The First Screening of Human Pharmaceuticals in Drinking Water in the Czech Republic

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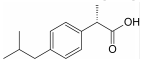
INTRODUCTION

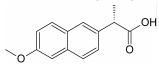
Increasing consumption of pharmaceuticals means increasing excretion and disposal and it leads to higher occurrence in surface waters. Traces may appear even in drinking water under certain conditions. Misinterpretation and exaggeration of existing knowledge from abroad by Czech media led to considerable concerns of public. Although, due to structure of water supplies in the Czech Republic (CR), situation was not considered as serious, it was decided to support communication to public with data from national monitoring. Therefore the first screening of human pharmaceuticals was completed within national research project "Occurrence of Residues of Human Pharmaceuticals in Drinking Water and their Health Risks" in 2009-2011.

IBUPROFEN (IBU)



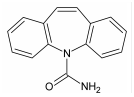
nonsteroidal antiinflammatory and antirheumatic agent

NAPROXEN (NAP)



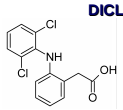
nonsteroidal antiinflammatory and antirheumatic agent

CARBAMAZEPINE (CARB)



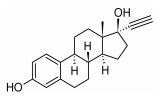
anticonvulsant

DICLOFENAC (DICL)



nonsteroidal antiinflammatory and antirheumatic agent

17α-ETHINYLESTRADIOL (EE2)



hormonal contraceptive

Figure 1.

METHOD

Substances. Five substances have been selected for screening: naproxen, ibuprofen, diclofenac, carbamazepine and 17α-ethinylestradiol. Their characteristics are given in **Figure 1**. Their occurrence was considered as the most probable due to findings abroad and structure of drug consumption in the CR. EE2 has been detected rarely in drinking water so far, but evoked the highest concern of public.

Analysis. The samples (2 x 2 litres), taken in specially treated (silanization) glass vessels and conserved with sodium azide, were concentrated with SPE method and derivatized with silylation reagent. Final analysis was done by GC/MS method [1]. Limit of quantification (LoQ) was 0.5 ng/l (but 2 ng/l for EE2 in the 2nd and 3rd sampling phases).

Sampling was completed in 3 phases. The 1st phase was representative screening involving all regions and all main supplies considering proportion of ground and surface water sources (see **Figure 2**). In total 92 different supplies were sampled (treated water from the tap within the network), 65 supplies using surface water or mixture of surface and ground water, 27 supplies using ground water only.

The 2nd phase was focused on critical localities – water treatment plants (WTP) abstracting surface raw water (quality categories A2 or A3) directly from middle or lower reaches of rivers or from reservoirs situated there (20 WTPs) or through bank filtration/artificial infiltration on lower reaches of rivers (3 WTPs). See **Figure 3**. In total 24 samples of treated water were taken on the outlet from WTPs.

The 3rd phase was thought as verification of higher results from the 2nd phase with repeating sampling. Samples (15 from 8 different supplies) were taken both on the outlet from WTPs and in the networks.

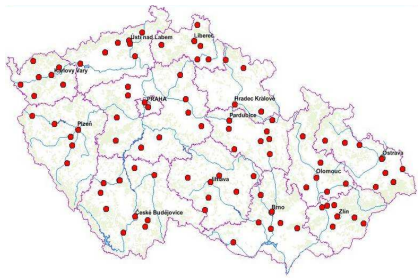


Figure 2. Sampling points in the first phase (August – November 2010)

RESULTS

All results are summarized in the **Tables 1 - 3**.

No positive findings in any sample (all values found below LoQ < 0.5 ng/l) was found in drinking water from the tap in the 1st phase.

In critical localities, only at 4 WTPs all values were below LoQ, Treated water from other ones (19 WTPs) contained 1 to 3 drugs above LoQ. IBU was found most frequently (12 x) in concentrations from 0.7 to 20.7 ng/l (median 2.0 ng/l), followed by CARB (9 x) from 2.2 to 18.5 ng/l (median 5.5 ng/l).

Results obtained in the 3rd phase was mostly lower than previously. Regarding samples taken from the taps, only in 3 cases any result was above LoQ: IBU 3 x (0.5 – 1.2 ng/l) and CARB 1 x (4.0 ng/l).

Concentrations of EE2 were in all cases below LoQ. Even the LoQ was quite low, probably it was not low enough to detect theoretical traces [2].

Using any methods for health risk assessment [3], when e.g. margins of exposures[1] ranged from 10^6 to 10^8 (but 3750 for EE2 if occurrence on LoQ level was thought), there is no known health risk from this exposure. We can illustrate this minute exposure to public on the example of ibuprofen: even using rare maximum (20 ng/l) found by us, one should drink such water (2 l/day) for 26,000 years to receive the dose equivalent to one pill (400 mg) as minimum therapeutic dose which is not perceived as harmful by Czech public.

[1] MOE calculated as proportion of minimum daily therapeutic dose and daily exposure of drug from drinking water (as worst case scenario).

Figure 3. Sampling points in the second phase (May – July 2011)

Tables 1 - 3. Number of findings in defined concentration ranges, minimums, maximums and medians

1st phase	IBU	NAP	CARB	DICL	EE2
< 0.5[a] ng/l	92	92	92	92	92
0.5 – 3 ng/l	0	0	0	0	0
3 – 10 ng/l	0	0	0	0	0
> 10 ng/l	0	0	0	0	0
minimum[b]	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5
maximum[b]	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5
median[b]	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5

2nd phase	IBU	NAP	CARB	DICL	EE2
< 0.5[a] ng/l	12	19	15	22	24
0.5 – 3 ng/l	8	4	2	1	0
3 – 10 ng/l	2	1	6	1	0
> 10 ng/l	2	0	1	0	0
minimum[b]	0.7	0.5	2.2	0.6	< 2.0
maximum[b]	20.7	3.0	18.5	3.9	< 2.0
median[b]	2.0	2.2	5.5	2.25	< 2.0

3rd phase	IBU	NAP	CARB	DICL	EE2
< 0.5[a] ng/l	8	12	12	14	15
0.5 – 3 ng/l	6	2	1	1	0
3 – 10 ng/l	1	1	1	0	0
> 10 ng/l	0	0	1	0	0
minimum[b]	0.5	0.8	1.4	1.9	< 2.0
maximum[b]	3.6	3.0	13.6	1.9	< 2.0
median[b]	1.1	1.2	4.0	1.9	< 2.0

[a] for EE2 in the 2nd and 3rd phase was LoQ 2.0 ng/l

[b] from values above LoQ

CONCLUSION

Situation regarding the occurrence of pharmaceuticals in Czech drinking water supplies seems to be favourable as we found selected substances – which may be thought as kind of indicators considering their high probability to be found – in the samples taken from consumer´s taps only very rarely (3 samples of more than 100) and in very low concentrations (IBU up to 1.2 ng/l; CARB up to 4.0 ng/l). Although there is no known health risk from such exposure, it seems to be worthwhile to inform public about the situation and educate them on responsible use and proper disposal of unused pharmaceuticals because of known risk for environment.

Favourable situation is mainly given by structure of water supply: about 49 % water is produced from groundwater sources and about 51 % from surface water sources. However, surface water is mostly (~ 80 %) taken from protected reservoirs with no or very low wastewater load, built on upper reaches of rivers; the rest is direct intake from rivers, ponds, unprotected reservoirs.

References

[1] Yu Z., Peldszus S., Huck P.M. *Journal of Chromatography A* 2007; 1148: 65-77.

[2] Hannah R., D'Aco V.J., Anderson P.D. et al. *Environmental Toxicology and Chemistry* 2009; 28: 2725–2732.

[3] Pharmaceuticals in Drinking water. WHO/HSE/WSH/11.05. WHO, Geneva 2011.

Acknowledgement

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