Dioxins in Vietnamese, Vietnamese Food and Environment: Potential Relevance of Hot Spots from Recent Findings

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University of Texas, Dallas, USA
ERGO is situated at the Northern part of Germany, Europe.
Some Words about ERGO

• More than 20 years experience in dioxin analysis
• Capacity: 3000 dioxin analyses per year
• 3 (4) HRGC/HRMS instruments
• Authorized by WHO and European Union for Food and Feeding stuff analysis for dioxin
• Accreditation in accordance with EN/ISO 17025
• Customers in more than 30 countries worldwide
The ERGO Team

- Chemists: 19
- Engineers: 7
- Laboratory technicians: 12
- Technicians: 5
- Support: 6
- Administration: 9
Thanth’s New: eurofins and ERGO

- ERGO belongs to the eurofins Group, a globally working group in the bioanalytical market with about 2,500 employees in 40 laboratories in EU and US and Asia.
- 5,000 different analytical methods.
- Investments in R&D and state-of-the-art equipment.
- Expertise in technical & regulatory matters.
Components of Question

• Polychlorinated dioxins and furans
  PCDDs and PCDFs
  \[ \text{C}_{12}\text{H}_x\text{C}_{l y}\text{O}_2 \ (x+y=8; \ n=75) \]
  \[ \text{C}_{12}\text{H}_x\text{C}_{l y}\text{O} \ (x+y=8; \ n=135) \]

  • Relevant for Humans:
    • 17 congeners with 2,3,7,8-chlorine substitution

• Polychlorinated biphenyls
  PCBs
  \[ \text{C}_{12}\text{H}_x\text{C}_{l y} \ (x+y=10; \ n=209) \]

  • Relevant for Humans:
    • 12 dioxin-like congeners, 4 non-ortho, 8 mono-ortho
Outline

• Sources
• Tissue selection
• Relevance of food
• Tolerably daily intake
• Body burden
• Time trends

• Factors of influence
• Special consumptions
• Occupational exposure

• Vietnam
  – general
  – early investigations
  – recent investigations
  – hot spots
Chemical & Industrial Sources for Dioxins

- Waste incineration
- Thermal industrial processes
- Metal production
- Chlorine industry
- Bleaching of pulp
- Traffic
- Domestic furnaces
Humans may become contaminated with PCDD/Fs and dioxin-like PCBs by:

- Environmental exposure
- Occupational exposure
- Accidental exposure
Incineration Industry
Chlorine Industry
Thermal Metal Process
Car Exhaust

Rain/deposition
Dump Site/Sledge
Plants, gras
Soil / Water

Beef, cow milk/-products
Pork/-products
Chicken/-products
Fish/-products
Vegetables

Humans
Importance of Food

• Consumption of food contributes normally with >95% to the daily total PCDD/PCDF and PCB uptake

• Food rich in fat, especially originating from animals, is most important

• Food originating from plants usually shows a lower content of PCDDs/PCDFs
Adipose and Serum 2,3,7,8-TCDD Levels

Adipose 2,3,7,8-TCDD pg/g - lipid weight basis

Serum 2,3,7,8-TCDD pg/g - lipid weight basis

Patterson et al., 1988
PCDDs/PCDFs in Humans
Comparison of Blood and Milk, 1994
Values in pg/g, lipid based

Päpke, 1998
Tolerable Intake for Dioxins and Dioxin-like PCBs

WHO, 1998
1 – 4 pg TEQ/kg b.w./day

SCF, 2001
14 pg TEQ/kg b.w./week

JECFA, 2001
70 pg TEQ/kg b.w./month
Average daily PCDD/F intake via food in pg TEQ, Comparison 1989/1996

Forst et al., 1997
Reduction of Dioxin Emissions by different Ordinances in Germany

- Ban of PCBs and PCP, 1989
- Limiting value for combustion plants, 1990: 0,1 ng I-TEQ/m³
- Ban of scavengers, 1992
- Limiting values for sewage sludge, 1992: 100 ng I-TEQ/kg
- Maximum workplace concentration, 1993: 50 pg I-TEQ/m³
- Prohibition on certain chemicals, 1994
Dietary Intake of PCDD/Fs (I-TEQ, pg/d) from Major Food Groups, Adults or Total Population

*other foods = cereals, cereal products, vegetables, fruit, vegetable fats and oils

European Commission
SCOOP Task 3.2.5
June 7, 2000
Contribution of PCDDs/PCDFs and Dioxin-like PCBs to Total WHO-TEQ

(Fürst and Päpke, 2002)

- PCDDs/PCDFs: 49%
- non-ortho PCBs: 30%
- mono-ortho PCBs: 21%
RESULTS OF THE WHO-COORDINATED EXPOSURE STUDY ON THE LEVELS OF PCBs, PCDDs AND PCDFs IN HUMAN MILK

Rainer Malisch and Rolaf van Leeuwen, Dioxin 2003
Temporal Trend of PCDD/Fs in Human Milk

Rolaf van Leeuwen and Rainer Malisch, Dioxin 2002

Graph showing the temporal trend of PCDD/Fs in human milk for various countries from 1988 to 2002.
Biological Half-Lives of PCDD/Fs and PCBs in Humans

Problem: Most PCDD/Fs and PCBs have long half-lives, e.g.

2.3.7.8.-TCDD: 7 years

Consequence: Permanent intake of PCDD/Fs via food leads to high concentration in humans (accumulation)
Factors of Influence on Human Body Burden for PCDDs/PCDFs

- Consumption habits
- Severe weight loss
- Age dependency
- Women: nursing / non nursing
- Infants: nursed / not nursed
PCDDs/Fs in Selected Population, Influence of Fish Consumption, Kivivanta et al., 2000

Finland Fishermen n=26
Finland Controls n=45
Germany Background n=44

- 2.3.7.8-TCDD
- 2.3.4.7.8-PeCDF
- I-TEQ

(pg/g lipid base)
Increase of PCDD/PCDF Concentration (TEQ) in Human Blood due to Weight Loss in pg/g, lipid based

Zober, Päpke, 1995

Time between 1st and 2nd analysis: 8 months

weight loss: 20 - 25kg
TEQ Concentrations from PCDDs, PCDFs, n-orthoPCBs, m-orthoPCBs in the Blood of Fukuoka residents (n=152), different Age and Sex

Masuda et al., 2002
Influence of Nursing Time for Human Milk PCDD/PCDF Burden

Mean decrease per week: 0.1 pg I-TEQ/g

Source: Fürst, CLUA Münster, 1991
Daily Intake of PCDDs/Fs of Nursed Infants in Germany, 1998

Basis: 5 kg BW          800 ml/day         3 % lipids
Human milk :   13,6 pg/g TEQ

Total Intake
• in pg/day: 324
• in pg/kg BW/day: 65
PCDDs/PCDFs in Human Blood
Comparison of nursed and not nursed infant
Values in pg/g, lipid based

Päpke, 1998
Early Investigations in Vietnam
Vietnam, Background

• **Agent Orange**, a mixture of
  - 2,4,5-T (2,4,5-trichloro-phenoxy-acetic acid)
  - 2,4-D (2,4-dichloro-phenoxy-acetic acid)

was sprayed as defoliant on about

• **10% of southern Vietnam** during the Vietnam war between **1962-1971**

  2,4,5-T was contaminated with
  2,3,7,8- TCDD in the low ppm-range.
Previous studies have documented intake of TCDD in among US war veterans and in Vietnamese 

Samples collected between 1970 and 1973 documented elevated levels for TCDD in milk samples from southern Vietnamese woman as well as in fish and shrimp samples from areas in this region.

Baughman et al., 1973
2,3,7,8-TCDD Levels in Fish and Shrimp Samples from Southern Vietnam, collected 1970, pg/g, wet weight

Baughman R.W., Messelson M., 1973
2,3,7,8-TCDD in 18 Human Milk Samples from South Vietnam

Values in pg/g, lipid based, Baughman et al, 1974
Further Investigations: 2,3,7,8-TCDD in Adipose Tissue
Schecter, Ryan, Constable, 1986, Values in pg/g lipid based

n = 15
positives = 12

n = 3
positives = 0
LOD = 4-10

South of Vietnam

North of Vietnam
Further Investigations: 2,3,7,8-TCDD in Human Milk
Schecter, Ryan, Constable, 1987, Values in pg/g lipid based

Collected in 1973
- South Vietnam: n=9, pos.=6, LOD=2-5

Collected in 1985
- USA, Controls: n=3, pos.=0, LOD=2-5
- Ho-Chi Minh City: n=7, pos.=4, LOD=23-50
- Hanoi: n=2, pos.=0, LOD=23-50
- Dong-Nai River: n=2, pos.=0, LOD=23-50
Further Investigations: PCDDs/Fs in Human Milk from Vietnam and other Countries

Schecter, Fürst et al. 1991, Values in pg/g, lipid based

![Graph showing PCDDs/Fs levels in human milk from various countries.](chart.png)
2,3,7,8-TCDD in Human milk
Original data from Baughman et al., 1973

Concentration measured originally

- Samples collected: 1970
- Values, pg/g lipid:
  - (a) 1832
  - (b) 333
  - (c) 100

Concentration estimated in 2005

- Half life for TCDD: 7 years
- Time between Coll./Estimate: 35 years
- No of elimination cycles: ~ 5
- Resulting values, pg/g lipid:
  - (a) ~ 60
  - (b) ~ 10
  - (c) ~ 3
Examples for HOT SPOTS (Local Reservoirs) in Vietnam
## Recent Investigations for Human Exposure in Vietnam

<table>
<thead>
<tr>
<th>Origin</th>
<th>No of samples (n)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hanoi (comparison)</td>
<td>6 (inclusively pool of n = 100)</td>
</tr>
<tr>
<td>Biên Hòa</td>
<td>43</td>
</tr>
<tr>
<td>Binh My</td>
<td>12</td>
</tr>
<tr>
<td>Tra Noc</td>
<td>12</td>
</tr>
<tr>
<td>An Tay Commune, Ben Cat District, Binh Duong Province</td>
<td>20</td>
</tr>
<tr>
<td>Phu My Hung Commune, Cu Chi District, Ho Chi Minh City</td>
<td>20</td>
</tr>
<tr>
<td>Trương Đồng Commune, Hoa Thanh District, Tây Ninh Province</td>
<td>20</td>
</tr>
<tr>
<td>Can Gio</td>
<td>20</td>
</tr>
<tr>
<td>Phu My (Dien Khan – Nha Trang)</td>
<td>20</td>
</tr>
<tr>
<td>Phu Cat</td>
<td>20</td>
</tr>
</tbody>
</table>
b) Area of Biên Hòa
(area of former air base)
Samples from Area of Former Air Base Biên Hòa

Time of collection: 1999 – 2002

Type, number and origin of samples:

- Soil: 3 Biên Hòa
- Sediment: 11 10 Biên Hòa, 1 Hanoi
- Human Blood: 49 43 Biên Hòa, 6 Hanoi
- Food: 16 Biên Hòa, Biên Hung

Publications:

- Blood samples: JOEM, 2002
- Food samples: JOEM, 2003
Levels of 2,3,7,8-TCDD and TEQ in Soil Samples collected from Bin Hoa Air Base, a Former Agent Orange Storage Facility

Concentration in ng/kg d.m. (Schecter et al. 2001)

<table>
<thead>
<tr>
<th></th>
<th>Sample 1</th>
<th>Sample 2</th>
<th>Sample 3</th>
</tr>
</thead>
<tbody>
<tr>
<td>2,3,7,8-TCDD</td>
<td>1,164,699</td>
<td>603,968</td>
<td>N.D.</td>
</tr>
<tr>
<td>Total PCDDs</td>
<td>1,645,017</td>
<td>714,740</td>
<td>39.4</td>
</tr>
<tr>
<td>Total PCDFs</td>
<td>146,721</td>
<td>38,852</td>
<td>N.D.</td>
</tr>
<tr>
<td>TEQ (PCDDs/PCDFs)</td>
<td>1,180,737</td>
<td>610,874</td>
<td>0.04</td>
</tr>
</tbody>
</table>
PCDDs/PCDFs in Sediment, Vietnam
Values in ng/kg, dry matter

Schecter et al., 2004
* = born after 1980 (end of Agent Orange spraying: 1971)
** = pool sample, n = 100

Schecter et al., 2004
TEQ in Human Blood from Agent Orange Sprayed Areas - Bien Hoa -

- TEQ in pg/g, lipid base
- Mean n= 5
- Pool n= 100

Hanoi
Bien Hoa
PCDD/Fs in selected Population,
Finland: Kivivanta et al., 2000 -- Vietnam: Schecter et al., 2002

2.3.7.8-TCDD
I-TEQ

Finland Fishermen
n=26
Finland Controls
n=45
Germany Background
n=44
Vietnam, Bin Hoa,
min, n= 43
Vietnam, Bin Hoa,
mean, n=43
Vietnam, Bin Hoa,
max, n=43

pg/g, lipid based
TCDD Blood Levels in Vietnam, Bin Hoa, 1999
Values in pg/g, lipid based

- Family 1 (had high level of fish consumption):
  - Father: 164 pg/g TEQ, lipid based
  - Mother: 271 pg/g TEQ, lipid based
  - Child 1: 87 pg/g TEQ, lipid based

- Family 2 (moved from North Vietnam after the spraying in southern Vietnam ended):
  - Father: 74 pg/g TEQ, lipid based
  - Mother: 68 pg/g TEQ, lipid based
  - Child 1: 62 pg/g TEQ, lipid based
  - Child 2: 67 pg/g TEQ, lipid based
PCDDs/Fs in Selected Vietnamese Food

TEQs values in pg/g, wet weight based
(Schecter et al., 2003)

Area / Species

Bin Hung Lake and around
Bin Hung Market
Bin Hoa Market
WHO-PCBs in Vietnamese Food (WHO-TEQs)
Values in pg/g, wet weight based, Schecter et al., 2003

Area / Species

Bin Hung Lake and around
Bin Hung Market
Bin Hoa Market
Area of Tra Noc

Tra Noc
TEQ (PCDD/PCDF) in Human Blood from Agent Orange Sprayed Areas - Tra Noc -

Schecter et al., 2004
Ratio 2,3,7,8-TCDD : TEQ - Tra Noc -

![Bar chart showing the ratio of 2,3,7,8-TCDD to TEQ in % for Hanoi and Tra Noc.]
PCDD/F Pattern in Human Blood

Vietnam, Hot Spots (in ng/g, lipid based)

- Hanoi, n = 5
- Tra Noc, n = 12
- Bien Hoa, n = 25
c) Area of Binh My

Binh My
PCDD/F Pattern in Human Blood Vietnam
Hot Spots, Binh My, values in ng/g, lipid based

Schecter et al., 2004
Ratio 2,3,7,8-TCDD : TEQ - Binh My -

Ratio TCDD/TEQ in %

Hanoi

Binh My
PCDD/F Pattern in Human Blood
Vietnam, Hot Spots (in ng/g, lipid based)

- Tetra-CDD
- Penta-CDD
- Hexa-CDD
- Hepta-CDD
- Octa-CDD
- Tetra-CDF
- Penta-CDF
- Hexa-CDF
- Hepta-CDF
- Octa-CDF

Legend:
- Green: Hanoi, n = 5
- Orange: Binh My, n = 12
PCDDs/PCDFs in Exposure Material
Totals of ONLY 2,3,7,8-substituted Congeners

- Soil (I-TEQ : 2378 pg/g)
- Eggs (I-TEQ : 746 pg/g, lipid)
- Chicken (I-TEQ : 513 pg/g, lipid)

Wuthe et al, 1992
PCDD/F Pattern in Human Blood Vietnam
2 new Hot Spots, values in ng/g, lipid based

Binh My
Pattern!!!

Tra Noc
Schecter et al., 2004
Recent Investigations for Human Exposure in Vietnam

- An Tay Commune, Ben Cat District, Binh Duong Province
- Phu My Hung Commune, Cu Chi District, Ho Chi Minh City
- Truong Dong Commune, Hoa Thanh District, Tay Ninh Province

- For each of the 3 areas 20 blood samples were collected
- Data were published at Dioxin 2005 in Toronto, August 05
New findings: TEQ in Human Blood
An Tay Commune, Ben Cat District, Binh Duong Province
Values in TEQ, pg/g lipid, Schecter et al., 2004

TEQ range: 12.9 – 63.3 pg/g. TCDD range: 1 – 4.6 pg/g
New findings: TEQ in Human blood
Phu My Hung Commune, Cu Chi District, HCM City.

TEQ, pg/g lipid, Schecter et al., 2004

TEQ range: 20.5 – 53.6 pg/g. TCDD range: 2.3 – 13.2 pg/g
New findings: TEQ in Human blood
Truong Dong Commune, Hoa Thanh District, Tay Ninh Province

TEQ, pg/g lipid, Schecter et al., 2004

TEQ range: 7.7 – 39 pg/g. TCDD range: 0.5 – 3.3 pg/g
New Findings: TCDD in Human blood
Concentration in pg/g lipid, Schecter et al., 2004

An Tay
1 – 4.6 pg/g

Phu My
2.3 - 13.2 pg/g

Truong Dong
0.5 - 3.3 pg/g
Recent Investigations for Human Exposure in Vietnam

- Can Gio
- *Phu My (Dien Khan – Nha Trang)*
- Phu Cat

  - For each of the 3 areas 20 blood samples were collected
  - Only TCDD was analyzed
  - Data were published at Dioxin 2005 in Toronto, August 05
New Findings: TCDD in Human Blood
Concentration in pg/g lipid, Schecter et al., 2005

Can Gio
- Range: 1.7 - 13
- Median: 5.45

Dien Khan-Nha Trang
- Range: nd(1) – 3.3
- Median: 1.5

Phu Cat
- Range: nd(1) – 9.7
- Median: 2.8
Conclusions - 1

• The daily consumption of low-level contaminated food leads to the accumulation of PCDD/Fs and dioxin like PCBs in human lipids.
• The dominant source of dioxins and dioxin like PCBs for humans is food.
• The daily dioxin intake of breast fed infants is – for the nursing period – 50 -fold higher than in adults.
Conclusions - 2

- High consumption of contaminated food may result in a distinct difference in PCDD/F and dioxin like PCB levels compared to normal consumption habits.
- The dioxin concentrations – based on lipids – in human adipose tissue, blood and milk are quite similar.
- Dioxin background levels in humans living in industrialized countries, show a distinct decline over a period of about 15 years.
Conclusions - 3

• In Vietnam a number of local reservoirs (hot spots) could be identified

• It can not be excluded that these hot spots still have a permanent influence on the contamination locally produced food resulting finally in a exposure of residents

• Despite declining PCDD/F trend, the exposure of babies during the breast feeding period is still matter of concern and justifies taking measures to reduce dioxin emission into the air. This is highly relevant for nursed infants living in hot spot areas of Vietnam.
Outlook

- For further information on the pathway of human exposure in HOT SPOT areas it is recommended to analyze local food and environmental samples.
- For source identification (other than Agent Orange) further investigations are recommended for the Bin My area.
- For a better estimation of background contamination in Hanoi (controls) a larger number of samples needs to be analyzed.
Thank you for your attention