

**^{210}Po and ^{210}Pb in the Netherlands:
releases to air from the thermal phosphorus plant
compared to environmental monitoring**

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Thermal Phosphorus plant, Vlissingen, NL

Tallinn, 06 June 2012



Overview

- RIVM (NL Institute for Public Health and the Environment):
Po-210 and Pb-210 measurements (EURATOM)
- The only thermal phosphorus plant in Europe: NORM to air
- NORM emissions of the same order of natural background
- USE OF MODELLING (air dispersion
+ IAEA dose estimate for regulatory purposes)
- ASSESSMENT OF DOSE TO GENERAL PUBLIC:
 - dose from actual emissions < permit
 - plant permitted limit 40 $\mu\text{Sv/y}$
 - about $\frac{1}{4}$ of Po-210 and Pb-210 background with same model

the Netherlands

Thermal
phosphorus
plant



RIVM

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Environmental measurements: RIVM (Utrecht)

High Volume Sampler: from 1990 to 2011



4 filter units (glass fiber 18x23 cm²)
50000 m³/week

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Environmental measurements: from 1990 to 2011



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High Volume Sampler: 2011 onwards



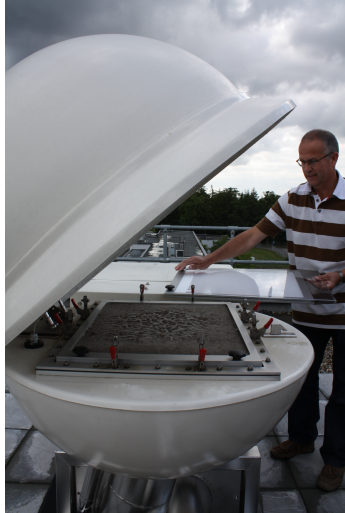
125000 m³/week – filter type G3 (polypropylene) 57x46 cm²
Efficiency 96±1% for Pb-210 and Be-7

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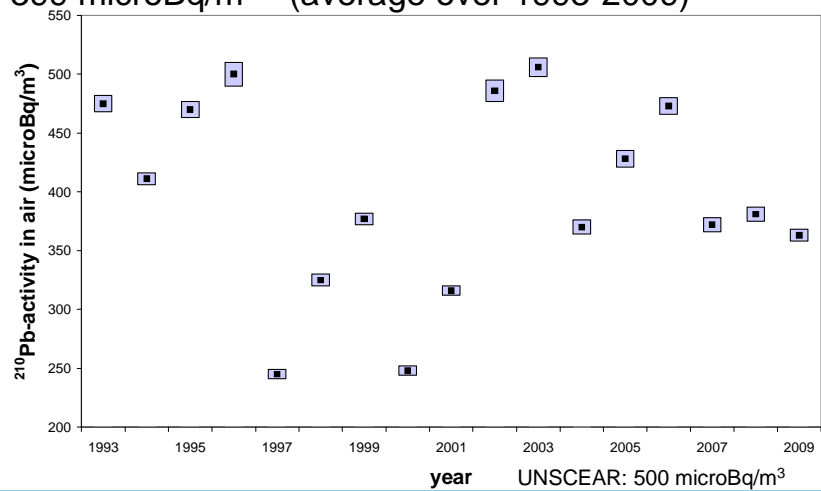
High Volume Sampler: 2011 onwards



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Pb-210 activity in air γ -spectrometry on 50000 m³/week HPGe-detector 399 microBq/m³ (average over 1993-2009)



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Environmental measurements: RIVM (Utrecht) deposition:



Weekly:
Pb-210 (γ -spectrometry)



Monthly (1 to 20L volume):
Po-210
+100 ml HCl (6 mol/L) carrier

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Deposition: Po-210 analysis

- Rinse with 1L demineralised water
- Po-208 or Po-209 tracer solution added
- Evaporate to 100 mL on cooking plate
- Reduce to residue on water bath
- Dissolve in HCl (6 mol/L)
- Add $\text{NH}_2\text{OH}\cdot\text{HCl}$ and adjust to pH 1,5
- Auto-electrodeposition on silver disk at 90°C for 4h
- α -spectrometry

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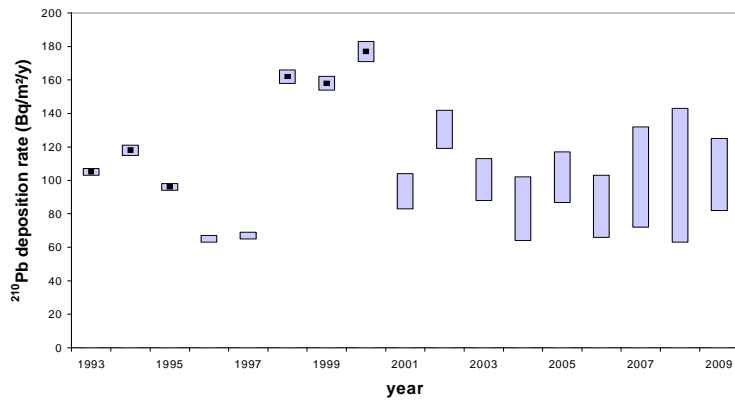
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Yearly Pb-210 activity deposited in Bilthoven, NL

60 to 180 Bq/m²/y

bar: 2σ

no dot if at least one weekly measurement ≤ DL



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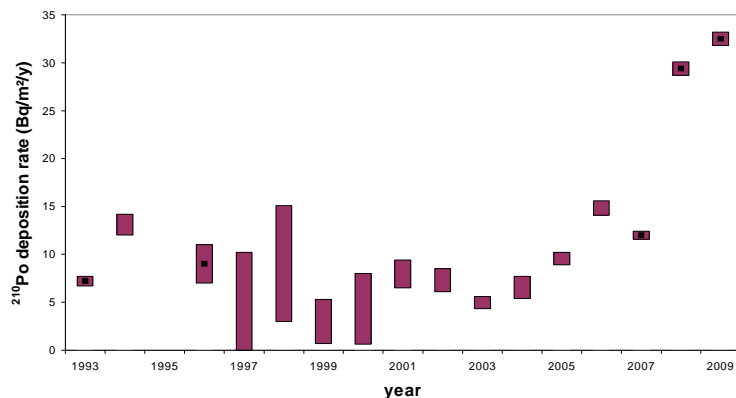
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Yearly Po-210 activity deposited in Bilthoven, NL

0 to 33 Bq/m²/y

bar: 2σ

no dot if at least one monthly measurement ≤ DL



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Thermal elemental phosphorus plants

MONSANTO: USA (Idaho, Soda Springs)

Kazakhstan (Kazphosphate)

India

China? (small operators?)

Europe

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The only **thermal** phosphorus plant in Europe



1963: NL Nuclear Energy Act

1973: NPP on river estuary

1983: NPP monitoring shows
Po-210 > background

CAUSE IDENTIFIED:
Hoechst (now Thermphos)

1985: operating permit granted

1987: plant reports emissions

the granted emission limits are
based on dose assessment
to the public

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Phosphorus production in the past



The Alchemist in Search of the Philosophers' Stone discovers Phosphorus

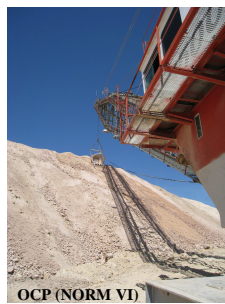
H. Brand, 1669

Credit: Joseph Wright of Derby, 1771

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Phosphorus production now (thermal)

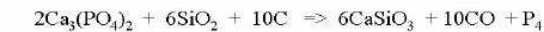
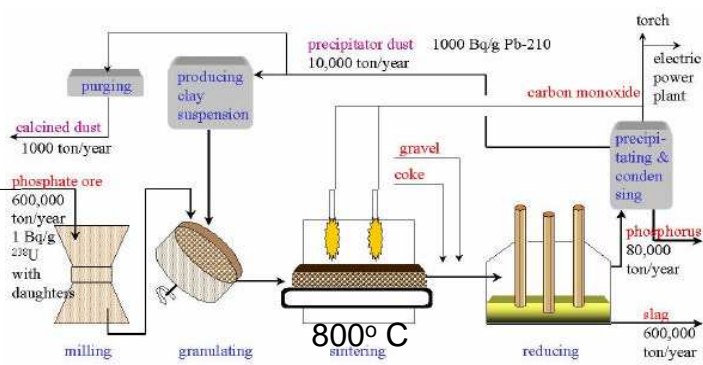
see García-Tenorio, these proceedings, p. 202



OCP (NORM VI)
phosphate ore:

apatite

$\text{Ca}_5(\text{PO}_4)_3(\text{F}/\text{OH})$



phosphate ore and gravel and coke => slag and carbon monoxide and phosphorus

PHOSPHORUS PRODUCTION AT THERMPHOS INTERNATIONAL

From Erkens

- slag: incidental release to ground
- sintering: continuous release to air of Po-210, Pb-210, Cd, Pb, Zn, Cu, Cr, Hg, dioxine



Only **thermal** phosphorus plant in Europe

450 employees (in NL)

• **Elemental phosphorus (sintering)**

• Phosphoric acid and phosphate (wet process): DAP, MAP

• NTTP (sodiumtripolyphosphate)

+ Marketing and sales

1997: independent within Hoechst

2000: independent

2003: acquired by (the same group which owns) Kazphosphate

DAP, MAP: diammonium, monoammonium phosphate

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Thermphos phosphorus products (list from 1997)

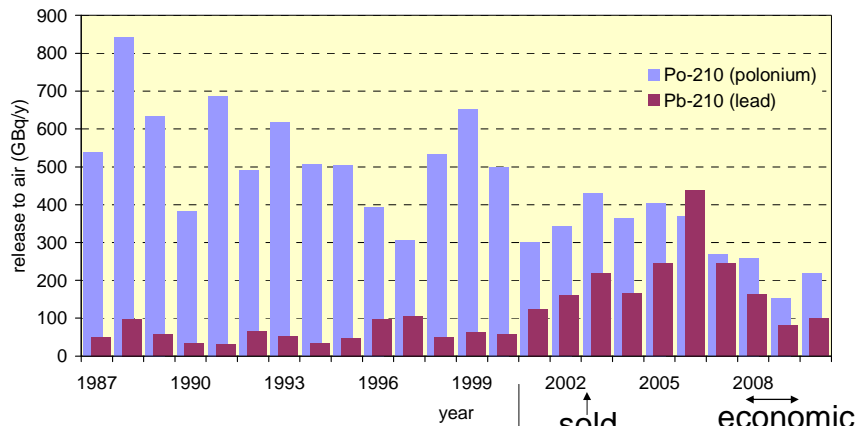
kton	product	use
53	P ₄	export to industries producing all of the following:
42	NTTP	detergents
18	H ₃ PO ₄	foodstuff + acid pickling baths (part returns to wet process)
0,4	PCl ₅	pharmaceutical products
1,4	POCl ₃	plastics (plasticiser, stabiliser, flame retardant)
3,2	PCl ₃	pesticide (mainly)
2,0	P ₂ S ₅	(car) industry (lubricant, pneumatic oil)
0,8	FeP	additive to steel
120	Total	

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Po-210, Pb-210 emissions to air



USA: phosphate ore export stop
(stop of Florida ore)

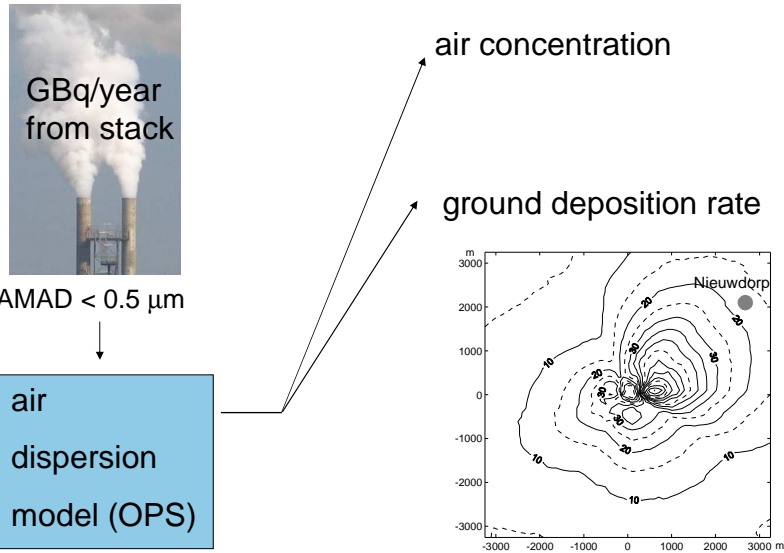


Phosphorus production in the future of Thermphos

from secondary fosfor sources (not ores but return streams
– e.g. manure from water purification)

2010	15%
horizon 2020	100%

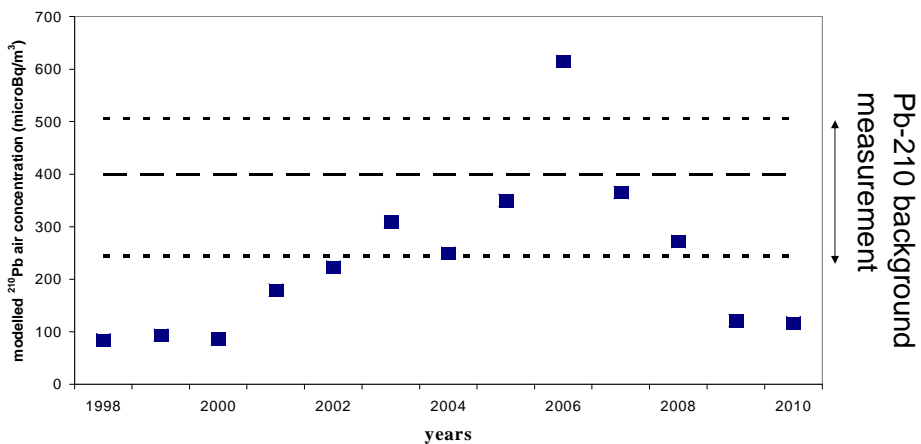
Model of environmental concentration,
from reported emissions to air



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**air dispersion model (OPS) at Nieuwdorp village,
4 km downwind
microBq/m³**



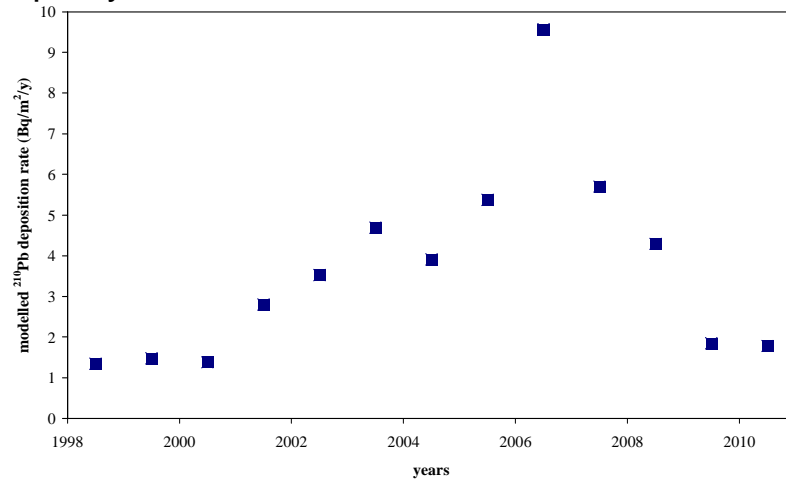
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**air dispersion model (OPS) at Nieuwdorp village,
4 km downwind**

Bq/m²/y

Pb-210

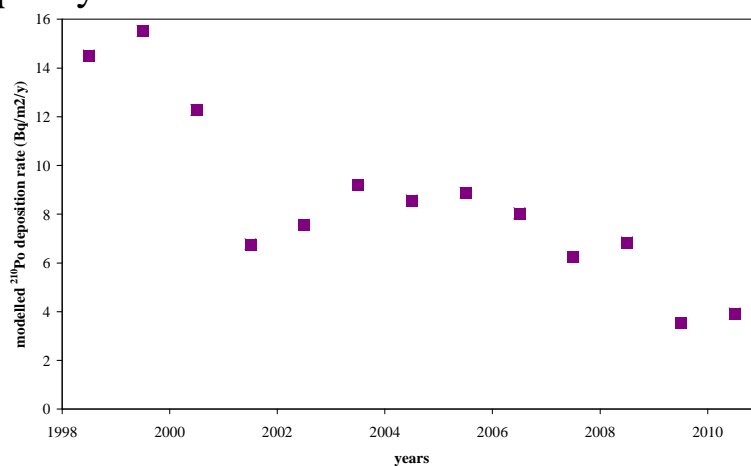


negligible compared to 60 - 180 Bq/m²/y background level

**air dispersion model (OPS) at Nieuwdorp village,
4 km downwind**

Bq/m²/y

Po-210



compare to 0 - 33 Bq/m²/y background level

Assessment of effective dose

from reported emissions to air



GBq/year
from stack

AMAD < 0.5 μm

air
dispersion
model (OPS)

air concentration → inhalation

ground deposition rate

→ ingestion pathway

follows IAEA SRS No. 19, 2001

emission constant over 25 y

food basket:

grain, potatoes, meat, milk,
leafy vegetables, + FRUIT

+ full ingrowth calculation

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Ingestion dose

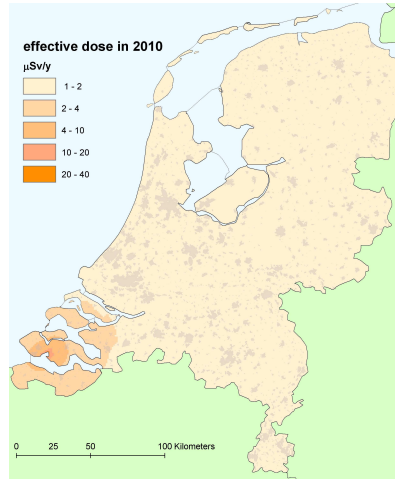
	potatoes	cereals	fruit	veg	milk	meat	total
Yearly consumption (kg)	51	71	19	42	147	10	340
<u>25 years plant operations</u>							
Pb-210: 500 GBq/y							
Effective dose (microSv/y)	0.7	0.3	0.6	0.7	<0.1	<0.1	2.3
Po-210: 500 GBq/y							
Effective dose (microSv/y)	0.3	0.2	0.6	0.6	<0.1	<0.1	1.7
<u>Natural background</u>							
Pb-210 100 Bq/m ² /y							
Effective dose (microSv/y)							140
Po-210 10 Bq/m ² /y							
Effective dose (microSv/y)							7

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Effective dose in NL

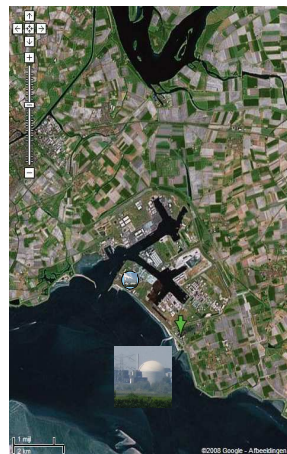
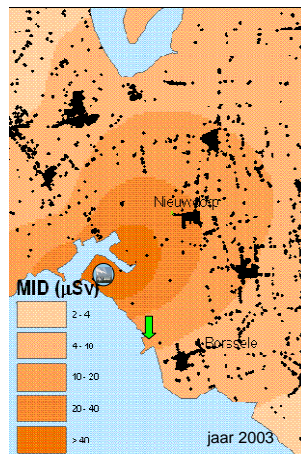


Permitted level
phosphorus plant:
40 $\mu\text{Sv}/\text{year}$ at
closest dwellings

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Thermal phosphorus: effective dose from air emissions



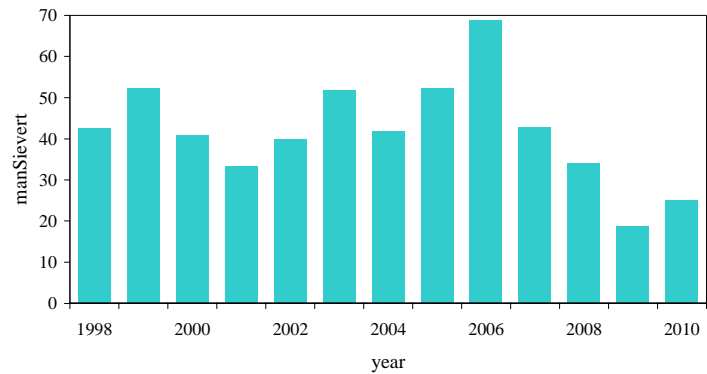
Collective dose: \int effective dose x inhabitants

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Collective effective dose in NL from emissions to air in NL

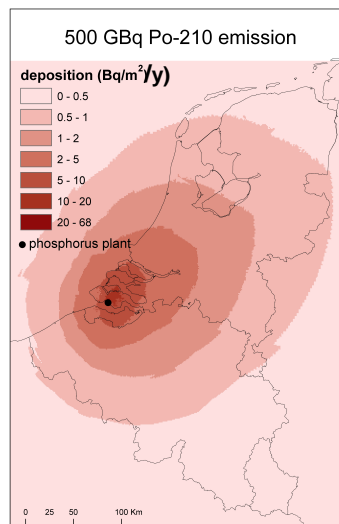


the model predicts increased Po-210 and Pb-210 in Belgium

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Thermal phosphorus: deposition rate in Belgium from NL air emissions



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Summary

Thermal phosphorus plant emissions to air

Before 2000:

500 GBq Po-210

50 Gbq Pb-210

After 2000:

250 GBq Po-210

200 GBq Pb-210 (>400 GBq in 2006)

Model of environmental load comparable to background level

Dose assessment

1. reported emissions to air
2. air transport modelling
3. dose from inhalation, ingestion
(food basket:
grain, potatoes, vegetables, meat, milk, + FRUIT)

→ in 2010 < 40 microSv/y (permitted level)

collective dose: 30 manSv/year in the Netherlands