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Temporal variation of iodine in Danish groundwater

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Abstract

Iodine is an essential element for human health, and both high and low iodine intake could have negative health outcomes. The spatial variation of iodine in Danish groundwater has been studied before, but to the author's knowledge, this is the first time that the temporal variation is characterised. Nationwide data from the Danish groundwater monitoring programme (GRUMO) were analysed between 2011 and 2021, including 2924 samples from 1242 well screens at 893 wells. The sampling frequency varied and so the robust coefficient of variation (*rCV*) was calculated for 930 (75%) of well screens, and time-series analysis was performed for 23 (2%). Key findings are (1) iodine in Danish groundwater varies over time (0–124%, median = 10%), (2) in one quarter of the well screens *rCV* exceeds 20% and (3) this variation cannot be attributed solely to analytical uncertainty at 14% of the well screens. The impact of temporal variation of iodine in Danish drinking water of groundwater origin should be evaluated in future exposure or epidemiological studies with respect to the study goal, location and time period. Since the temporal variation could not be quantified over the entire concentration range, monitoring of iodine in Danish groundwater should continue.

Introduction

Iodine is an essential element for the proper functioning of the thyroid, and either high or low iodine intake could result in adverse human health outcomes, given that there is a U-shaped relationship between iodine intake and thyroid disorders (Laurberg *et al.* 2009) or autoimmunity (Wang *et al.* 2019). Iodine deficiency disorders (IDD) are a major public health problem globally (De Benoist *et al*. 2004), and even though levels of global iodine nutrition have improved since the 20th century, Andersson *et al.* (2012) estimated that 1.88 billion people, including 241 million school children, still had insufficient dietary iodine intakes. Iodine deficiency has been described as "the greatest cause of preventable brain damage in childhood" (De Benoist *et al*. 2004). Iodine deficiency could also cause miscarriages or stillbirths (at the foetal stage); neonatal goitre or hypothyroidism, endemic mental retardation (at the neonatal stage); goitre, hypo- or hyperthyroidism, impaired mental function or retarded physical development (in children, adolescents and adults; De Benoist *et al*. 2004). Excessive iodine intakes, on the other hand, may result in hypo- or hyperthyroidism, goitre and/or thyroid autoimmunity for some individuals (Farebrother *et al.* 2019).

The recommended daily intake and the tolerable upper iodine intake vary with age (Table 1; Institute of Medicine 2001; WHO & FAOUN 2004; EFSA 2006; WHO 2007).

Drinking water could be a significant contributor to the daily iodine intake for some populations (Voutchkova *et al.* 2014; Farebrother *et al.* 2019; Ma *et al.* 2022). Recently, a meta-analysis by Azevedo *et al.* (2023) showed that iodine status is directly correlated to iodine content in drinking water and concluded that iodine concentration in drinking water can be used as an indicator of dietary intake. Therefore, the spatial variation of iodine in both drinking

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Abbreviations
b.g.l.: below ground level
GAM: generalised additive model
GEUS: Geological Survey of Denmark and Greenland
GRUMO: Danish national groundwater monitoring programme
ICP-MS: inductively coupled plasma mass spectrometry
IDD: Iodine deficiency disorders
IQR: interquartile range
LOD: limit of detection
LOESS: local polynomial regression fit
MAD: median absolute deviation
rCV: robust coefficient of variation
U: analytical uncertainty

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Table 1 Recommended daily intake (RDI) and tolerable upper intake (TUI) for iodine.

Age	RDI[a] (µg/day)	TUI (USA)[b] (µg/day)	TUI (EU)[c] (µg/day)
Children	120[1]	300–900[2]	200–500[3]
Adults	150	1100	600
Pregnant or lactating women	250	1100	600

For children, the provided values and ranges are for: [1]school children; [2]children in the age 4–18 years; [3]children in the age 1–17 years. References are as follows: [a]World Health Organization 2007; World Health Organization & Food and Agriculture Organization of the United Nations 2004. [b]Institute of Medicine, Academy of Sciences & USA 2001. [c]EFSA 2006.

water and groundwater and its importance for human health have been discussed in depth in the scientific literature: either with respect to the need for optimising dietary iodine intake of a given population subjected to a universal salt iodisation programme (Voutchkova *et al.* 2014; Ma *et al.* 2022) or because of the potential for the formation of unwanted iodinated by-products when implementing certain advanced methods of drinking water treatment (Sharma *et al.* 2019). However, less attention has been given to temporal aspects of iodine variation, which may be important regarding iodine exposure from drinking water.

In Denmark, drinking water is of entirely groundwater origin and undergoes mostly simple treatment (aeration and sand filtration) without chlorination or ozonation. It has been hypothesised that iodine concentrations in Danish groundwater are stable over time. This hypothesis stems from limited evidence for iodine in treated drinking water (tap water) and was mostly based on qualitative assessments, which concluded the following:

1. Day-to-day variation within a period of 10 days at two locations in Denmark was "small" (Pedersen *et al.* 1999)
2. "No significant" difference between samples collected in January and June (Rasmussen *et al.* 2000)
3. Concentration at a waterworks in the northernmost part of Denmark (Skagen) was "unaltered" in the period 1997–2000, based on one sample taken every 2 months for a duration of 6 months and one sample taken every year for a period of 4 years (Andersen *et al.* 2002)
4. No conclusive results concerning short-term variation were observed in eight samples taken in a 2-week period at a waterworks supplying Copenhagen (Voutchkova *et al.* 2014).

The purpose of this article is therefore to quantify the temporal variation of total dissolved iodine in Danish groundwater. This assessment is based on nationwide groundwater-quality monitoring data covering a 10-year period, which has not been reported and systematically analysed previously.

Data

Total dissolved iodine ('iodine' for brevity) was analysed in 2924 samples obtained during the period 2011–2021 (31 January 2011 – 11 May 2021) as part of Denmark's national groundwater monitoring programme GRUMO (Thorling *et al.* 2023). These samples were taken at 1242 sampling points (well screens) belonging to 893 GRUMO wells located throughout the country (Fig. 1). The well screens were relatively short: 1 or 2 m long (Q25 = 1 m, median = 1 m, Q75 = 2 m, min. to max.: 0.05–69 m).

Following the sampling protocols of the Geological Survey of Denmark and Greenland (GEUS), well purging was carried out until pH, conductivity, O_2 and temperature were stable (Thorling 2012). Samples were filtered in the field through 0.45-µm filters and placed in glass bottles without conservation or other sample pre-handling, after which the samples were stored in cold (0–4°C) and dark conditions for a maximum of 36 h before analysis (Ministry of Environment of Denmark 2011).

Iodine was analysed with ICP-MS (Ministry of Environment of Denmark 2011) at nationally accredited laboratories. The national requirements for analytical uncertainty of all environmental measurements for iodine are 1.5 µg/l absolute expanded uncertainty (U_{abs}) for low concentrations and 20% relative expanded uncertainty (U_{rel}) for high concentrations (Ministry of Environment of Denmark 2021). As Ministry of Environment of Denmark (2021) has not defined what is considered a low concentration, the highest of the two ($max\,(U_{rel}, U_{abs})$) is used here as the analytical uncertainty for individual samples (U). The actual values of U are unknown for this monitoring data set. For 71% of the samples (n = 2082), $U_{abs} \geq U_{rel}$, so $U = U_{abs}$. For the remaining 29% (n = 842), U_{rel} was higher, so $U = U_{rel}$.

Fewer than 1% of the samples (n = 26) were below the LOD. Two different LODs were used: 2 µg/l (10 samples) and 0.3 µg/l (16 samples). The values below LOD were handled by substitution with 0.5 × LOD, which is equal to 1 µg/l or 0.15 µg/l, respectively.

Statistical methods

The sampling frequency varied at the different well screens from 1 to 11 times during the study period (Figs 1 and 2a). The temporal variability could only be assessed for sampling locations with at least two samples (n = 930, 75%) using the robust coefficient of variation (rCV, equation 1; Arachchige *et al.* 2022).

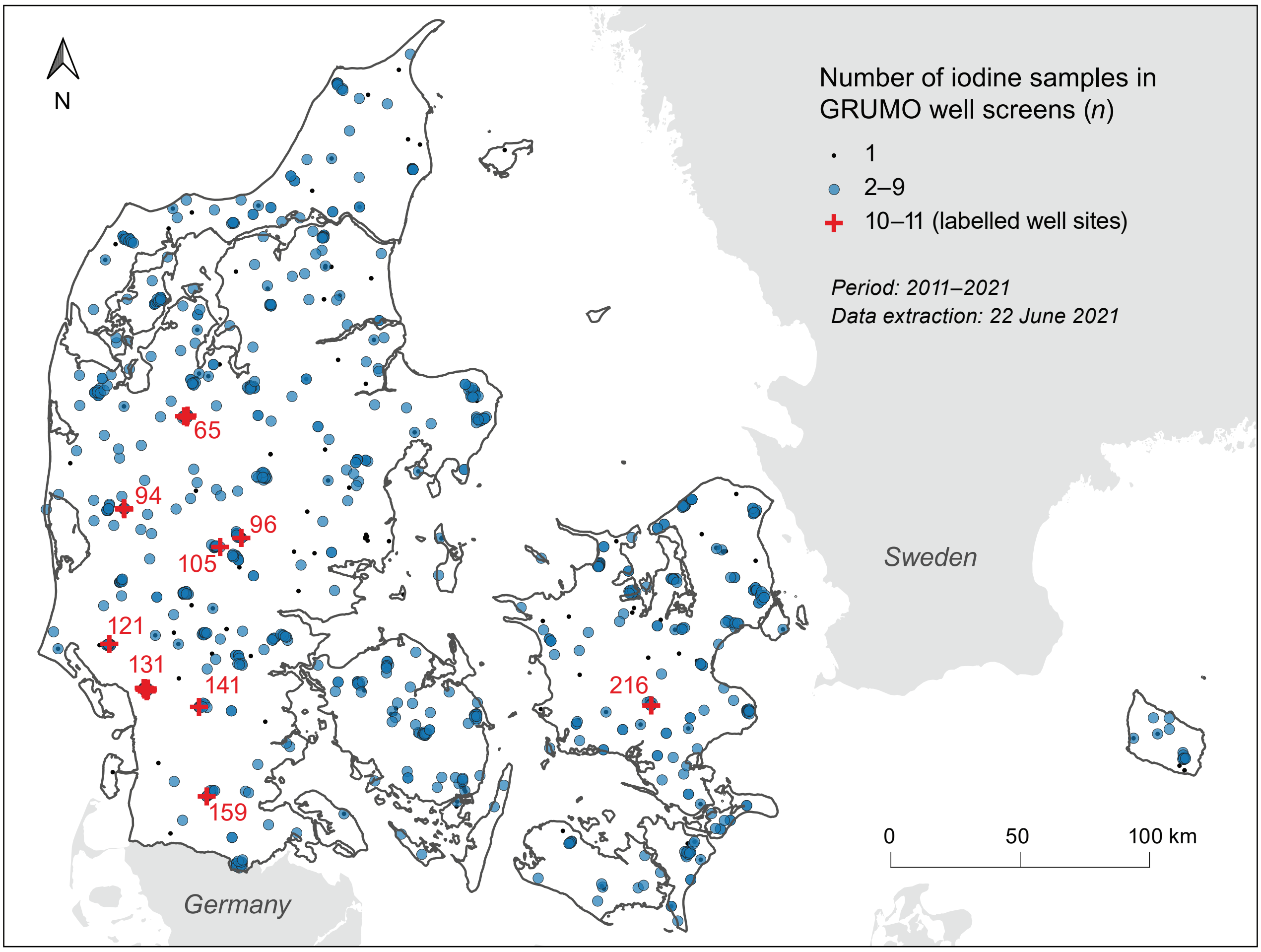

Fig. 1 Spatial distribution of iodine sampling sites in the study period at well screens of the Danish national groundwater monitoring programme (GRUMO).

The *rCV* is a measure of relative dispersion similar to the coefficient of variation but is based on the median and median absolute deviation (*MAD*) as follows:

$$rCV = 1.4826 \times MAD/m \tag{1}$$

where *m* is the median and *MAD* is a robust measure of variability, similar to the standard deviation but using the median instead. *MAD* (equation 2) is defined as the median of the absolute deviations from the median of the data (*m*), such that:

$$MAD = \text{median}(|X_i - m|) \tag{2}$$

where X_i are the individual iodine measurements at a given sampling location.

These metrics for central tendency and variability are more robust against outliers or skewed distributions and so were preferred for this study.

Next, *rCV* was compared against the analytical uncertainty across Denmark to provide an indication as to whether the observed iodine variability might be associated with environmental factors or else most probably because of the analytical uncertainty. This was done as follows:

$$\text{if} \begin{cases} rCV_j > \text{median}\left(\dfrac{U_i}{X_i} \times 100\%\right) \rightarrow \text{environmental} \\ rCV_j \leq \text{median}\left(\dfrac{U_i}{X_i} \times 100\%\right) \rightarrow \text{analytical} \end{cases} \tag{3}$$

where *i* is the index of an individual sample, *j* is the index of the well screen, *X* is iodine concentration and *U* is analytical uncertainty. Here the analytical uncertainty is converted to a percentage, so that it could be compared to *rCVj*. This comparison is only used as a screening tool to provide a preliminary estimate of the potential cause of the observed variation. A more comprehensive assessment at a local scale involving an increased number of samples, as well as additional hydrogeochemical, hydrogeological and environmental data would be needed to provide further details.

In addition, a trend analysis was performed solely for well screens with ≥10 years of data (*n* = 23; 2%). Linear regression and local polynomial regression (LOESS with a generalised additive model, GAM, with integrated smoothness estimation as implemented in Wood (2023)) and their 95% confidence intervals were used to interpret the trends (Wickham 2016). All statistical analyses and summaries were implemented in R v. 4.2.1 (R Core Team 2022).

Results and discussion

Nationwide temporal variability

The results of this nationwide assessment showed that the robust coefficient of variation (*rCV*) for iodine at the 930 sampling points in Denmark had a wide range (0–124%) with a median of 10% (IQR 4–21%; Fig. 2b). This means that at half (quarter) of the sampling locations, iodine variation in the period exceeded 10% (20%). Part of the observed variation could be due to

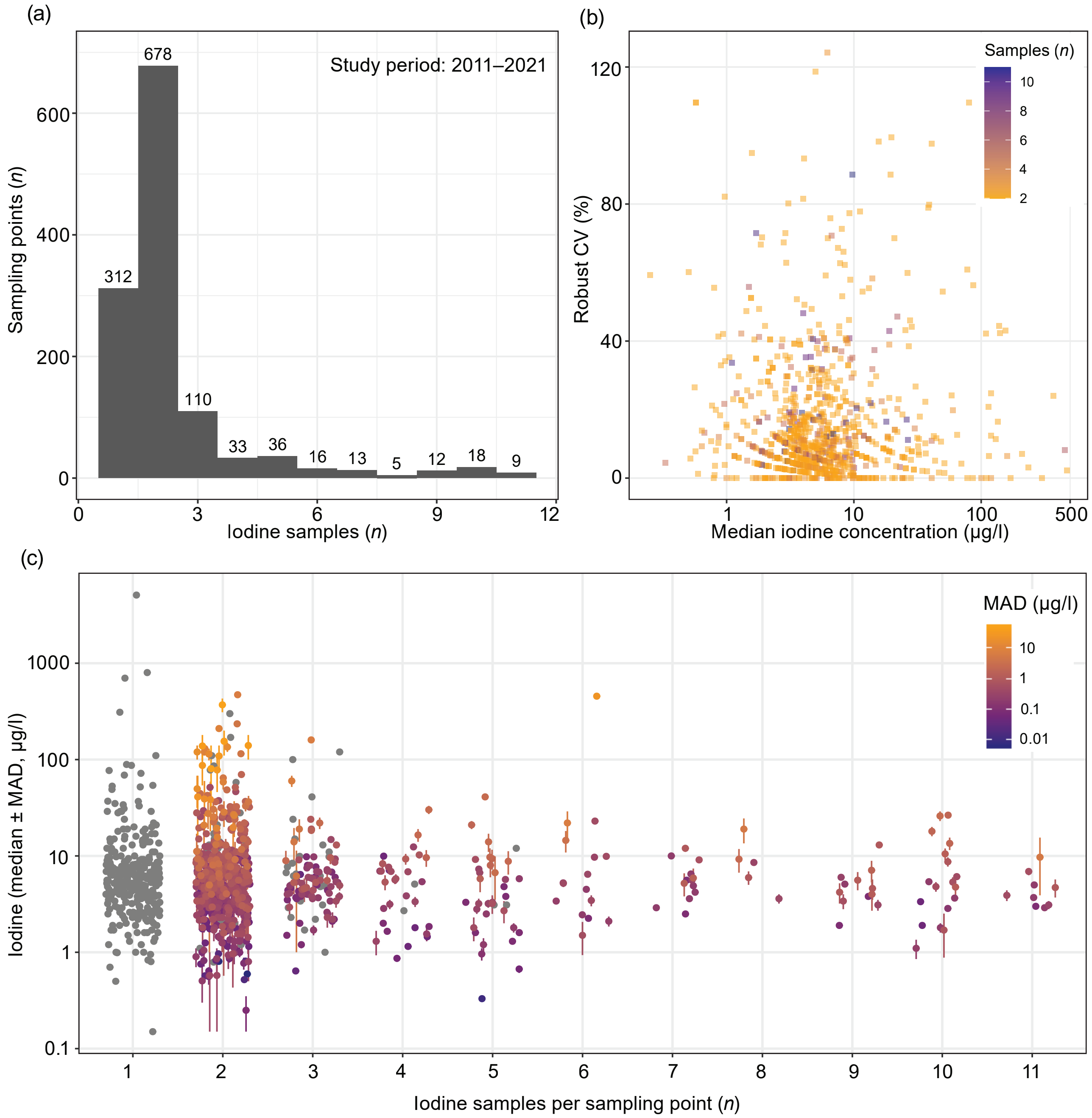

Fig. 2 Iodine in groundwater wells from the Danish national groundwater monitoring programme (GRUMO). **a**: Histogram of number of iodine samples per well screen. **b**: Scatter plot of robust coefficient of variation (*rCV*) against median iodine concentration, where each symbol refers to a well screen. **c**: iodine concentration and its variation (expressed by the median and median absolute deviation, *MAD*). Grouped according to the number of samples per sampling location, where horizontal jitter is added to minimise overlapping points. **c**: Grey colour is used when the *MAD* was not calculated (only one sample available) and if *MAD* = 0 (2–11 samples).

analytical uncertainty, and so comparison between *rCV* and *U* was used as a screening tool at the national scale. This comparison showed that at 14% of the sampling points (n = 127, Fig. 3), iodine variability could not be attributed solely to analytical uncertainty. It could be inferred that at those locations, the variability was, at least partially, caused by environmental factors such as local hydrogeological conditions and/or iodine variability in precipitation. No concentration dependency or depth dependency (not shown here) or spatial pattern (Fig. 3) was associated with the observed iodine variability at the national scale. Further, more focused investigations could elucidate the governing environmental factors at specific locations.

This assessment showed that there is a temporal variability of iodine in groundwater, but its importance in exposure and epidemiologic studies cannot be inferred purely from these data alone. As all drinking water in Denmark originates from groundwater, variability in groundwater iodine may have a significant effect on the concentration of iodine in finished (i.e. treated) drinking water, especially in parts of the country where high levels of iodine are observed. Other factors could also contribute to iodine variability in drinking water. For example, the well sites, wells and/or the pumping strategies could change in time, resulting in the use of other aquifers or parts of the aquifer where the iodine concentration is different. It is also possible that the water treatment and its performance over time affect the iodine content of the finished product. Therefore, the recommendation is to consider temporal aspects when designing future studies, so that the significance of iodine variability can be assessed with respect to the specific location of interest.

Even though the GRUMO programme has yielded plenty of new iodine data over the past decade, it is still challenging to assess variability for the full range of observed concentrations. The highest iodine concentrations (>50 µg/l) and absolute variations occur at locations with only two or very few samples (Fig. 2c), which limits analysis solely to a *rCV* to *U* comparison. At 25% of the sampling locations (n = 312), only a single sample was acquired, and the variability could therefore not be assessed at those sites. It is therefore recommended that iodine continues to be monitored as part of the GRUMO programme. It should be noted that iodine is not analysed in drinking water in Denmark because

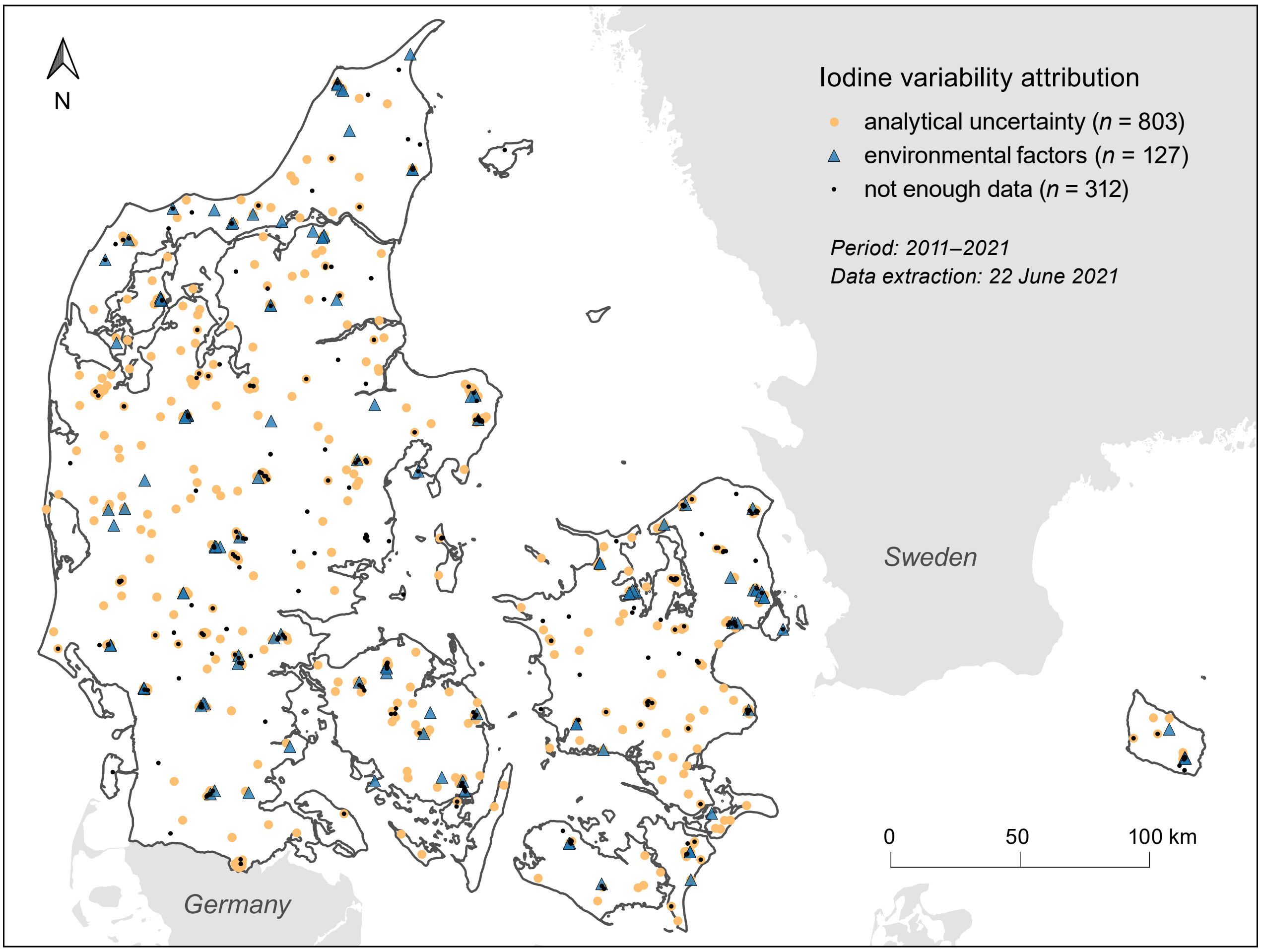

Fig. 3 Iodine variability attribution at sampling sites throughout Denmark.

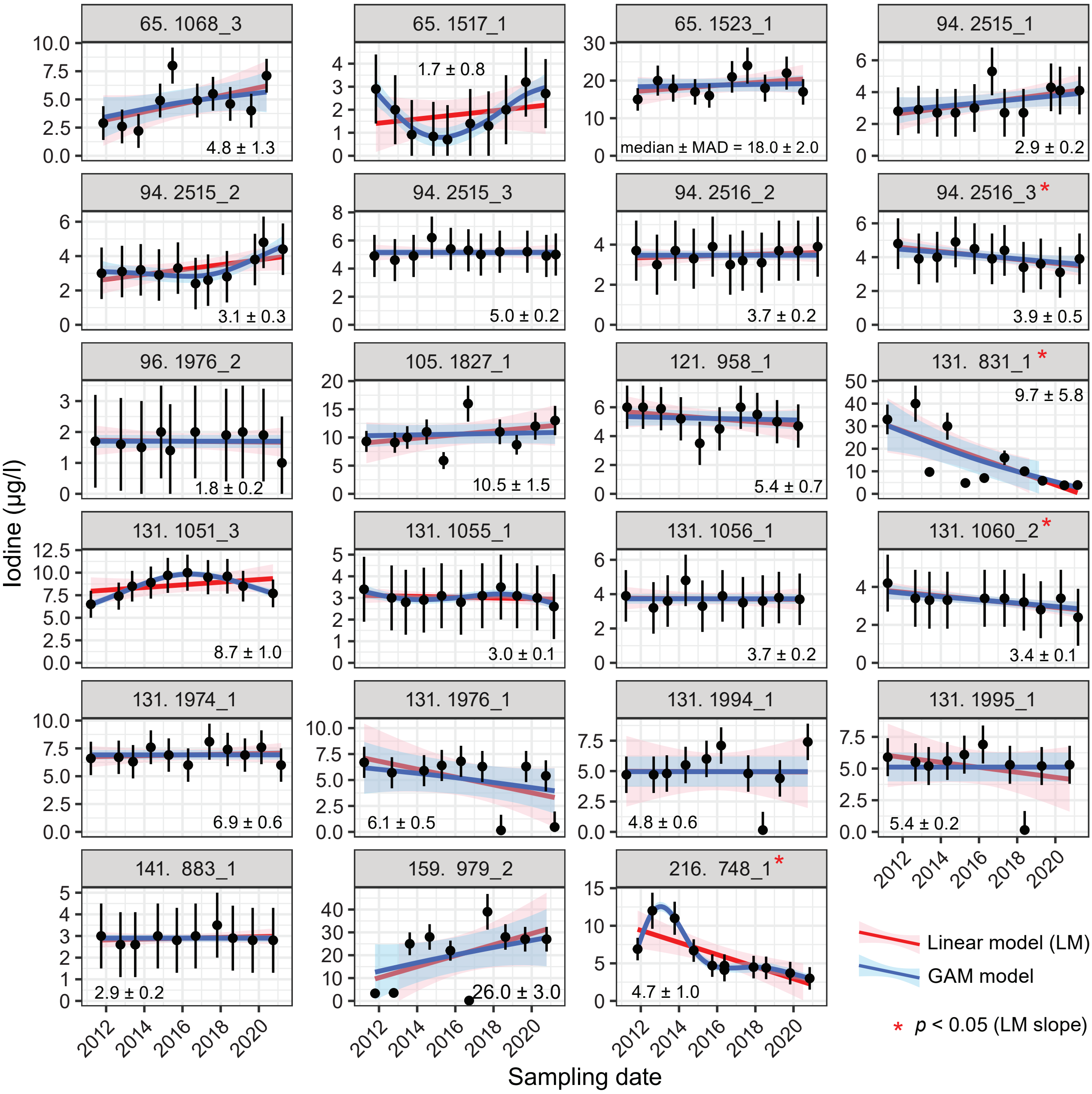

Fig. 4 Iodine time series for well screens with data of at least a 10-year duration (n = 23; Fig. 1; Table S1 in supplementary information). The error bars show the analytical uncertainty. 95% confidence intervals are displayed for both the linear and generalised additive (*GAM*) models.

there is no legal requirement to do so as part of quality control at waterworks.

Time series analysis

Time series analyses could provide further insight into the way iodine concentrations vary over time. However, there is presently insufficient data coverage to conduct time series analyses across Denmark; a time series analysis could be performed at only 23 of the well screens, which have at least 10 years of iodine data. All these well screens are relatively shallow (3–17.6 m b.g.l.) and located at nine GRUMO well sites (Fig. 1), one in East Denmark, the rest in West Denmark. These screens are in Quaternary sand aquifers except for one in a pre-Quaternary sand aquifer (named '96.1976_2'). They are associated with 10 different groundwater bodies, as defined by Troldborg (2020). The parent sediment (at 1 m depth), where known, is either proglacial (n = 4), glacial (n = 14) or postglacial (n = 1). There are mostly low iodine concentrations at these sampling points: at 18 of the well screens (78%), the median iodine concentration is ≤10 µg/l and only at three it is >20 µg/l. See Table S1 for more details.

Figure 4 presents the time series analysis for iodine at these well screens. Based on the *rCV* to *U* comparison, the variability in iodine concentration could be attributed to environmental factors at three of these wells ('65.1068_3', '105.1827_1', '131.831_1'). At '65.1068_3', there is a positive, but not statistically significant linear trend, no non-linear trend and potentially an outlier. However, the concentrations at this well are low overall at 4.8 ± 1.3 µg/l. The well screen is shallow (4.5–5.5 m b.g.l.), located in a Quaternary sand aquifer. The well screen '105.1827_1' is also shallow (3–4 m b.g.l.), without a significant trend, with a possible outlier, but the iodine concentrations over the period are a bit higher at 10.5 ± 1.5 µg/l. However, at '131.831_1', there is a significant decreasing trend, the screen is a bit deeper (14.5–20.5 µg/l), and the concentrations are higher, especially at the beginning of the monitoring period (9.7 ± 5.8 µg/l). Two other well screens have significant linear trends ('94.2516_3' and '131.1060_2'), but both have low concentrations (3.9 ± 0.5 µg/l and 3.4 ± 0.1 µg/l, respectively) and are relatively shallow (6.5–7.5 m b.g.l. and 10.5–11.5 m b.g.l.). Their variability can potentially also be attributed to analytical uncertainty. Similarly, four other well screens exhibit non-linear trends ('65.1517_1', '94.2515_2', '131.1051_3', '216.748_1'), and their variability can also be attributed to analytical uncertainty. In addition, Fig. 4 shows how the analytical uncertainty of each sample (error bars) compares to the variability, concentration level and linear/non-linear trends. The observed variability, when attributed to environmental factors, was relatively low at these well screens and so could most probably be explained by variation in the precipitation concentration and potentially by varying amounts of leaching from the soil. Unfortunately, none of the locations with higher iodine concentrations had time series that were sufficiently long, and so it is presently impossible to quantify and explain the variability of the full range of iodine concentrations found in Danish groundwater.

Conclusions

This is the first time that the temporal variation of iodine in Danish groundwater has been characterised systematically. The data spanned a decade (2011–2021), but the sampling frequency varied, according to the GRUMO programme. This nationwide assessment showed that temporal variability of groundwater iodine in Denmark exceeded 20% during 2011 to 2021 at 25% of the well screens that had at least two samples (n = 930). Based on comparison between the robust coefficient of variation and the analytical uncertainty, it was found that the observed variability cannot be attributed solely to analytical uncertainty for 14% of the well screens. Although there were insufficient data to undertake a trend analysis at a national scale, the time series (n = 23) indicated that there could also be statistically significant linear and non-linear trends in iodine concentration at some locations. Unfortunately, the time series did not include sampling locations with high iodine concentrations. Moreover, the calculation of robust coefficient of variation was also limited for well screens with iodine >50 µg/l. For a quarter of the GRUMO well screens, there was only one sample available in the 2011–2021 period, so the variability could not be assessed at all. It is therefore recommended that the temporal variation analysis be repeated when more data have been collected as part of the GRUMO programme. In short, iodine in Danish groundwater varies temporally; thus it may be important to evaluate this aspect in future exposure or epidemiological studies focusing on iodine in drinking water.

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Author contributions

DV is the sole author.

Competing interests

No competing interests.

Additional files

Three supplementary files are available with this manuscript at *https://doi.org/10.22008/FK2/ELLKNC*

1. Aggregated_data.xlsx – Data set, aggregated at well-screen-level, as used in this study.
2. Table_S1.docx – Characteristics of the well screens included in the time-series analysis
3. READ_ME.docx: A short description of the data set parameters in 'Aggregated_data.xlsx'.

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