Science of the Total Environment 493 (2014) 432–444

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Assessment of spatial variation in drinking water iodine and its implications for dietary intake: A new conceptual model for Denmark

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HIGHLIGHTS

- Nationwide study on iodine concentration and speciation in treated drinking water
- Complex spatial pattern of both the iodine concentration and speciation
- Spatial variance governed by both geology and treatment procedures
- Proposed conceptual model of geographical differences in drinking water iodine
- Estimated contribution of drinking water to the dietary iodine intake

ARTICLE INFO

Article history:
Received 3 April 2014
Received in revised form 26 May 2014
Accepted 2 June 2014
Available online xxxx

Editor: D. Barcelo

Keywords:
Iodine
Iodate
Iodide
Local Moran's I
Dietary iodine intake

ABSTRACT

Iodine is essential for human health. Many countries have therefore introduced universal salt iodising (USI) programmes to ensure adequate intake for the populations. However, little attention has been paid to subnational differences in iodine intake from drinking water caused by naturally occurring spatial variations. To address this issue, we here present the results of a Danish nationwide study of spatial trends of iodine in drinking water and the relevance of these trends for human dietary iodine intake.

The data consist of treated drinking water samples from 144 waterworks, representing approx. 45% of the groundwater abstraction for drinking water supply in Denmark. The samples were analysed for iodide, iodate, total iodine (TI) and other major and trace elements. The spatial patterns were investigated with Local Moran's I. TI ranges from <0.2 to 126 $\mu g\ L^{-1}$ (mean 14.4 $\mu g\ L^{-1}$, median 11.9 $\mu g\ L^{-1}$). Six speciation combinations were found. Half of the samples ($n = 71$) contain organic iodine; all species were detected in approx. 27% of all samples. The complex spatial variation is attributed both to the geology and the groundwater treatment. TI >40 $\mu g\ L^{-1}$ originates from postglacial marine and glacial meltwater sand and from Campanian–Maastrichtian chalk aquifers. The estimated drinking water contribution to human intake varies from 0% to >100% of the WHO recommended daily iodine intake for adults and from 0% to approx. 50% for adolescents. The paper presents a new conceptual model based on the observed clustering of high or low drinking-water iodine concentrations, delimiting zones with potentially deficient, excessive or optimal iodine status.

Our findings suggest that the present coarse-scale nationwide programme for monitoring the population's iodine status may not offer a sufficiently accurate picture. Local variations in drinking-water iodine should be mapped and incorporated into future adjustment of the monitoring and/or the USI programmes.

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1. Introduction

Iodine plays an essential role in human metabolism and the early development of most organs, including the brain (WHO, 2007). Insufficient or excessive iodine intake can both cause health problems. Excess ingestion of iodine (iodine excess, IE) may increase the prevalence of thyroid enlargement and goitre, subclinical hypo- and hyperthyroidism,

Abbreviations: RNI, recommended daily nutrient intake in this paper the nutrient is iodine; TI, total iodine; DOI, dissolved organic iodine; DWQC, drinking water quality criteria; IDD, iodine deficiency disorder; ID, iodine deficiency/deficient; IE, iodine excess/excessive; WHO, World Health Organisation; UI, urinary iodine; USI, universal salt iodising; LISA, local indicators for spatial association.

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http://dx.doi.org/10.1016/j.scitotenv.2014.06.008
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iodine-induced hyperthyroidism and thyroiditis (WHO, 2007), and it may give rise to autoimmune thyroid diseases, iodine allergies and poisoning, and loss of intelligence (Shen et al., 2011). The WHO (2007) grouped the wide spectrum of iodine deficiency disorders (IDD) by age: foetus—abortions, stillbirths, birth defects, perinatal mortality, endemic cretinism; neonate—hypothyroidism, endemic mental retardation; child and adolescent—goitre, hypo- and hyperthyroidism, impaired mental function, slower physical development; adult—goitre with its complications, hypo- and hyperthyroidism, and impaired mental function. Furthermore, an increased vulnerability of the thyroid gland to nuclear radiation irrespective of age is another characteristic of iodine deficiency (ID) (WHO, 2007). Even mild iodine deficiency could result in learning disabilities, poor growth and diffuse goitre in school children (Soldin, 2009). Worldwide, the focus falls mainly on the ID, as only 10 countries are classified as excessive iodine intake countries (Pearce et al., 2013). ID is not confined to developing countries (Zimmermann, 2011); it is estimated that iodine intake is insufficient in 43.9% ($n = 30.5$ million) of 6–12-year-old children and 44.2% ($n = 393.1$ millions) of the general population in the WHO European Region (Zimmermann and Andersson, 2012).

Denmark (Fig. 1) is one of the countries classified to have a mild ID status (epidemiological criteria: median urinary iodine (UI) excretion 50–99 μg L^{-1} (WHO, 2007)). However, this classification is not based on a nationwide UI survey. Universal salt iodising programmes (USI) are recognised as the most efficient method for ID prevention. In Denmark, such a programme was recommended in 1996 (Rasmussen et al., 1996). However, Denmark was considered neither to be deficient, nor to have endemic goitre; rather, the country was described as an area with "marginal iodine deficiency" (Rasmussen et al., 1996). A voluntary USI programme therefore started in 1998 (Laurberg et al., 2006; Rasmussen et al., 2002) aiming at increasing the daily intake by 50 μg I (Laurberg et al., 2006). Two years later, the programme was found insufficient, and mandatory iodine addition to salt for household consumption and for cake and bread production was therefore introduced in April 2001 (Laurberg et al., 2006).

Drinking water is not normally regarded a major contributor to the average dietary iodine intake, providing in general 10% only (Fuge, 2005). Still, in Denmark where drinking water is of groundwater origin, on average 25% (Rasmussen et al., 2000) or 24% (Rasmussen et al., 2002) of the overall iodine intake before 2001 was derived from drinking water and other beverages (w/o milk and juice). After implementing the USI programme, this percentage fell to 14% (Pedersen et al., 2010).

Regional variations in iodine content in the Danish drinking water were first reported in 1999 (55 tap water samples; iodine variation <1 - 139 μg L^{-1} (Pedersen et al., 1999)). These variations were later confirmed in a more representative study based on drinking water samples taken at waterworks ($n = 22$) (Andersen et al., 2002)). Based on these two studies, a difference between East (Zealand) and West Denmark (Jutland and Funen), with respect to iodine content in drinking water has been postulated (Fig. 1) (Andersen and Lauberg, 2009).

Two cohorts have been chosen to follow up on the effects of the Danish USI programme (DanThyr) (Laurberg et al., 2006): the Aalborg cohort representing Western Denmark (lower iodine intake) and Copenhagen representing Eastern Denmark (higher iodine intake) (see Fig. 1B). Nevertheless, no detailed studies of the spatial patterns of iodine in drinking water in Denmark have been published. Thus, a main purpose of the present study is to investigate the spatial trends that may have been missed due to the low number of locations included in the previous studies (e.g. Andersen et al. (2002)).

The last nationwide study of iodine status of the Danish population was conducted by Munkner in the late 60s (Munkner, 1969) who collected 24-hour UI from about 6000 young men from all over the country. Pedersen et al. (1999) compared these data to the data from tap water samples they collected in 1999. They found a correlation between the two datasets ($r = 0.68$, $P < 0.001$; regression line $y = 1.7x + 43.2$). However, drinking-water iodine has not been monitored for spatial and/or temporal variations as iodine is not part of the monitoring programmes of the Danish waterworks and a water quality criterion has not been set. By performing detailed high-resolution national studies of the spatial patterns of iodine concentration (and speciation), potentially ID/IE geographical areas can be identified for further epidemiological studies or for adjustment of the current ID prevention measures. For example, in China where USI is used as an IDD prevention measure, a national criterion for classifying high-iodine regions has been adopted by using median water iodine (150–300 μg L^{-1}—likely IE area; >300 μg L^{-1} high iodine endemic area) (Lv et al., 2013), and an association between drinking water and IE has been found (Lv et al., 2013; Shen et al., 2011). A second purpose of the present study was therefore to provide updated data on the spatial patterns of iodine concentration in drinking water for the entire country and on that basis to identify areas with different iodine status.

Iodine speciation is another largely ignored, but important issue in the discussion of the bioavailability of iodine. In the hydrogeochemical cycle, iodine predominantly appears in the inorganic forms of iodide (I^-)

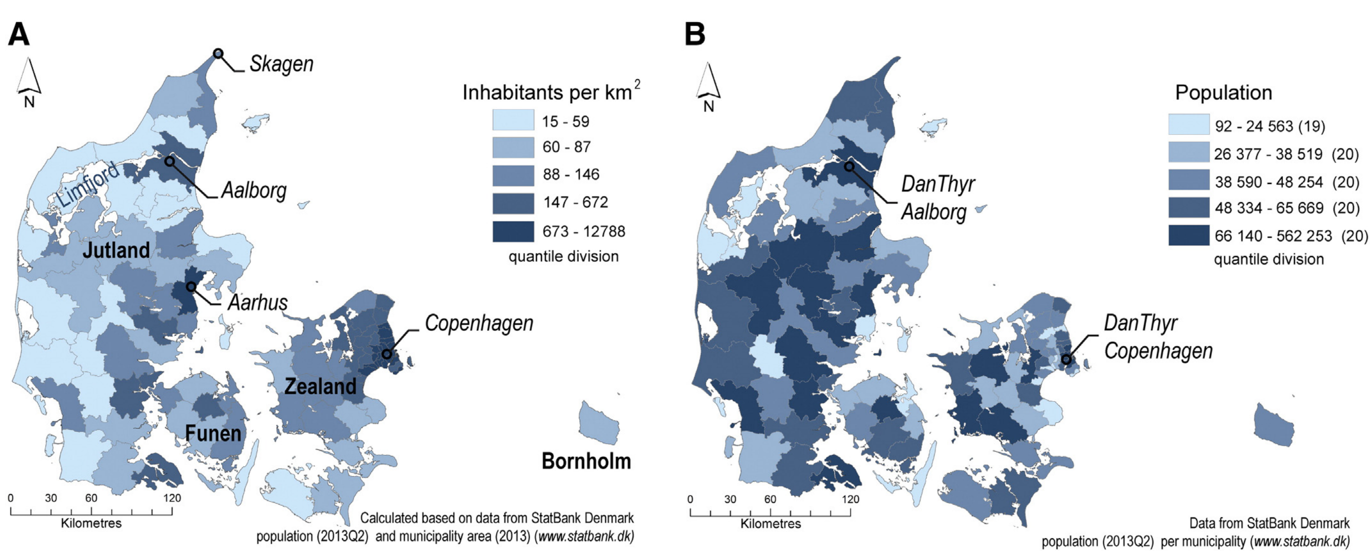

Fig. 1. Population density (A) and population distribution (B) over the 99 Danish municipalities (based on 2013 data); the number of municipalities included in each group for (B) is given in the legend too e.g. (19), and the two DanThyr cohorts are also shown; see *Supplementary data* for further details.

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