to what the impact of a stronger limitation of mercury emissions from the chlor-alkali industry would be on the future mercury pollution in the EEC. Table IV-A shows the figures for the mercury emissions per ton of chlor produced in 1972 and that expected to be realized in 1980 and 1990 28/, whereas Table IV-B indicates the corresponding figures for a stronger limitation of mercury emissions in this sector 20/.

Stronger limitation may reduce the total mercury consumption in the EEC to 810 tons/year in 1980 and to 610 tons/year from 1990 onwards. Consequently, the percentages for the mercury discharge in air, soil and sediments will change after 1980 and are estimated to average 59%, 15% and 26% respectively of the total mercury discharge (see Tables V and VI).

Table IV

Year	Production of chlor by mercury in tons x 10 ⁶	A Total emission gr Hg/ton Chlor		Emi gr H _g	B ission g/ton	Chlor		
			H ₂	NaOH	H ₂ O	Air	Tot	al
1972	5, 13	140	•	•	1	ł	1	
1980	7.00	55-70	1.5	1.2	2	æ	~	~
1990	6.50	45-60	1.5	0.6	1	4	2	~

<u>Mercury Discharge in the Common Market in metric tons/</u>	уеаг	Table	<u>v</u> - 1980			Table	<u> 1990 - 17</u>		
	Air	Soil	Sediments	Total	Air	Soil	Sediments	Total	
DISSIPATIVE, FUNCTIONAL									
flectro-technical, including dry batteries	235	75	,	310	180	60	1	240	3
Dentist ry	67	2	61	135	20	7	63	140	5-
Paint Industry	30	. 4	ı	30	10	1	ı	10	
Agriculture	8	38	ì	40	Π	19	ı	20	
Pha rma ceutics	,	н	19	20	1	•	10	10	
Chlor-alkali Industry (mercury in NaOH, Cl ₂ , H ₂)	9	2	11	19	S		80	14	· · · ·
Laboratory and Chemical Products	30	ł	40	20	20	ł	20	40	
Present and Future Applications	43	7	20	70	43	2	20	20	
Others	2	t	ñ	ŝ	ı	ł	ı	•	1
VON-DISSIPATIVE, FUNCTIONAL						<u> </u>			
Chlor-alkali Industry	56	1	14	70	26	I	Q	32	
Catalystics	1	E	40	40	B	P	30	30	
TOTAL tons/year	471	130	208	809	355	94	157	606	
	(58%)	(16%)	(26%)	(100%)	(26%)	(15%)	(26%)	(% 001)	



Fig. 10 : The mercury concentration levels as a function of time Hg in air = $A(ng/m^3)$, soil = S(ppb), mud = M(ppb)

The effect of such limitation on the future average mercury concentration levels in air, soil and sediments is demonstrated by Fig. 11. As can be seen from this figure, such a reduction will contribute to a much more rapid reduction of the average mercury content level in river sediments of approximately 40%, to be reached in the year 2100, whereas at the same time the average concentrations of mercury in air and soil will remain practically unmodified. Therefore, such a measure in itself, must be seen as a means to reduce, in an accelerated way, the average mercury concentration level in river sediments, or to reduce local pollution of air, soil and river sediments, which, however, cannot be predicted by the model.

Almost the same impact on the future average mercury concentration of river sediments could be obtained if the mercury release from the chloralkali industry were to be limited to 30 gr Hg/ton of chlor produced after 1980 or even only after 1990 as shown by the results in Fig. 11 a and 11 b.

Any stronger limitation appears to have proportionally less effect on the average mercury concentration of river sediments, due to the mercury wastes discharged into streams by other users.



Fig. 11 : The impact of reduced mercury emissions from the chlor-alkali industry after 1980. Hg in air = $A(ng/m^3)$, soil = S(ppb), mud = M(ppb)



Fig. 11a : The impact of reduced mercury emissions from the chlor-alkali industry limited to 30 gr Hg/ton of chlor produced after 1980. Hg in air = $A(ng/m^3)$, soil = S(ppb), mud = M(ppb).



Fig. 11 b : The impact of reduced mercury emissions from the chlor-alkali industry limited to 30 gr Hg/ton of chlor produced after 1990. Hg in air = $A/ng/m^3$, soil = S(ppb), mud = M(ppb)

3. The Average Mercury Concentration of the World's Oceans

3.1 Model for the Pathway of Mercury on a Global Scale

For the evaluation of the future average mercury content of the oceans, the computer model has been extended with an additional model for the pathway of mercury through the environment for non-industrialized regions and with a compartment representing the oceans, into which all the mercury is discharged by the streams from industrialized and non-industrialized regions in the world. In addition, in the model, the mercury concentration in the air over non-industrialized regions and over the oceans is influenced by the outgoing mercury from the air over industrialized regions, as previously assumed (see Part I - 1.).

The following simplifications will be taken into consideration. The total industrialized regions in the world will be considered as being concentrated in a unit over an area of $9 \cdot 10^{12} \text{ m}^2$, which is 6 times that of the EEC ($1.5 \cdot 10^{12} \text{ m}^2$), since the world's consumption of mercury and mercury-containing primary materials may be estimated to be 6 times that of the EEC. Given the pattern of the use and discharge of mercury in the EEC representative for the industrial regions in the world, the total yearly mercury transport out of the industrialized regions by air and via streams, represented by the model for the EEC, will be 6 ***** AMO and 6 ***** RSOM, respectively.

As, of the remaining 94% of the total surface of the continents, considered as being non-industrialized regions $(141 \cdot 10^{12} \text{ m}^2)$, only an area equal to that of the EEC will be represented by the model, the contribution of the outgoing mercury by air from the industrialized regions to the mercury concentration levels of air over the non-industrialized regions and the oceans $(350 \cdot 10^{12} \text{ m}^2)$ will be 0.02 ***** AMO and 4.2 ***** AMO, respectively.

Since the mercury run-off streams-ocean in the non-industrial model is represented by RNSOM, the total yearly run-off from all the non-indus-trialized regions in the world is represented by 94 \ddagger RNSOM.

Mercury evaporation from the ocean into the air will be ignored for the following reasons:

- 1) The lack of information on the evaporation of mercury from surface waters.
- 2) The assumption of a negligible mercury evaporation from the ocean surface waters will only contribute to pessimistic results for the mercury content of ocean water.

3) If mercury evaporation does contribute to the mercury concentration level in the air over the oceans and the continents, for an almost constant average mercury concentration level of ocean water with time, such a contribution may be considered to be included in the natural incoming mercury in air over the continents and oceans, attributed to volcanic activity, geysers, etc.

The natural incoming mercury in the atmosphere on a global scale is estimated to be:

EEC	(total surface $1.5 \cdot 10^{12} \text{ m}^2$)	=	19 tons/year
Continents Oceans	(total surface $150 \cdot 10^{12} \text{ m}^2$) (total surface $350 \cdot 10^{12} \text{ m}^2$)	= =	1900 tons/year 600 tons/year
	Total natural emissions	=	2500 tons/year /10/

Schematically, the pathway of environmental mercury on a global scale is represented by Fig. 12.

3.2 The Ocean as a Compartment in the Extended Model

Apart from direct volcanic activity, the natural and man-made mercury enters the ocean via run-off in river water and river sediments from the continents and by precipitation from the air over oceans (PROM).

Within the ocean mercury may be ingested by micro-organisms and passed up the food chain to larger biota.

The transport of mercury within the ocean is certainly on the increase, due to uptake by fish. Mercury is lost to the ocean bottom, by sedimentation or through fish excreta and by the death of fish (SEOM). The ocean bottom thus acts as a "sink".

3.3 Additional Numerical Assumptions

Air:	The average precipitation rate	over ocean and continents = $1000 \text{ mm/m}^2 \text{ year } / 28 $
	The average mercury content o state (1750)	over oceans in the natural = $\sim 0 \text{ ng/m}^3$.
Ocean:	The average mercury content of state (1750) The time-constant for the sedin ocean bottom	of ocean water in the natural = 0.03 ppb mentation of mercury to the = 16000 years $\sqrt{297}$.



Fig. 12 - The Model of the Pathway of Mercury on a Global Scale

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3.4 The Computer Model of the Pathway of Mercury on a Global Scale

VWEIKL=VWEN VWEN=0 AM_K=AM_J+DT*&VWE_JK+EVM_JK-PRSM_JK#IAN_JK+AMIIJK-AMO_JK) AM_AMB AM_AMB ROLNURODUACUURURURURULNURACUUURULNURURURUCUUATXXAAUUUUAUAT AM, K = AM, J + D T * 4VWE .JK + EVM. JK - PRSM. JK # IAN. JK # AMI I JK - AMU.JK) AM = AMB AME = 15 IAM IKL = PPAM _K + CM. K + CFR * C .K + OFR * O .K + PCZ .K + O I O 3 * RM .K + G .K * GFR CFR = 0.3 GFR = C .02 GFR AN E = 15 IA M I K L = PPAM "K *CM" K + CFR * C * K + OFR * O * K * & CZ * K + O 103 * & M * K + G * K * & FR XXAATXXATXXXAT XXATXLNCRRALNCRCACLRNRRLRNLRN* 0 M .K = 0 M .J + DT * (PROM .JK+6*KSUP .JK+94*KNSUM .JK-SECM .JK) 0 M = 4 + 0 5 E7 SEC M .KL = CM .K/PT RT = 16000 CCM .K = 0 M .K* 1E9/OCEAN OCEAN = 1 . 35E18 ANM .K = ANM .J + DT * (0 .02*AMO .JK+EVNM .JK-PNSM .JK+ANMI .JK) ANM .KL = ANM .K/PT EVNM .KL = ANM .K/PT EVNM .KL = SNM .J + DT * (PNSM .JK-EVNM .JK-RNSSM .JK) RNS SP .KL = SNM .K/RSSMT SNM .SP .KL = SNM .K/RSSMT SNM .K = MNM .J + DT * (RNSSM .JK-RNSOM .JK) RNS CM .KL = MNM .K/RSGMT MNM # MB DIRECTIONS TIME = 1750 CACM = Ef - 7, 1 J/COM = G(-C .6 .0 .2) DT = 0.01 /LENGTH = 2100/PRTPER = 0/PLTPER * 5 NCN - INDUSTRIALIZED REGIONS .+ UCEANS RUN PLOT SPEC

3.5 The Average Mercury Concentration in the Air over the Oceans, and that of Ocean Water, as a Function of Time

Assuming that the consumption of mercury and the emission from Pb, Cu, Zn refineries throughout the world will remain constant with time after 1980, with the exception of the EEC, the average mercury concentration level in the atmosphere over the oceans and that of ocean water as a function of time would be that indicated in Fig. 13.

	1750	<u>1975</u>	2100
Average Hg content in air over oceans in ng/m ³	0	0.6	0.6
Average Hg content of ocean water in ppb	0.03	0.03	0.03

Air over the Oceans

The only experimental data available for the mercury concentration in the atmosphere over the ocean is that measured 20 miles off-shore over the Pacific Ocean by Williston (1968), i.e. 0.6 to 0.7 ng/m³ (32). This conforms with the computer results for that time.

Ocean Water

Due to lack of information on the average mercury concentration level of ocean water in the past, the measured mercury concentration of sea water near Helgoland by Stock and Cucuel (1934) has been used as an initial value in the computer model.

Some recent experimental data for the mercury content of ocean water have been published by Bouquiaux $\sqrt{19}$, and demonstrate considerable variations in local mercury content, as reported by Hammond $\sqrt{21}$:

		Hg cont	ent in pp	b
Location	Period	min.	max.	average
Waddenzee (NL)	1970		0.2	
North Sea, Belgian				
shore line	1971-1972	0.03	0.76	0.15
English Channel	1970-1971			0.018
Irish Sea:				
- Central area	1971-1972		0.025	
- Northern and				
Southern areas	1971-1972	0.025	0.050	
- Some places in				
the Eastern area				
(Morecombe Bay)	1971-1972	0.050	0.400	

ing content in ppb (continued).	Hg c	ontent	in	ppb (continued):
---------------------------------	------	--------	----	-------	-----------	----

Location	Period	min.	max.	average
Open Ocean Surface Water:				
- Eastern Nor Atlantic mid	th			
latitudes	1972	0.067	0.127	0.088
– from West o	f			
Africa, arou	ınd			
Africa, to Ja	apan 1972	<0.0005	0.113	0.036

Evidently, continental coastal waters, particularly near estuaries, may contain relatively higher values, influenced by the mercury run-off streams-ocean.

Nevertheless, the logarithmic average of the two extremes of the above mentioned experimental data also show that it is in the same order as that measured in 1934 /5/, which may confirm that the mercury content of ocean water did not demonstrably increase as a result of human activity.

A more sensitive plot for the evolution of the average mercury concentration of ocean water, as demonstrated in Fig. 14, shows an increase of only 0.8% for the period 1965-1975. This conforms to the estimate given by Hammond (1971), who thought it unlikely that man could have increased the mercury concentration in the sea by as much as 1% [34]. The results also show a further increase of 2% up to the year 2100, based on the considerations for the future consumption of mercury and mercury-containing primary materials in the world, as outlined previously.



Fig. 13 : The mercury concentration levels as a function of time Non-industrialized region: Hg in air = $A(ng/m^3)$, soil = S(ppb), mud = M(ppb)Ocean: Hg in air = $B(ng/m^3)$, ocean water = O(ppb)



Fig. 14 : The mercury concentration levels as a function of time. Hg in air over oceans = $B(ng/m^3)$, Hg in ocean water = O(ppb).

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CONCLUSIONS

From this study the following conclusions can be drawn:

1) The average mercury concentration of air, soil and river sediments in the EEC have increased more rapidly during the past 50 years.

		1750	<u>1930-1935</u>	<u>1970-1975</u>
air	in ng/m ³	2	3	4
soil	in ppb	50	62	73
river sediments	in ppb	100	780	1470

- 2) The increase of the average mercury concentration levels in air and soil is attributable in approximately equal degree to the consumption of mercury and to that of mercury-containing primary materials.
- 3) The increase of the average mercury content of river sediments is mainly attributable to mercury wastes discharged directly into streams.
- 4) According to the model, man-made mercury discharged directly onto soil has not measurably influenced the mercury content of river water by leaching via ground water. This result is apparently confirmed by experiments as well /17/.
- 5) According to future predictions on mercury consumption, the consumption of mercury-containing primary materials and the expected recovery of mercury released from Pb, Cu and Zn refineries [9], the tendency of the average mercury content levels to increase rapidly in the EEC may be considered to be arrested in the near future and replaced by a diminishing one for air and river sediments.

Considering in the model a constant mercury emission level from the year 2000 onwards, the following results are obtained:

		1975	2000	2100
air	in ng/m ³	4	3	3
soil	in ppb	74	76	78
river sediments	in ppb	1470	1460	1200

- 6) The average mercury content of top soil will remain almost constant due to the long mercury run-off soil to streams (time-constant ~ 600 years), additionally hampered by the evaporation of mercury from the soil, which consequently precipitates at a later stage.
- 7) Any stronger limitation of the mercury emissions from the chloralkali industry after 1980 to less than 50 gr Hg/ton of chlor produced will have a negligible effect on the future average mercury content of air and soil, but may accelerate the decrease of the future average mercury content of river sediments.

A mercury release limited to 30 gr Hg/ton of chlor produced after 1980 or even only after 1990 may result in a reduction of the average mercury content of river sediments to approximately 940 ppb to be reached by the year 2100.

A much stronger limitation appears to have proportionally less effect on the average mercury concentration level of river sediments, due to the mercury wastes into streams, discharged by other users. Since the model only gives averages, no results can be obtained on the reductional effect on the local pollution of air, soil and river sediments.

8) The average mercury content of ocean water does not appear to have increased significantly with time as a result of human activity. Regarding the assumed initial mercury content of ocean water of 0.03 ppb in 1750, this level should have been increased by only 0.8% by the present time. Considering in the model an almost constant total man-made mercury emission in the world often the ware 10% of approximately 12,500.

emission in the world after the year 1980, of approximately 12,500 ton/year, the average mercury content of ocean water will have increased with respect to the initial value for the year 1750 by about 2% by the year 2100.

It should be stressed that the computer model only gives averages, so that no local pollution effects for the future mercury content of air, soil, river sediments and sea water can be predicted. In fact, the assumed initial values for the average natural mercury content levels for the year 1750 are also only averages, and in fact, locally, the concentrations may have been many times higher as a result of the degassing process of the earth's crust (mercury in soil gas and air is a tool in mineral exploration), volcanic activity or geothermal heat streams [40]. This is still the case at the present time [6], as demonstrated by the natural mercury content of river sediments around the Italian geothermal steam fields of Larderello and Monte Aniata, reported (1966) to reach as much as 50,000 ppb [39].

Consequently, the results given must only be seen as an indication of the future general trend.

UNITS AND NOTATIONS

1	ppb	= :	1	part per billion of solids or water,
1	ng/m^3	= .	1	nanogram per cubic meter of air,
1	10	_	1	we should have

1 ton = 1 metric ton.

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metric tons/year	
Market in	
e Common	
throughout th	
r Discharge	
Mercury	

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			1972				1 980				1990	
	Air	Soil	Sedi- ments	Total	Air	Soil	Sedi - ments	Total	Air	Soil	Sedi - ments	Total
FUNCTIONAL												
DISSIPATIVE												
Electro-technical, including dry batteries ⁰	330	100	1	430	235	75	1	310	180	60	ı	240
Dentistry	60	9	54	120	67	~	61	135	70	7	63	140
Paint Industry	100	ı	0	100	30	i	ı	30	10	1	I	10
Paper Industry	2	ı	9	80	1	ı	i	I	I	i	1	ı
Agriculture	3.5	63	1	66.5	2	38	1	40	I	19	I	20
Pharmaceutics	ı	ŝ	37	40	ı	-	19	20	ı	3	10	10
Chlor-alkali Industry (mercury in caustic soda, Cl ₂ , H ₂)	35	6	56	100	18	4	32	54	13	4	21	38
Laboratory and Chemical Products	ı	ı	1	1	30	ı	40	70	20	,	20	40
Present and Future Applications	l	ı	1	ı	43	2	20	70	. 43	2	20	20
Others	10	3	17	30	2	,	ά	5	1	'	1	•
NON-DISSIPATIVE		(
Chlor-alkali Industry	482	ı	238	720	265	1	131	396	189	I	93	282
Catalystics	-	I	60	60	1	1	40	40	1	ı	30	30
TOTAL tons/year	1022.5	184	468	1674.5	6 92	132	346	1170	526	76	257	880
NON-FUNCTIONAL DISSIDATIVE												
Hard Coal, Lignite	06	ı	ł	06	60	1	1	90	120	ı	J	120
Crude Oil	10.5	ł	ł	10.5	12	I	1	12	18	ı		18
Natural Gas	0.5	t	ı	0,5	1	ı	ı		1.5	1	1	1.5
NON-DISSIPATIVE												
Mercury in mining and smelting	27	ı	i	27	21	1	I	21	21	•	ı	50
Refinement of lead, copper, zinc	430	I	ı	430	60	'	I	60	50	ı	1	. 50
TOTAL tons/year	558.0	ł	-	558.0	184	'	ı	184	210.5	ľ	1	210.5
- FUNCTIONAL + NON-FUNCTIONAL	1580.5	184	468	2232.5	876	132	346	1354	736.5	26	257	1090.5

APPENDIX

EUR 6023 – On the future average mercury content of air, soil and river sediments in the EEC and in the world's oceans

by R.H. Van Enk

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This report deals with estimates for the future average mercury concentration levels in air, soil and river sediments in the EEC based on possible future decrease in mercury emissions.

A computer model, already described in EUR 5683, has been extented and used to simulate the pathway of mercury through the environment.

The results regarding future mercury concentrations given by the model are , averages which only indicate the general future trend, and therefore give no predictions for localized pollution. The model shows the possible effect of emission reductions in the various forms of consumption on future mercury pollution within the EEC and in addition it shows the effect of the total mercury released throughout the world on the average mercury concentration in the world's oceans.