HIGH LEVELS OF PCDD/F, PBDD/F AND PCB IN EGGS AROUND POLLUTION SOURCES DEMONSTRATES THE NEED TO REVIEW SOIL STANDARDS

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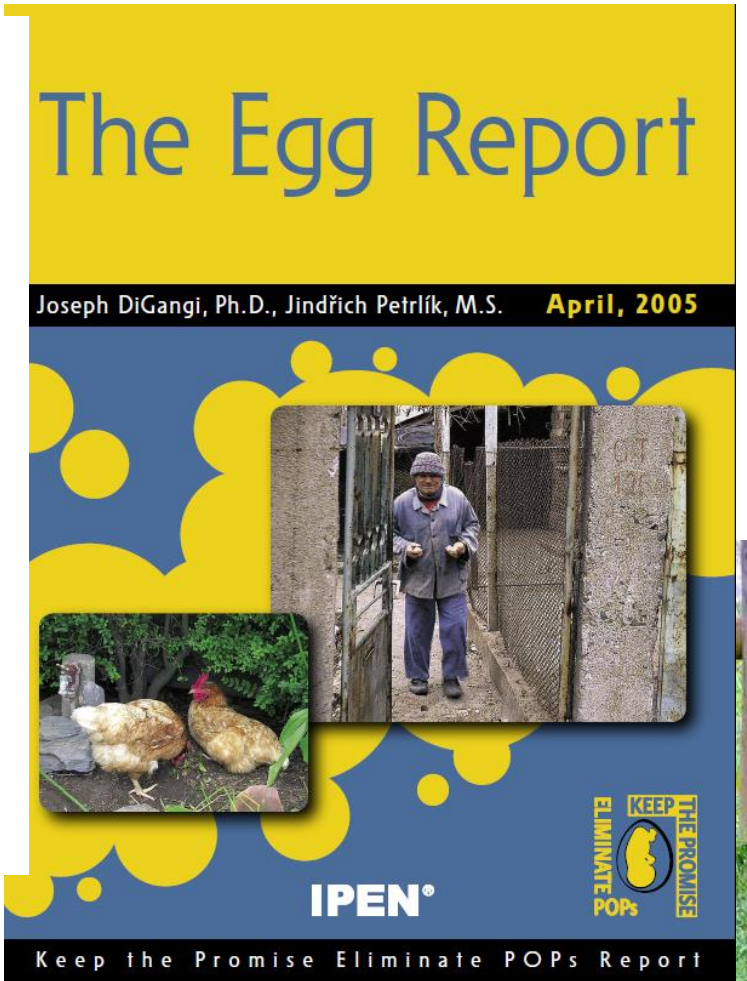
Background

- There have been an increasing number of reports on contamination of eggs with PCDD/Fs and dl-PCBs in recent years. In Netherlands more than 50% of eggs from small scale free range chicken holders were above EU limits (Hoogenboom et al.; Arkenbout Dioxin 2015).

- Eggs have been found to be sensitive indicators for PCDD/F and PCB contamination in soils and are an important exposure pathway from soil pollution to humans (IPEN egg reports).
- Chickens and eggs might therefore be ideal “active samplers” and indicator species for contaminated soils but there are, as yet, few systematic studies linking pollution sources, related soil levels and contaminants in food.



Materials and Methods

- Eggs were sampled at two potential hot spots in Germany; close to a condenser factory and the second close to a hazardous landfill site. For both sites two individual chicken flocks were sampled.
- Eggs from two flocks close to one municipal waste incinerator in China. Reference samples of eggs were also analysed after being purchased from a Chinese supermarket.
- Eggs from one flock at an e-waste site from Thailand.
- The egg samples were pooled in each case. For each of the German sites 10 to 20 eggs were pooled and for each flock in China and Thailand 3 or 4 eggs were pooled for analysis.

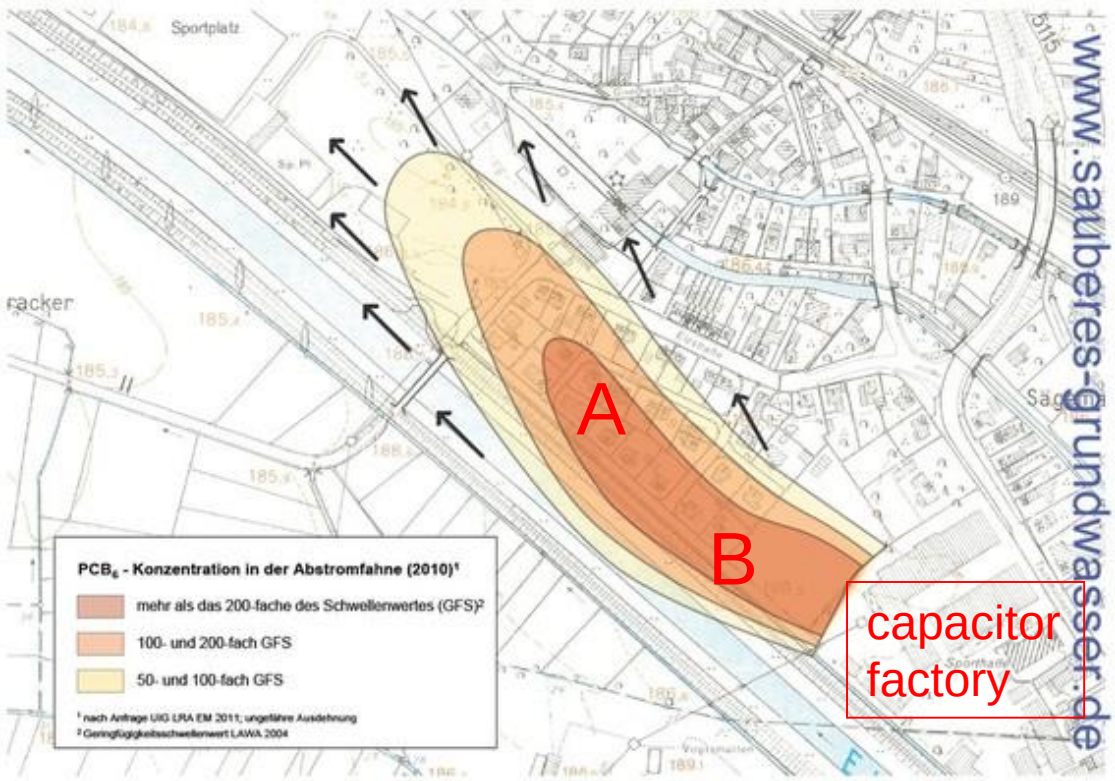
Materials and Methods

- Bioassay. All samples were analyzed at Bio Detection System (BDS) in Amterdam for dioxin-like activity according to the standard procedures of the DR CALUX® method from BDS.
- Instrumental analysis. All samples were analysed by HRGC/HRMS in ISO 17025 accredited laboratories with a resolution >10,000 using 13C isotope labelled standards. PCDD/F and dl-PCB analysis followed the European Union's methods of analysis for the control of levels of PCDD/Fs and dl-PCBs for levels in certain foodstuffs in Commission Regulation (EC) No 252/20129.
- Selected samples from China and Thailand were also analysed for PBDD/F using 13C isotope labelled standards.

Egg monitoring around a capacitor factory (Germany)

- Chicken eggs were sampled from two private chicken farmers (A and B) close to a factory in Köndringen/Teningen, a small town in South-West Germany which was used for the production of capacitors from 1932 on including PCB containing capacitors for some time.

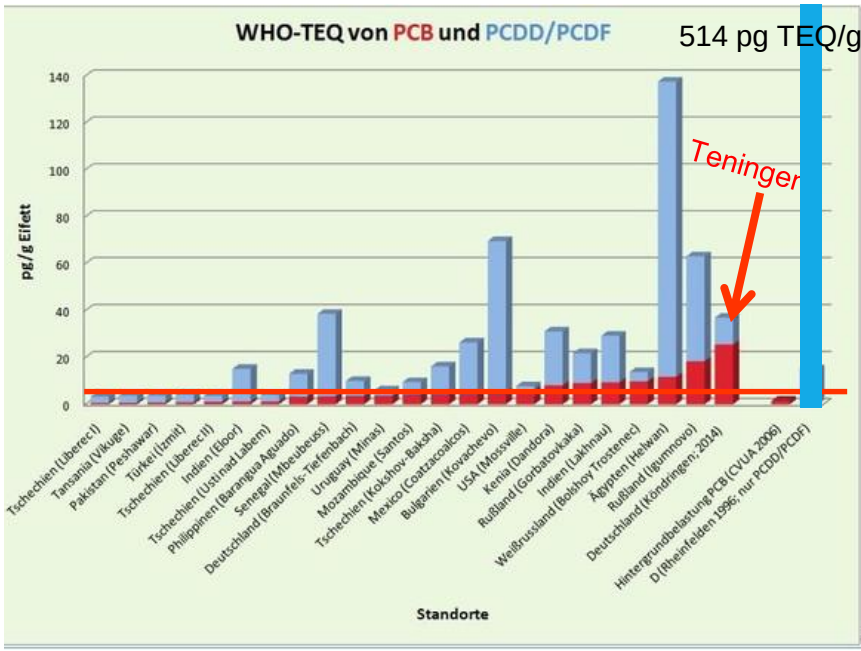
The groundwater below the former production site and the associated landfill is contaminated with PCBs and a PCB plume contaminates the ground water of the nearby town.



Chicken egg monitoring around a condenser factory

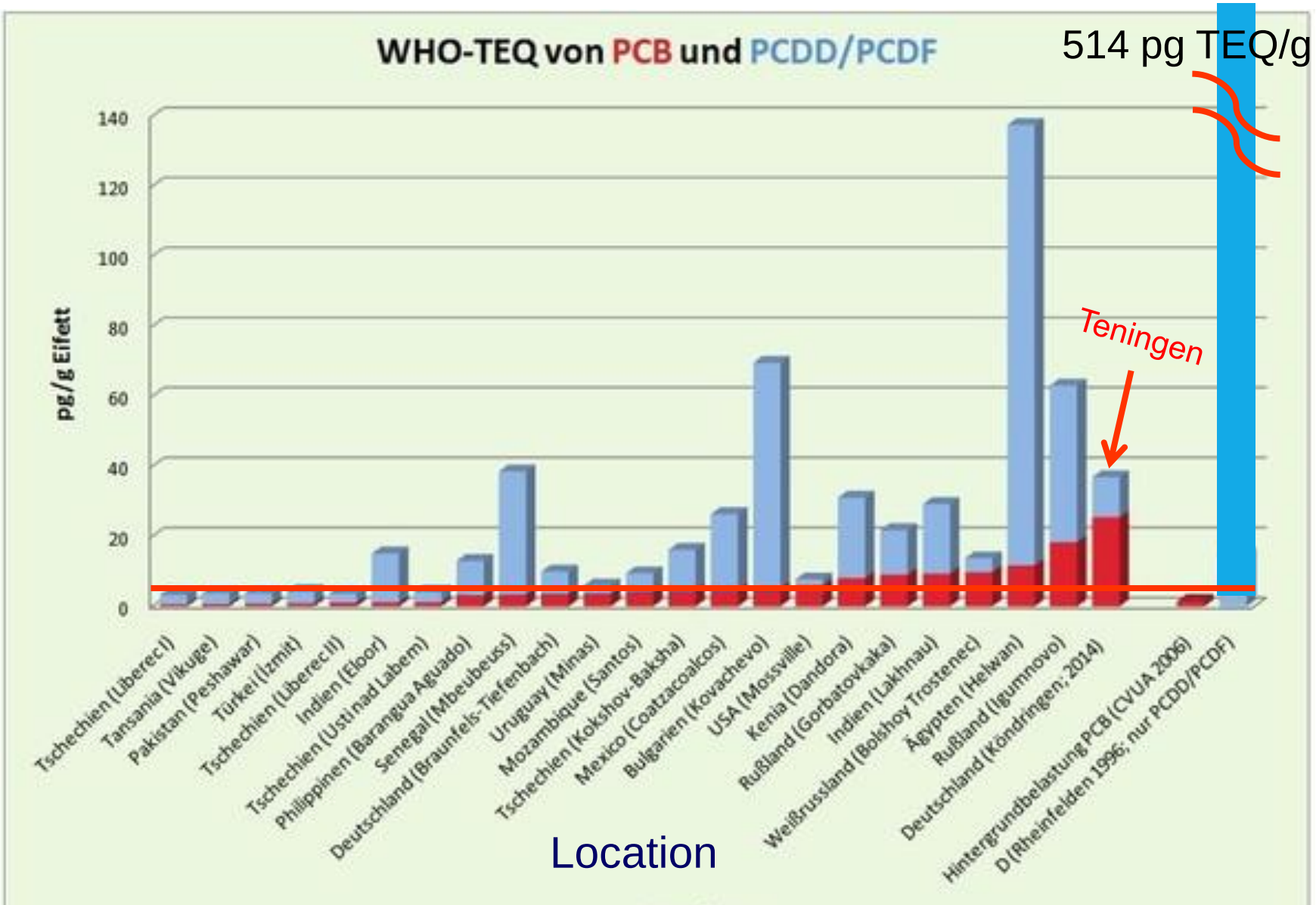
- Bioassay screening for dioxin-like toxicity was conducted for eggs from the two flocks/sites with DR CALUX. The BEQs from the bio-assays were 18 pg BEQ/g fat at both sites (considerably higher than the regulatory limit of 5 pg PCDD/F-PCB-TEQ/kg fat). PCBs were the main contributor to the BEQ for both samples.

- For confirmation the eggs were also analysed by instrumental analysis (HRGC/HRMS). The same egg samples from chicken holder A were highly contaminated (36.4 pg TEQ/kg fat) mainly from dl-PCB (25 pg TEQ/g fat). The levels in eggs from chicken farmer B were nearly as highly contaminated with 31.9 pg TEQ/kg fat mainly due to dl-PCB (25.5 pg TEQ/g fat).



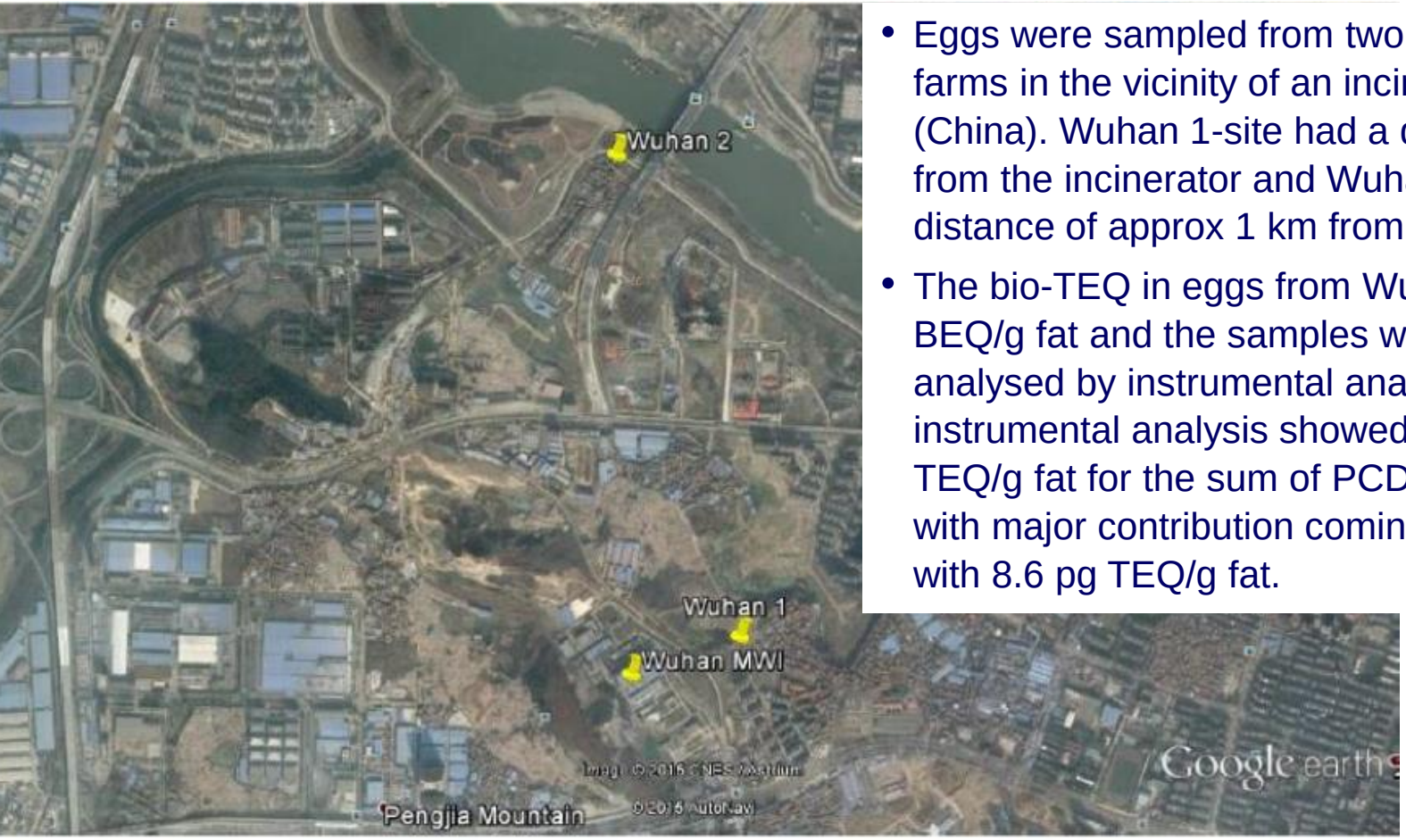
Chicken egg monitoring around a condenser factory

- Comparison with eggs from IPEN 2005 screening and German data.



Egg monitoring close to an incinerator (China)

- The egg sample from a Chinese supermarket showed low levels both in the DR CALUX bioassay (1.2 pg BEQ/g fat) and from instrumental analysis (total TEQ of PCDD/F and dl-PCB of 0.66 pg TEQ/g fat) and were comparable to, for example, background levels in Europe.



- Eggs were sampled from two private chicken farms in the vicinity of an incinerator in Wuhan (China). Wuhan 1-site had a distance <0.3 km from the incinerator and Wuhan 2-site had a distance of approx 1 km from the incinerator.
- The bio-TEQ in eggs from Wuhan 2 was 8.8 pg BEQ/g fat and the samples were therefore also analysed by instrumental analysis. The instrumental analysis showed levels of 13.3 pg TEQ/g fat for the sum of PCDD/F and dl-PCB with major contribution coming from PCDD/F with 8.6 pg TEQ/g fat.

Egg monitoring close to an incinerator (China)

- The bio-TEQ in eggs from Wuhan 1-site was 35 pg BEQ/g fat and the GC/MS showed levels of 16 pg TEQ/g fat for the sum of PCDD/F and dl-PCB with major contribution from PCDD/F with 12.2 pg TEQ/g fat.
- Due to the discrepancy between BEQ and TEQ, PBDD/F were also screened in the eggs and high levels of PBDD/F were found (29 pg TEQ/g fat). This PBDD/F contamination from Wuhan-1 can explain the large gap between BEQ and instrumental PCDD/F and dl-PCB TEQ.
- This egg contamination case demonstrates that also PBDD/F are bio-accumulating in eggs.

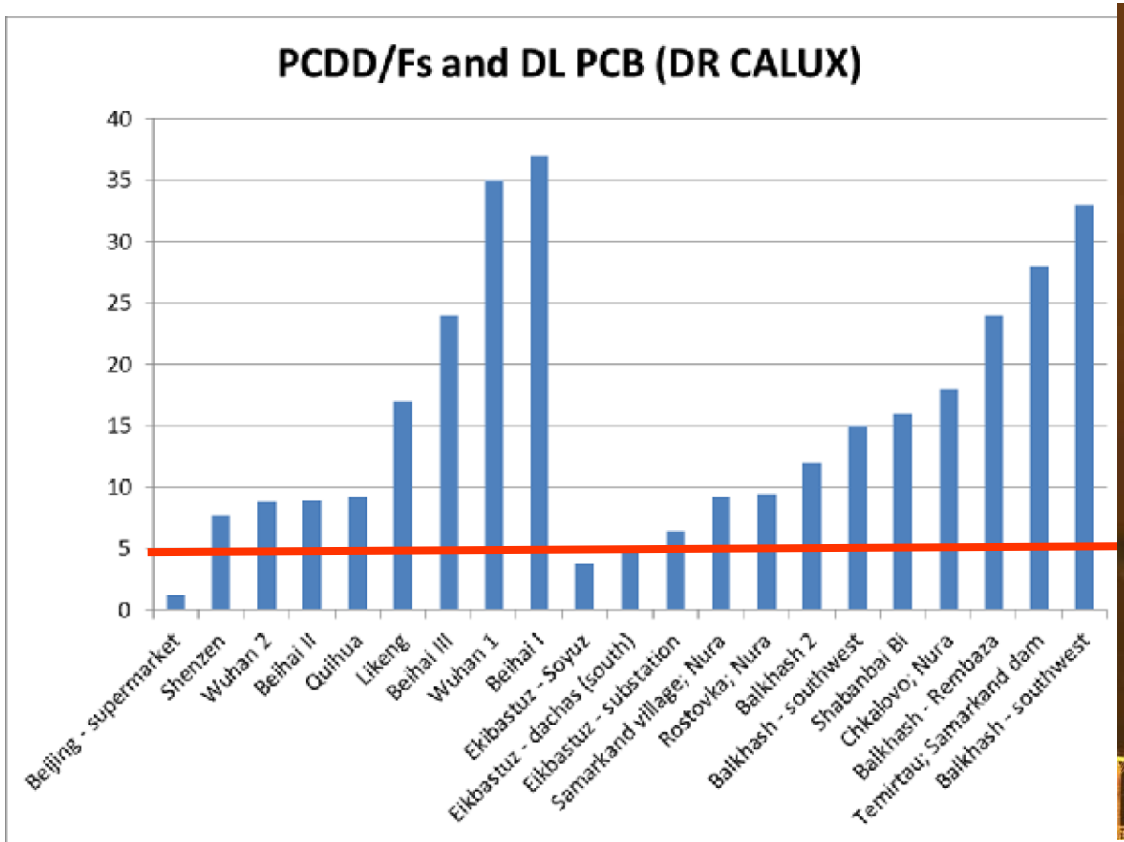
Egg monitoring at an e-waste site (Thailand)

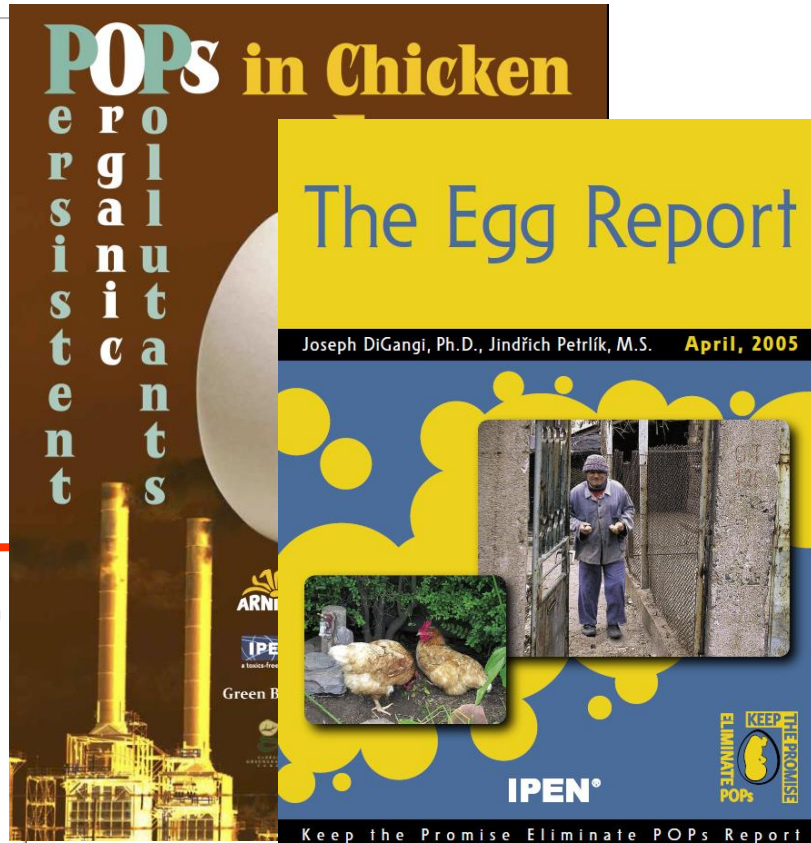
- Eggs were sampled at an area where different metallic waste including e-waste was treated including open burning activities. The bio-TEQ in eggs from the site showed extreme high levels of 100 pg BEQ/g fat (83 in Dioxin-; 17 pg TEQ/g in PCB-fraction) (EU limit: 5 pg TEQ/g fat).
- Instrumental analysis for PCDD/F: 55.6 pg TEQ/g fat.
- Instrumental analysis for PBDD/F: 22.9 pg TEQ/g fat

Analysis parameter	Unit	Result	Limit of quantification
PBDD 2378-congeners			
2378-TetraBDD[c]	pg/g fat	**nd**	0,40
12378-PentaBDD[c]	pg/g fat	**nd**	1,2
123478/123678-HexaBDD[c,d]	pg/g fat	**11,8**	1,8
123789-HexaBDD[c]	pg/g fat	**8,36**	0,9
1234678-HeptaBDD	pg/g fat	**15,9**	2,7
12346789-OctaBDD	pg/g fat	**81,2**	8,1
PBDF 2378-congeners			
2378-TetraBDF[c]	pg/g fat	**3,10**	0,3
12378-PentaBDF[c]	pg/g fat	**8,08**	0,6
23478-PentaBDF[c]	pg/g fat	**17,3**	0,6
123478/123678-HexaBDF[c,d]	pg/g fat	**49,7**	1,8
234678-HexaBDF[c]	pg/g fat	**nd**	16,4[f]
123789-HexaBDF[c]	pg/g fat	**nd**	37,6[f]
1234678-HeptaBDF	pg/g fat	**280**	2,3
1234789-HeptaBDF	pg/g fat	**nd**	4,4
12346789-OctaBDF	pg/g fat	**391**	8,1

Indication of critical soil levels from other studies and consequences for soil limit values

- Overall the IPEN global egg studies and the cases from Germany or Netherlands (Arkenbout 2015) on TEQ levels in eggs around industrial emission sources (PCB use/processing, incinerators, metal industries, etc.) demonstrated that in many areas soils can be polluted with PCDD/F or PCB levels at which eggs can be (highly) contaminated.





Problematic PCDD/F and PCB levels in soil for free range chicken and egg production

What are critical soil levels for impacting an egg above regulatory limit?

- For the two German sites also soils levels have been measured. Around the hazardous landfill the competent authority (LANUV) found dl-PCBs between 3.1 and 6.6 ng WHO-PCB-TEQ/kg dm which is ca. 6 to 10 times above background soil levels in German pasture land. The PCB-levels in soils from Teningen where chickens were picking were between 2.3 and 4.4 ng PCB-TEQ/kg (ca. 4-9 times background).
- Also studies in the Netherlands have indicated that eggs from free-range chicken on soils with levels of 2 to 4 ng PCDD/F-TEQ/kg dm frequently exceed EU limits (Kijlstra et al. 2007 Poult Sci. 86:2042-2048).
- However most chicken herds from rural areas in SW-Germany had low PCDD/F and PCB levels comparable to eggs from industrial production (Malisch et al. 2006).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Problematic PCDD/F and PCB levels in soil for free range chicken and egg production

- With a total uptake of 25 pg (50 pg) TEQ/day a chicken reaches the current EU-limit of 2.5 pg (5 pg) for PCDD/F (sum PCDD/F-PCB) TEQ/g fat in egg (Hoogenboom 2013; UBA Workshop Bonn).
- Free range chicken have a soil uptake of 10 to 30% of their feed which translates to approximately 12 to 36 g of soil/day.
- With a carry over of approx. 50% for the TEQ-relevant PCDD/F (and PCB) the problematic levels in soils are 1.4 to 4.2 ng TEQ/kg for PCDD/F and 2.8 to 8.4 for sum PCDD/F+PCB.
- These levels are extremely low and are exeeded in many areas of industrial emissions and residential areas.


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National PCDD/F Limits in Soil

- A major challenge is that the levels of contamination in the soil which result in excessive levels of contamination of chicken/egg are below the current regulatory soil limits. Therefore an update is needed.

Canadian Environmental Quality Guidelines			
4 ng/kg TEQ	**Alert level**	**CCME, 2005a**	**a**
New Zealand Interim Acceptance Criteria			
10 ng/kg TEQ	**Agricultural**	MoE, 1997	b
1,500 ng/kg TEQ	Residential	MoE, 1997	b
18,000 ng/kg TEQ	Industrial	MoE, 1997	b
Germany Federal and Lander Ministers of the Environment recommendations			
5-40 ng/kg TEQ	**Agriculture**	EU, 1999	c
100 ng/kg TEQ	Playgrounds	EU, 1999	c
1,000 ng/kg TEQ	Residential areas	EU, 1999	c
10,000 ng/kg TEQ	Industrial areas	EU, 1999	c
The Netherlands Guidelines			
1,000 ng/kg TEQ	Residential and agricultural areas	EU, 1999	c
10 ng/kg TEQ	**Dairy farming**	EU, 1999	c

- In Germany and Netherland, for example, the regulatory limit for soil for residential areas is 1,000 ng PCDD/F-TEQ/kg dm. If chickens were kept on land with these levels this could result in eggs with approx. 800 pg TEQ/g fat! For a 16 kg child a single egg (10 g fat) would exceed the TDI by 250 times.

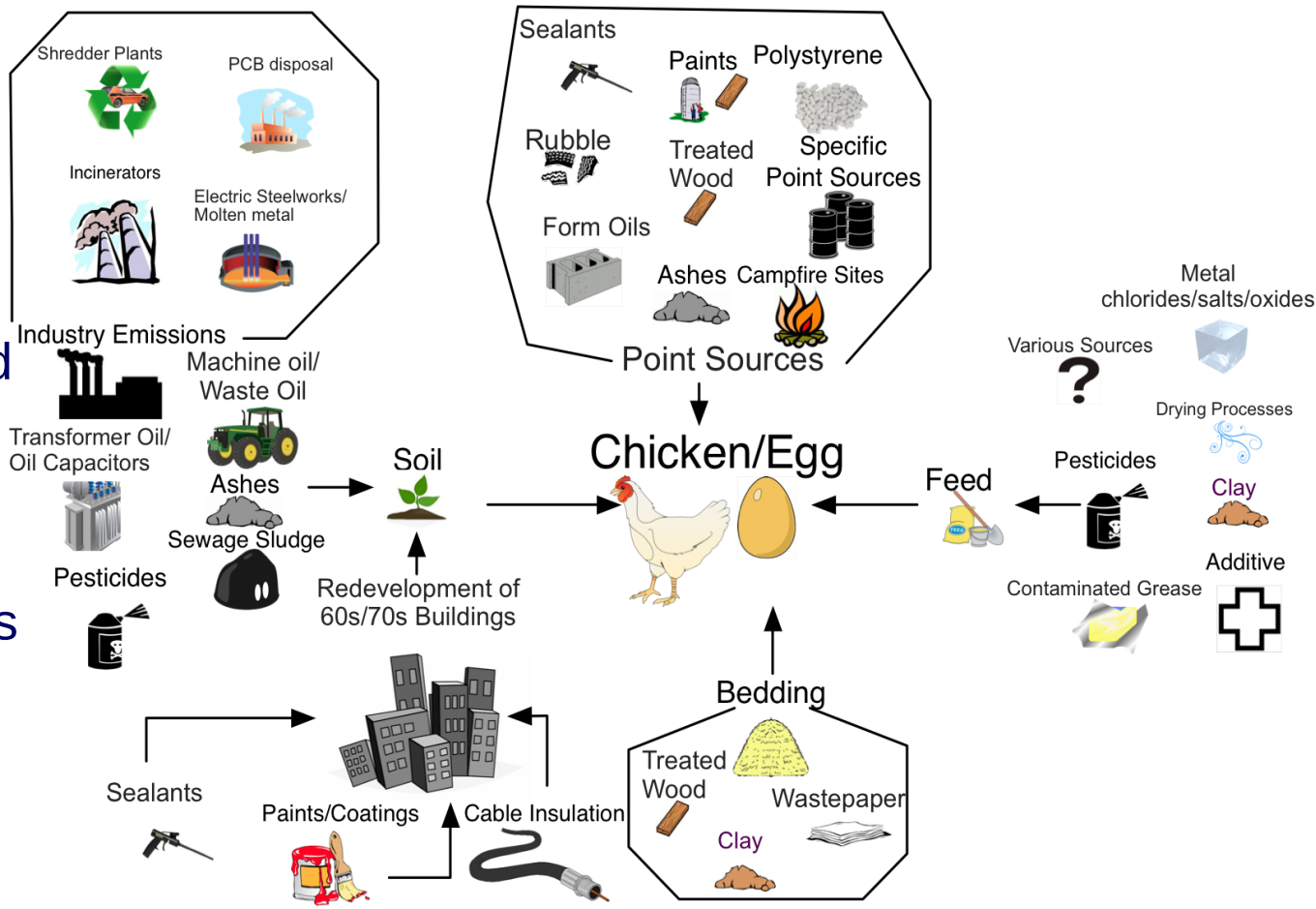
Consequences for industrial emissions and for controlling ashes from thermal processes

- The low PCDD/F and PCB levels in soil at which chicken/eggs can become contaminated above regulatory limits and health based limits highlights the need to strictly control industrial and other emissions.
- It is also important to ensure the safe disposal of residues from waste incinerators and ashes from residential sources where waste plastics/PVC or contaminated wood are co-incinerated.
- Residual ashes with contamination levels as low as 50 ng TEQ/kg can be a risk sources. Even if such ash is “diluted” on soils the PCDD/F can accumulate over time with repeated applications.
- It needs to be highlighted that the provisional Basel Convention low POPs limit for dioxin contaminated waste of 15,000 ng TEQ/kg is much too high and needs urgently to be re-evaluated and reduced: A single kilogram of ash meeting the Basel “low POPs” level could contaminate 7 tonnes of soil to a level where eggs would not meet EU regulatory limits if laying chickens were kept on it.

Consequences for industrial emissions and for controlling ashes from thermal processes

- When assessing contamination sources for a flock, potential local sources on the farm should be considered together with larger emission sources in the vicinity (and the feed and bedding).

- It is also useful to assess at least two independent flocks around pollution sources.
- Together with detailed soil investigations including assessments of fingerprints of sources and soils before firm conclusions can be reached.



Consequences for industrial emissions and for controlling ashes from thermal processes

- Contamination around current and former (industrial) Dioxin and PCB emission sources need to be assessed in respect to food safety considering problematic soil levels.
- The regulatory framework needs to be updated by the establishment of lower thresholds for soil contamination reflecting the levels at which land uses need to be restricted or managed to minimise exposure.
- There is a need for compensation of farmers/owners for loss of the use of land and in some cases for historic (and current) exposure considering the polluter pays principle.
- Stringent emission standards and residue treatment can reduce long-term impact and (external) costs associated with the loss of productive land due to emission sources.

Conclusion on monitoring using bioassay and role of brominated dioxins and other DLCs

- The study demonstrates the utility of using bioassay for monitoring of chicken eggs. Bioassays have the dual benefit of being both a cheap and useful tool to measure PCDD/F and PCB in eggs and can indicate contamination in soils via the egg levels.
- The bioassay approach also detect PBDD/F and mixed-halogenated PXDD/F in eggs (and associated soils). Due to the complexity of instrumental analysis of the mixed halogenated PXDD/F currently only total dioxin-toxicity measured by appropriate bioassays can address this challenge at reasonable cost.
- PBDD/F and PXDD/F are not yet regulated in foodstuffs, sewage sludge or soils. Due to the increasing stocks of PBDD/F precursors (PBDEs & other brominated aromatic chemicals) and increasing thermal treatment of flame retarded waste (for e-waste often by open burning) they should also be evaluated for possible consideration in regulatory development (total dioxin-like toxicity?).

Thank you for your attention

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