Creating a map of stable isotopes in tap water across South Africa for hydrological, ecological and forensic applications

Report to the
Water Research Commission

by

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Summary

We present a map of the stable H- and O-isotope ratios of tap water samples from across South Africa. This map represents the first spatially extensive database of this kind for Southern Africa. The datasets made available here include spatially interpolated $\delta^{2}H$, $\delta^{18}O$ and deuterium-excess maps based on an observation set of 410 tap water samples from across South Africa. Our tap water samples spanned a considerable isotopic range. For $\delta^{2}H$, the range was 77.4‰ (-54.5‰ to +22‰). For $\delta^{18}O$, the range was 13.1‰ (-7.7‰ to 5.4‰). For *d* the range was -30.9‰ and 22.7‰. Tap water prediction maps were produced following the methods of Bowen et al. (2007), involving a direct comparison with modeled precipitation isotope values. In general, coastal areas have more isotopically enriched tap water than inland areas. The most enriched values occur on the eastern seaboard of the Eastern Cape and KwaZulu-Natal. The most depleted values occur in the Kalahari and southwestern interior. Over extensive areas of the country, tap water and precipitation are isotopically similar. However, there are many areas where considerable discrepancies between these datasets prevail. The nature of these discrepancies potentially provides information about the tap water source (borehole or surface water) and its vulnerability to climate change. Our maps reveal a spatially coherent picture of the isotopic composition of tap water across South Africa. They provide an important baseline for future monitoring efforts as well as opening the door to novel ecological and forensic research.

All GIS data are freely available for download and use from http://waterisotopes.org.

Introduction

Managing all water resources is a key objective for government. As water resources increasingly come under pressure from the multiple stresses of population growth, pollution and changing climate, so does the need to identify and quantify potential changes to these resources. Tap water is a key deliverable for water resource management. Tap water represents the interface between human and hydrological systems and has a direct effect on human geography and socio-economic development. The stable hydrogen and oxygen isotopic composition of water is a valuable recorder of both hydrological pattern and process (Gat, 1996). Tap water has been shown to retain the signature of local precipitation as well as incorporating information on processes such as evaporation and chemical interaction with minerals or soils (Bowen et al., 2007). Differences between the isotopic composition of tap water and local precipitation (derived from well-developed geostatistical models – www.waterisotopes.org) provide information on the coupling of water resources to precipitation source and can provide insight into resources that might be vulnerable to changes in climate (Bowen et al., 2007).

Creating a spatially explicit map of the stable isotope ratios in tap water (hereafter an "isoscape") serves several important functions. In an era of rapid global change, we need spatially explicit and process-oriented baseline information in order to monitor changes to our water resources. Our tap water isoscape presented here provides an important baseline of the hydrological patterns and processes influencing tap water across South Africa. This isoscape will be an important reference for future hydrological studies, allow further novel questions to be asked and provide a springboard for future research efforts. In addition to water resource management, our tap water isoscape has other potential uses. Stable isotopes are wonderful recorders of ecological pattern and process (West et al., 2006). The isotopic composition of tap water transfers to the tissues of plants and animals that use this water in a predictable manner. As such, a spatially explicit map of the stable isotope ratios in tap water provides an important tool for ecological studies and forensic tracing (Bowen et al., 2005; Ehleringer et al., 2008; Kennedy et al., 2011).

Methods

Sample Collection

410 tap water samples were collected from around South Africa during the period April 2009 to December 2010 (Figure 1). We used two techniques to gather samples. The first technique involved a return mail campaign with the cooperation of the South African Post Office. We sent sample kits to 336 post offices nationally. The sample kit contained a letter stating the purpose of the study together with a brief instruction sheet, two empty vials, some Parafilm™ to seal the vials with and a postage-paid, return-addressed, padded envelope. An example of the sampling kit is contained in Appendix 1. Of the 336 sample kits sent out, 280 were returned successfully. This represents an 83% success rate for our sampling campaign and indicates this is an efficient and cost-effective strategy for repeat sampling campaigns. The second sampling strategy involved targeted and opportunistic sampling. Sampling kits were supplied to colleagues visiting specific locations within South Africa, or mailed to colleagues in remote locations. Additionally, the researchers visited some specific locations personally in order to collect samples in poorly sampled areas. Samples were collected in this manner at 130 locations.

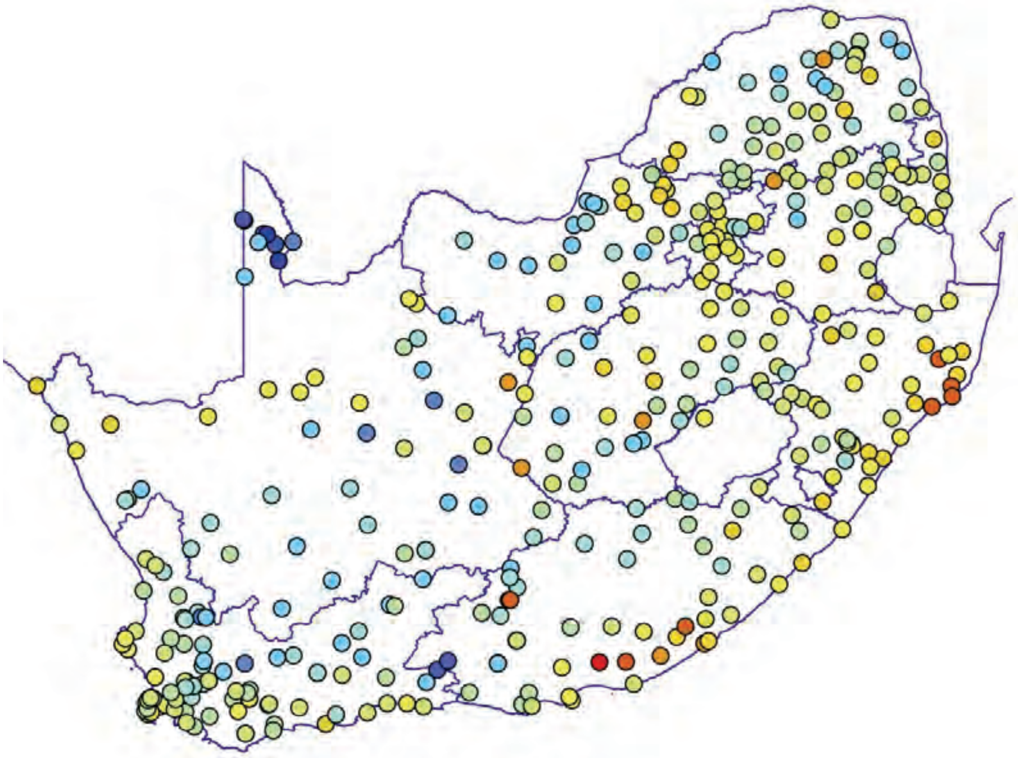

Figure 1: Locations of tap water samples collected across South Africa.

All tap water samples were collected in 8ml borosilicate glass vials with rubber-lined screw-top caps (LASEC, South Africa). As an extra-precaution, all filled vials had Parafilm™ wrapped around the caps to prevent caps from coming loose and the sample becoming evaporatively enriched.

Isotope analyses

Samples were analyzed by two different isotopic methods. A considerable fraction of the collected samples were analyzed by traditional, offline isotope ratio mass spectrometry (IRMS) methods. δ^2H analyses were performed using the closed-tube Zn-reduction method (Coleman et al., 1982). δ^{18}O was measured using the CO_2-equilibration method (Socki et al., 1992). The isotopic ratio of the H_2 and CO_2 gases was analyzed via dual-inlet on a Finnigan MAT 252 mass spectrometer (Bremen, Germany) in the Archaeometry Laboratory, University of Cape Town, South Africa.

During this time consuming process, a new water isotope analyzer (L2120-i; Picarro Inc., 480 Oakmean Parkway, Sunnyvale, California, 94085, USA; www.picarro.com) became available for stable isotope analysis at the University of Cape Town. In contrast to IRMS, the L2120-i utilizes wavelength-scanned cavity ring-down spectrometry (WS-CRDS) to determine the isotopic composition of water directly via an analysis of infra-red absorbance in specific wavelengths. As such, the L2120-i is able to conduct high quality isotopic analyses on micro-liter quantities of water, without the time consuming chemical conversion steps required by traditional IRMS (Brand et al., 2009b). Duplicate samples were analyzed on the L2120-i and cross-checked with the existing IRMS data (see Figure 2). Once the instrument had been validated, remaining samples were analyzed on this instrument.

For each WS-CRDS analysis, microliter quantities of water were injected into a vaporization chamber and then passed into an infrared absorbance cavity. Isotope ratios (δ^2H and δ^{18}O) are calculated from the ring-down time in specific wavelengths using WS-CRDS (Gupta et al., 2009). These analyses were conducted in the Botany Department, University of Cape Town. WS-CRDS measurements have been shown to be susceptible to organic contaminants (Brand et al., 2009b; West et al., 2010a). However, spectral analysis software is able to accurately identify problematic samples (West et al., 2011). Following the methodology of West et al. (2011), all WS-CRDS samples were screened post-analysis by Chemcorrect™ version 1.0.0

using analysis file “chemcorrect_inst avg_orgeval_06.csv” (Electronic supplement). None of the tap water samples were identified as problematic. Nevertheless, selected samples were cross-checked on IRMS to confirm the Chemcorrect™ results. All comparisons were within the convolved error of the two analytical methods.

For all methods, isotope ratios are expressed in ‰ (Brand et al., 2009a) as:

$$\delta^{N}E = \left(\frac{R_{sample}}{R_{standard}} - 1 \right) \qquad (1)$$

where N represents the heavy isotope of element E and R is the ratio of the heavy to light isotope ($^{2}H/H$ or $^{18}O/^{16}O$). For all methods, two calibration standards that spanned the range of isotopic values expected for our samples were used to adjust the delta values relative to V-SMOW. For the WS-CRDS data, precision and accuracy obtained via quality control standards with identical isotope ratios introduced into every run was 0.2‰ and 1.5‰ for $\delta^{2}H$ and 0.07‰ and 0.13‰ for $\delta^{18}O$ respectively.

Spatial analyses

Isotope data were analysed spatially following the methodology of Bowen et al. (2007). Briefly, tap water data were compared with $\delta^{18}O$ and $\delta^{2}H$ values of precipitation at the sampling sites as obtained from the Online Isotopes in Precipitation Calculator (OIPC, http://waterisotopes.org, accessed June 2011) The OIPC calculates long-term average precipitation $\delta^{18}O$ and $\delta^{2}H$ at specified locations through geostatistical modeling of precipitation isotope data collected from 1960-2004 (Bowen and Wilkinson, 2002; Bowen and Revenaugh, 2003; Bowen et al., 2005). To create prediction maps of average tap water isotope ratios, differences were calculated between measured isotope ratios and OIPC precipitation isotope ratios at each tap water collection site. Surfaces of tap – precipitation for $\delta^{2}H$, $\delta^{18}O$, and *d* were generated by ordinary kriging using a spherical semivariogram with nugget. The $\delta^{2}H$, $\delta^{18}O$ and *d* prediction maps were created by summing the OIPC precipitation isotope layer for each element and the corresponding tap minus precipitation difference layer. Cross-validation of the Kriged difference layers using Geostatistical Analyst was used to estimate prediction errors (root mean square error) for the tap water $\delta^{2}H$ and $\delta^{18}O$ maps. All files were created or analyzed in ArcGIS Desktop for Windows 9.3.

Results

Comparison of IRMS and WS-CRDS data

The two isotopic methods used (IRMS and WS-CRDS) compared very favourably with each other (Figure 2). Certainly there is no systematic bias from using one technique over the other. If anything, the IRMS data is noisier than the WS-CRDS data most likely due to the chemical conversion steps that can introduce error in the laboratory. In particular, the $\delta^{18}O$ IRMS data is noisier than the $\delta^{2}H$ data probably due to the potential for minor evaporation in the samples when prepared offline for analysis. The CO_2 equilibration method is prone to such potential problems to a far greater extent than the Zn-reduction method. This is supported by the fact that the IRMS data appears to be slightly more enriched than the WS-CRDS data at more positive isotope ratios (Figure 2A). Thus, we are confident that the WS-CRDS data is of good quality and these data are used for the remainder of the study.

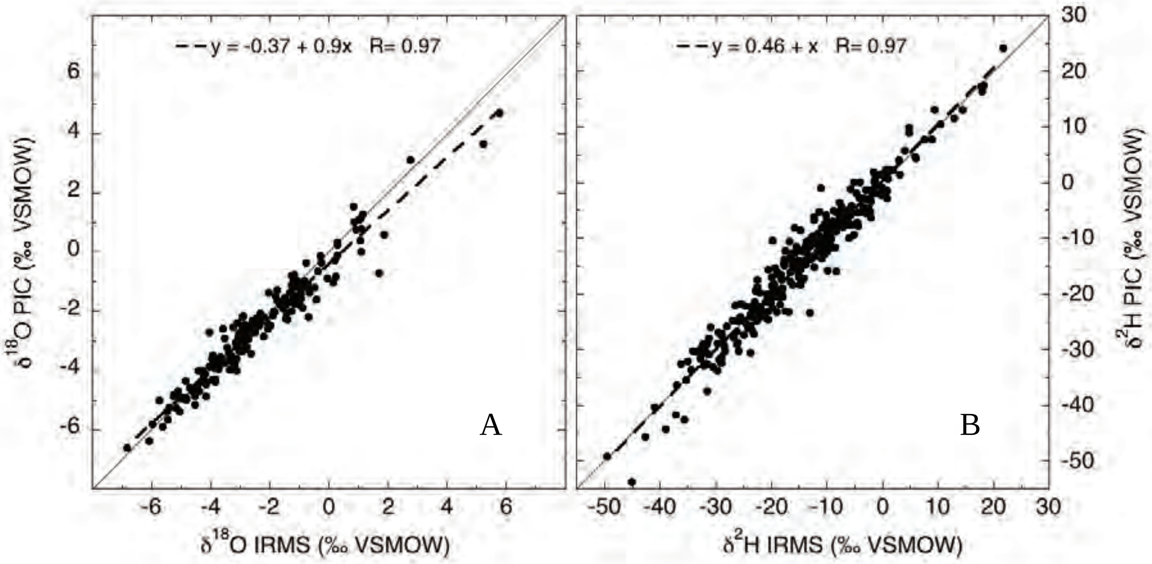

Figure 2: A comparison of the $\delta^{18}O$ and $\delta^{2}H$ data produced via IRMS techniques and the Picarro L2120-i WS-CRDS instrument. In each plot the regression is shown (dashed line), as is a 1:1 line for reference (solid line).

Range and distribution of tap water data

Our tap water samples spanned a considerable isotopic range. For δ^2H, the range was 77.4‰ (-54.5‰ to +22‰). For δ^{18}O, the range was 13.1‰ (-7.7‰ to 5.4‰). Deuterium excess (*d*, calculated as $d = \delta^2H - 8*\delta^{18}O$) ranged between -30.9‰ and 22.7‰. As revealed in a dual isotope plot (Figure 3), these numbers are indicative of waters of both meteoric origin (samples close to the Global Meteoric Water Line, y = 8x + 10) as well as some evaporated surface waters (samples with low *d*-excess).

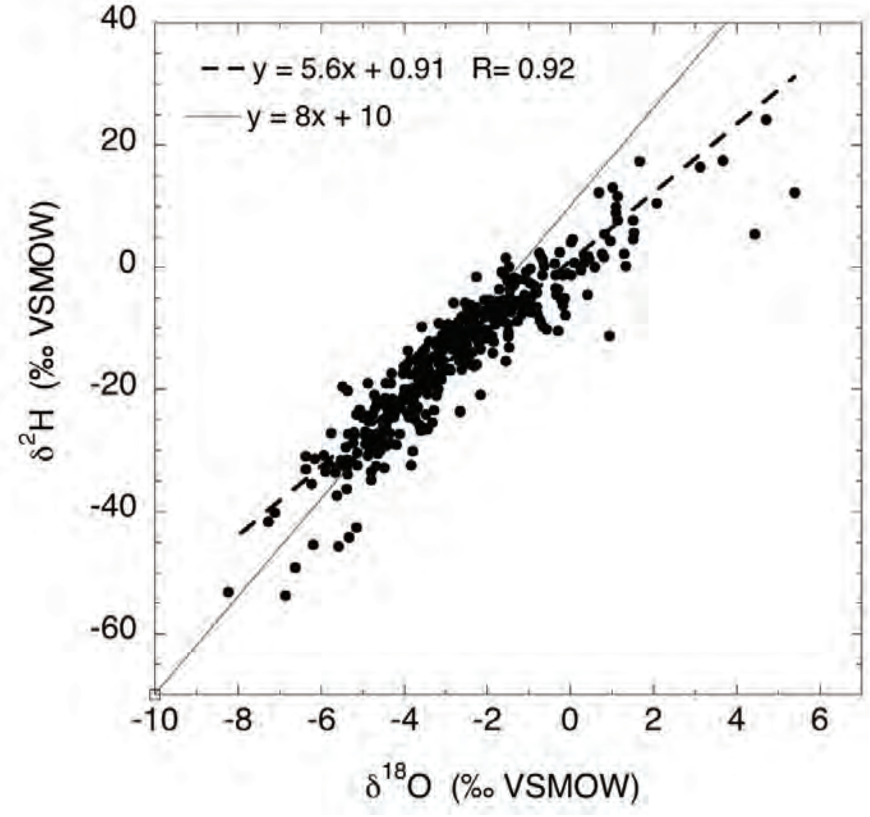

Figure 3: δ^2H vs. δ^{18}O plot of the tap water samples analyzed in this study. The regression through the data points is shown (y = 5.6x + 0.91). The Global Meteoric Water Line (y = 8x+10) is plotted for reference.

Although samples were collected over a period of time (April 2009 to December 2010), the majority of samples were collected in the summer months (DJFMA = 344). Furthermore, there was no deliberate spatial bias during any of the sampling campaigns and samples were collected from a wide range of locations at all time periods. Figure 4 shows the isotope data plotted by sampling time. There does not appear to be any specific variation by sampling time, thus we do not feel that the temporal range of our sampling effort significantly compromises our spatial analyses.

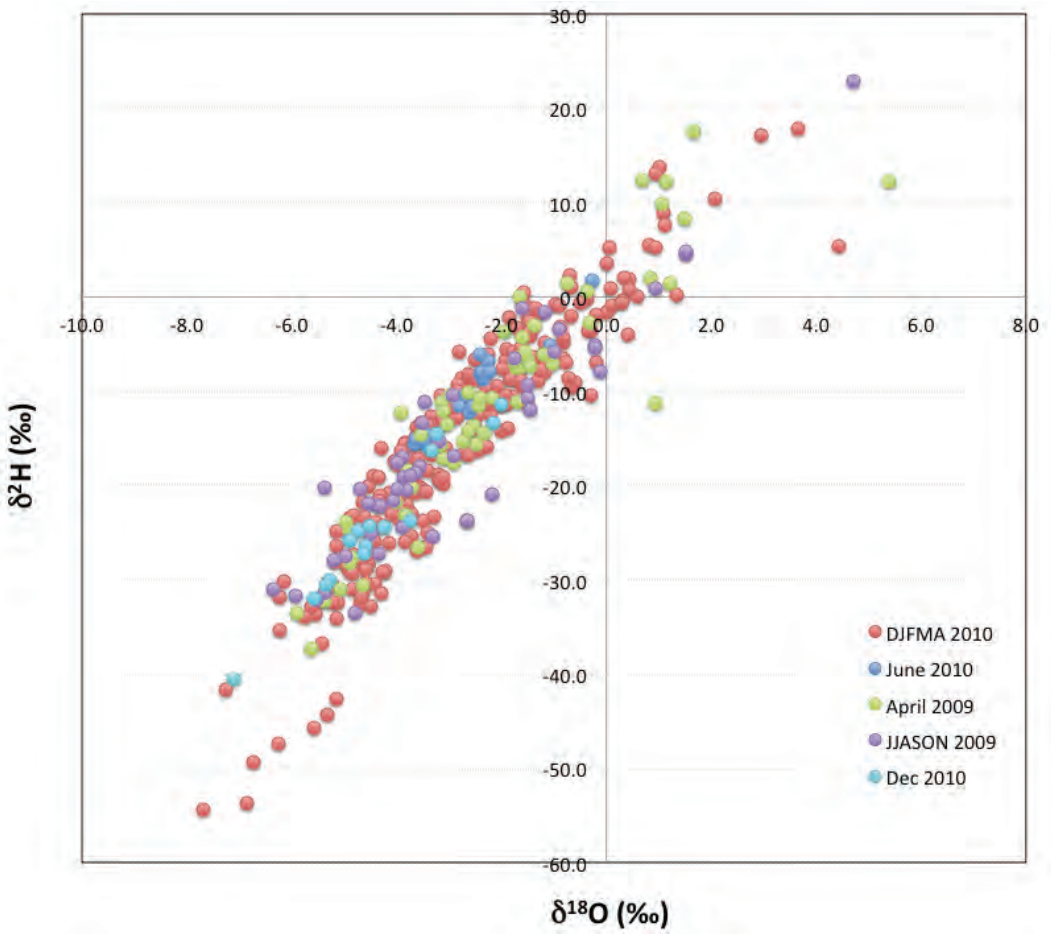

Figure 4: δ^2H and δ^{18}O of tap water samples plotted by collection date. DJFMA = December, January, February, March, April. JJASON = June, July, August, September, October. There appears to be little effect of sampling time on the distribution of the isotope data.

Spatial results

Our spatial results are presented in sets of four figures for δ^2H, δ^{18}O and d-excess. Each set contains: 1) Prediction map of tap water isotopes across South Africa, 2) Actual tap observations plotted over the modeled precipitation field, 3) Tap water residuals (tap – precipitation) plotted as point and interpolated values, 4) Standard error of the interpolated tap water results. A list of the figures follows (Table 1):

Table 1: A list of the spatial figures presented in this report.

Figure	Data source	Prediction map	Observations and precipitation layer	Residuals	Standard errors
5	δ^2H	X			
6	δ^2H		X		
7	δ^2H			X	
8	δ^2H				X
9	δ^{18}O	X			
10	δ^{18}O		X		
11	δ^{18}O			X	
12	δ^{18}O				X
13	d-excess	X			
14	d-excess		X		
15	d-excess			X	
16	d-excess				X

The considerable range observed in the isotope data emerges as a spatially coherent pattern across South Africa (Figures 5, 9). Very broadly speaking, for δ^2H and δ^{18}O:

- More negative values occur in the winter rainfall dominated areas (western side of South Africa) and the interior (Karoo and Kalahari), with the most negative values occurring in the Kalahari.
- More positive values occur in the summer rainfall regions (Free State, Gauteng, KwaZulu-Natal, Mpumalanga) with very positive values along Eastern Cape and KwaZulu-Natal coast.
- Slightly more negative values occur in the north-eastern part of the country (Limpopo, northern Mpumalanga).

Low d-excess was observed in the central and northern areas of the country, particularly in the Free State, Northern Cape and Northwest Provinces (Figure 13). The residuals of tap water – precipitation indicate that the greatest differences between tap water and precipitation occur in the higher elevation areas of South Africa. The maps of standard error (Figures 8, 12, 16) show that confidence in our predicted surfaces decreases in the Northern Cape, northern Limpopo and the Lesotho highlands due to low sample density.

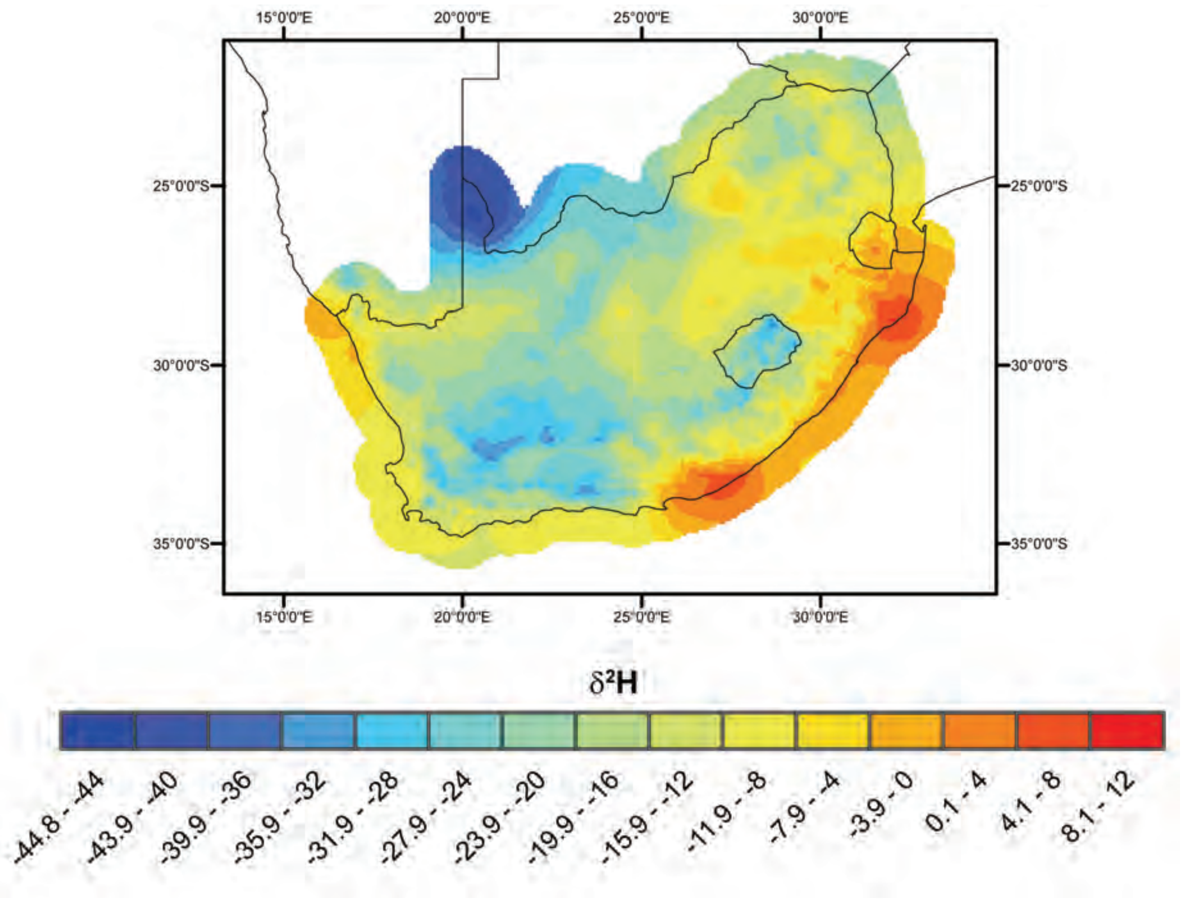

Figure 5: Prediction map of tap water δ^2H across South Africa

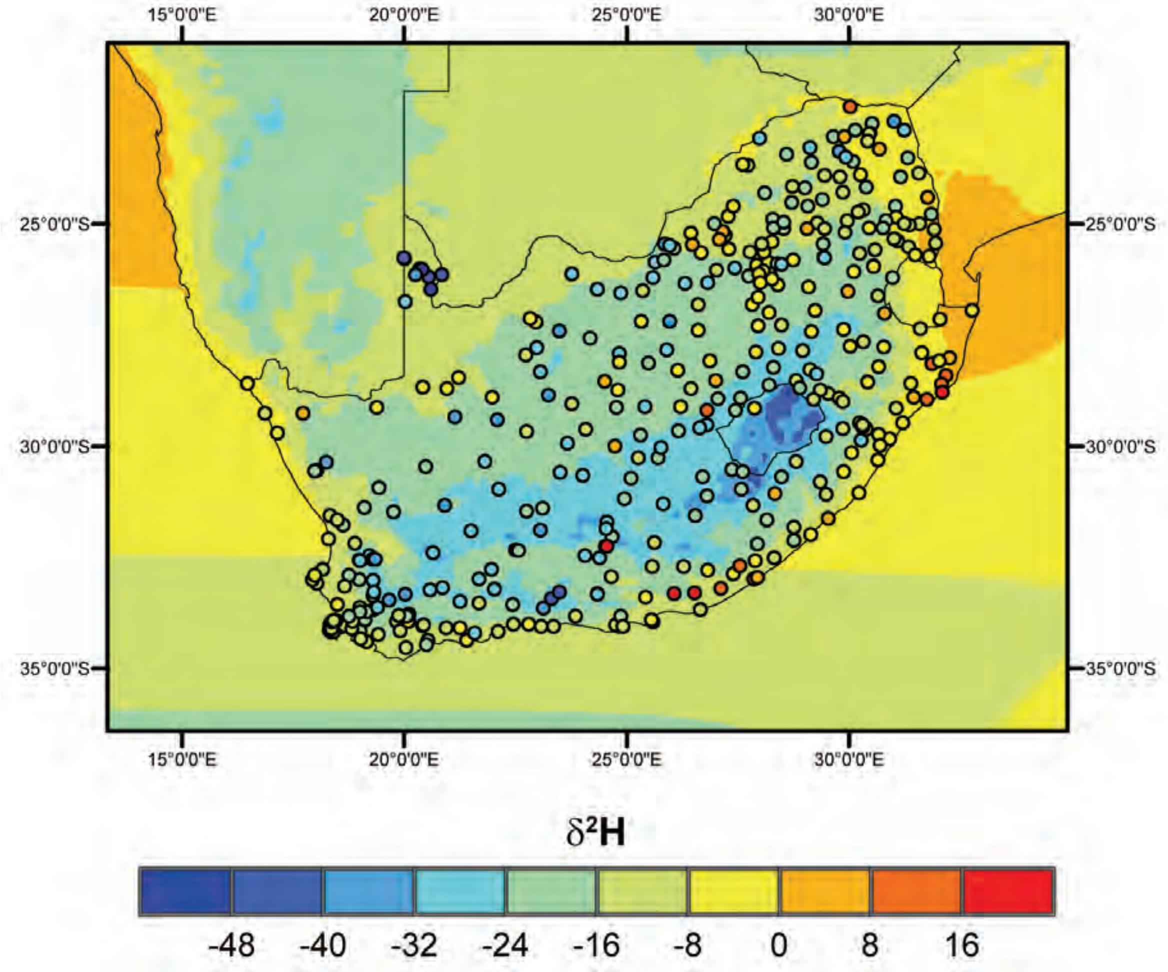

Figure 6: Actual tap observations (δ^2H) plotted over the modeled precipitation field

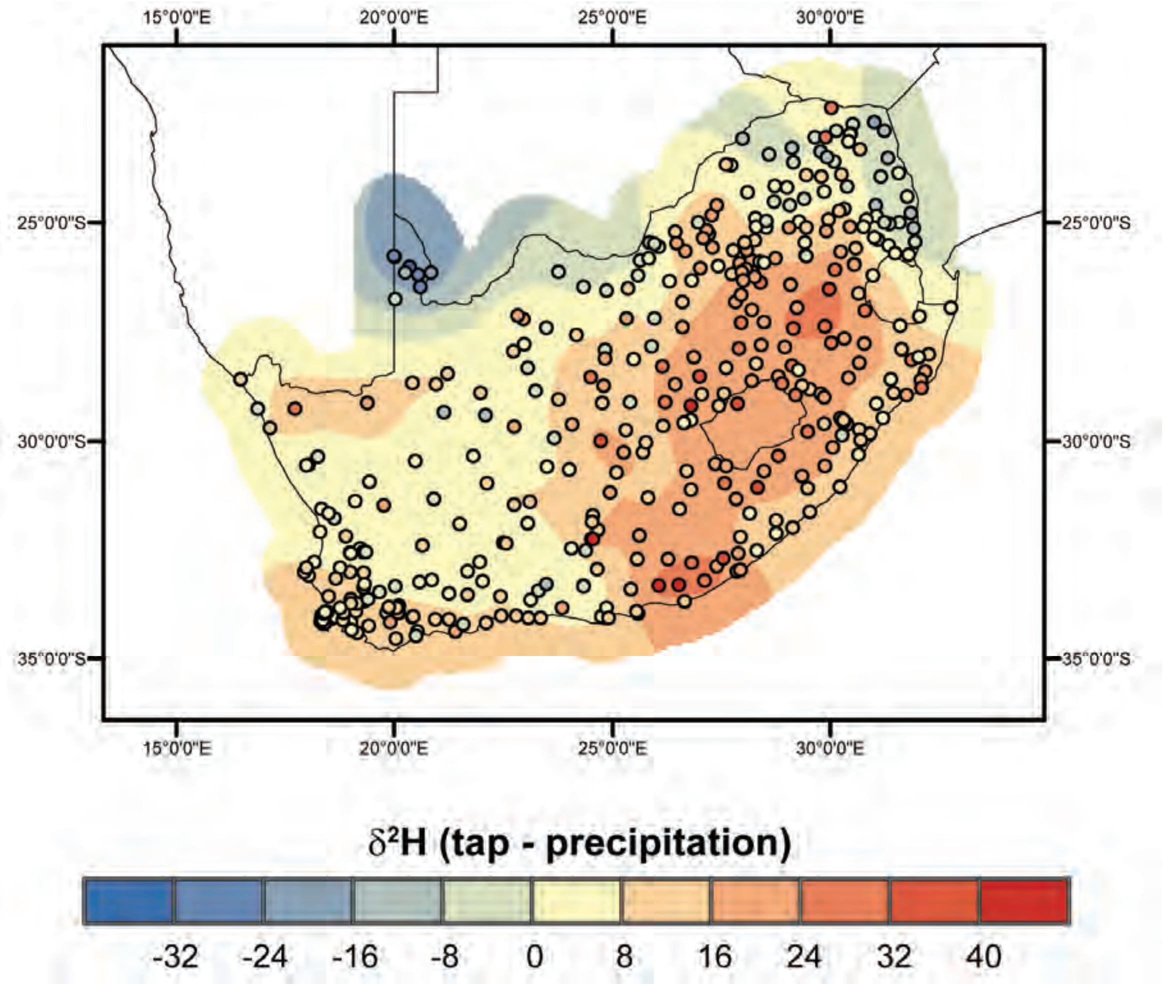

Figure 7: Tap water residuals (tap δ^2H – precipitation δ^2H) plotted as point and interpolated values.

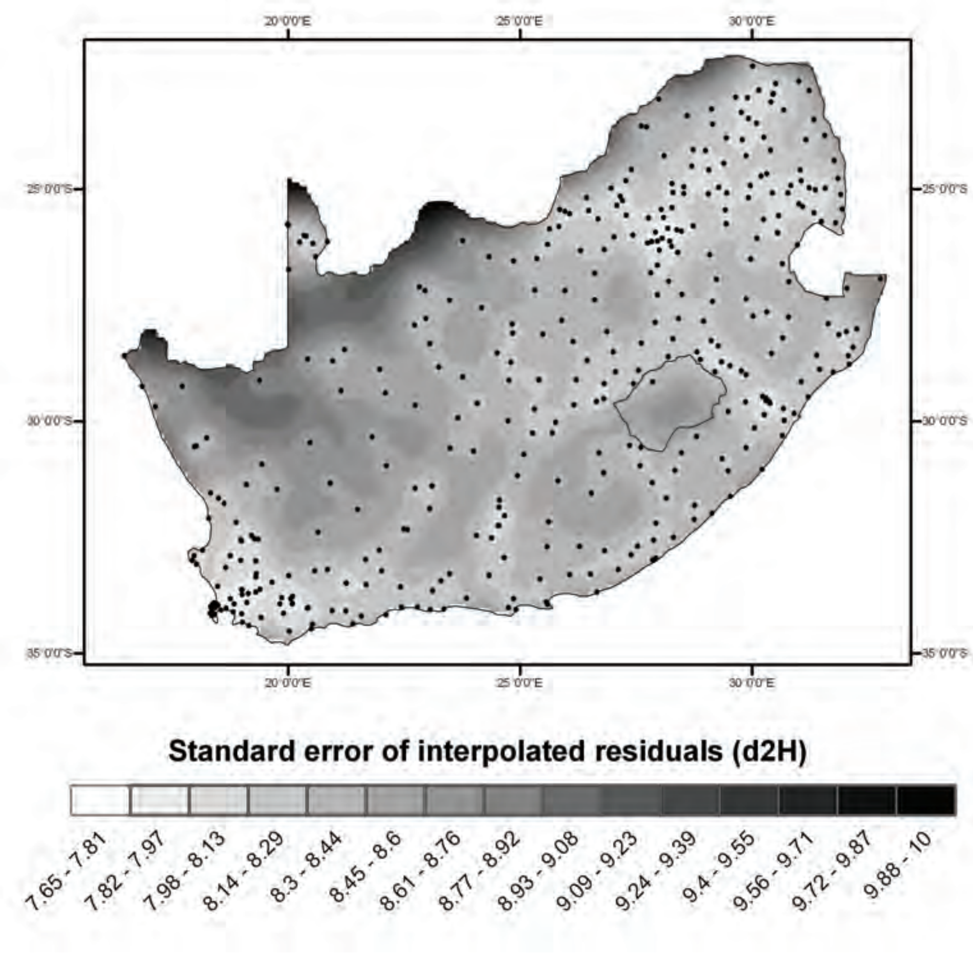

Figure 8: Standard error of the interpolated δ^2H tap water results

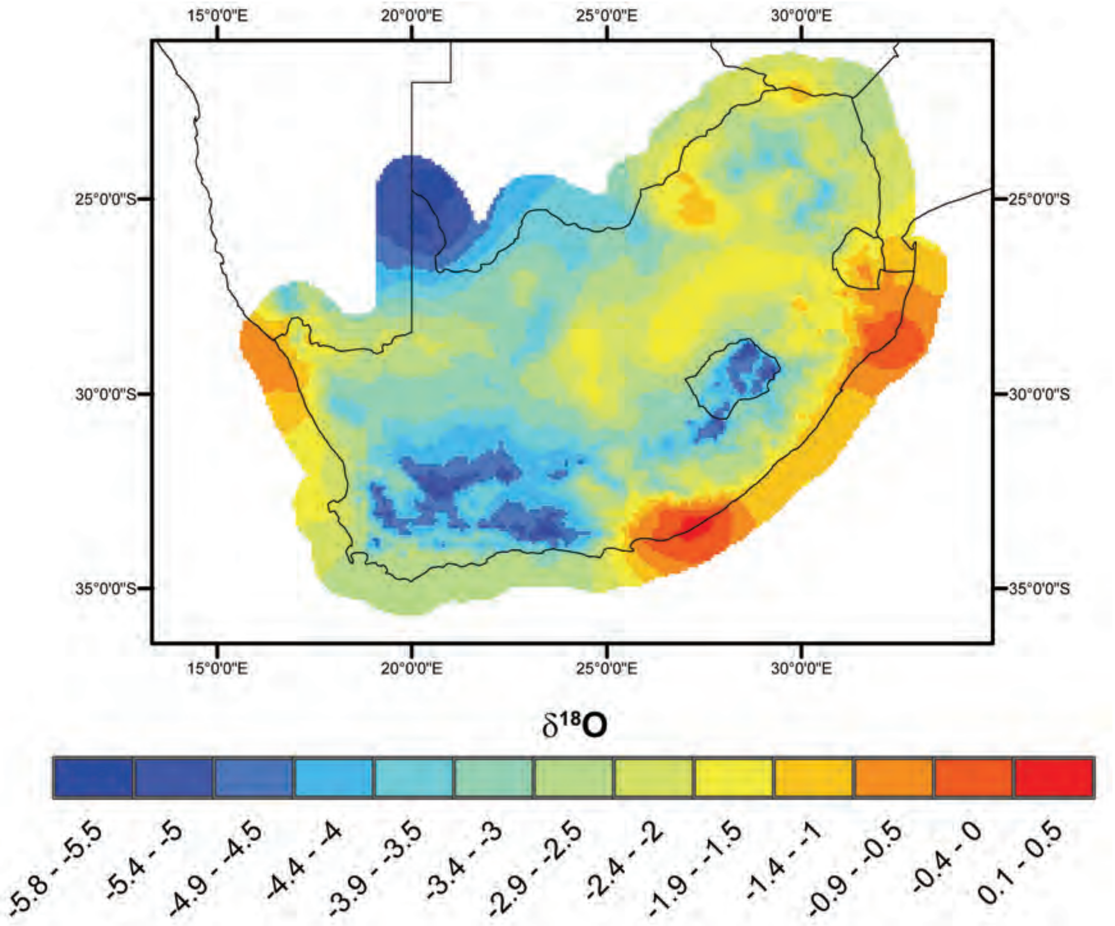

Figure 9: Prediction map of tap water δ^{18}O across South Africa

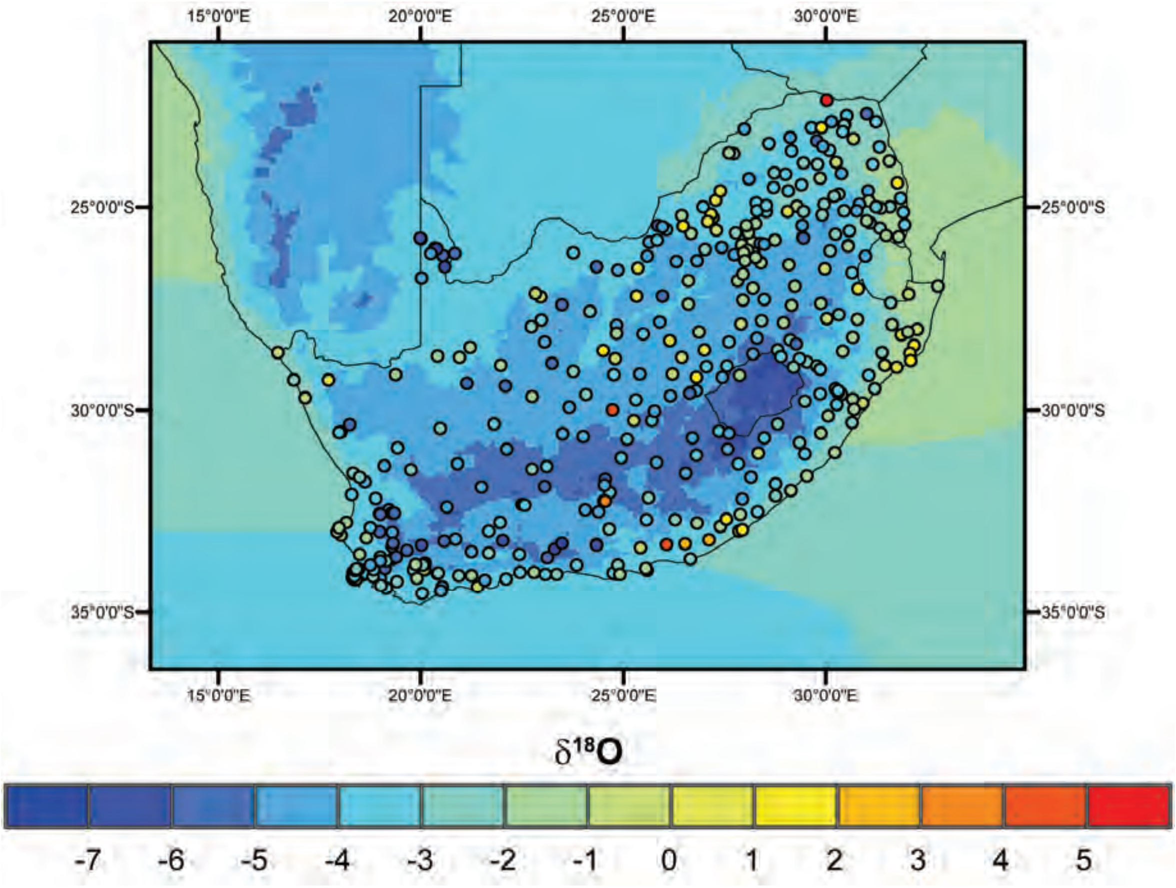

Figure 10: Actual tap observations ($\delta^{18}O$) plotted over the modeled precipitation field

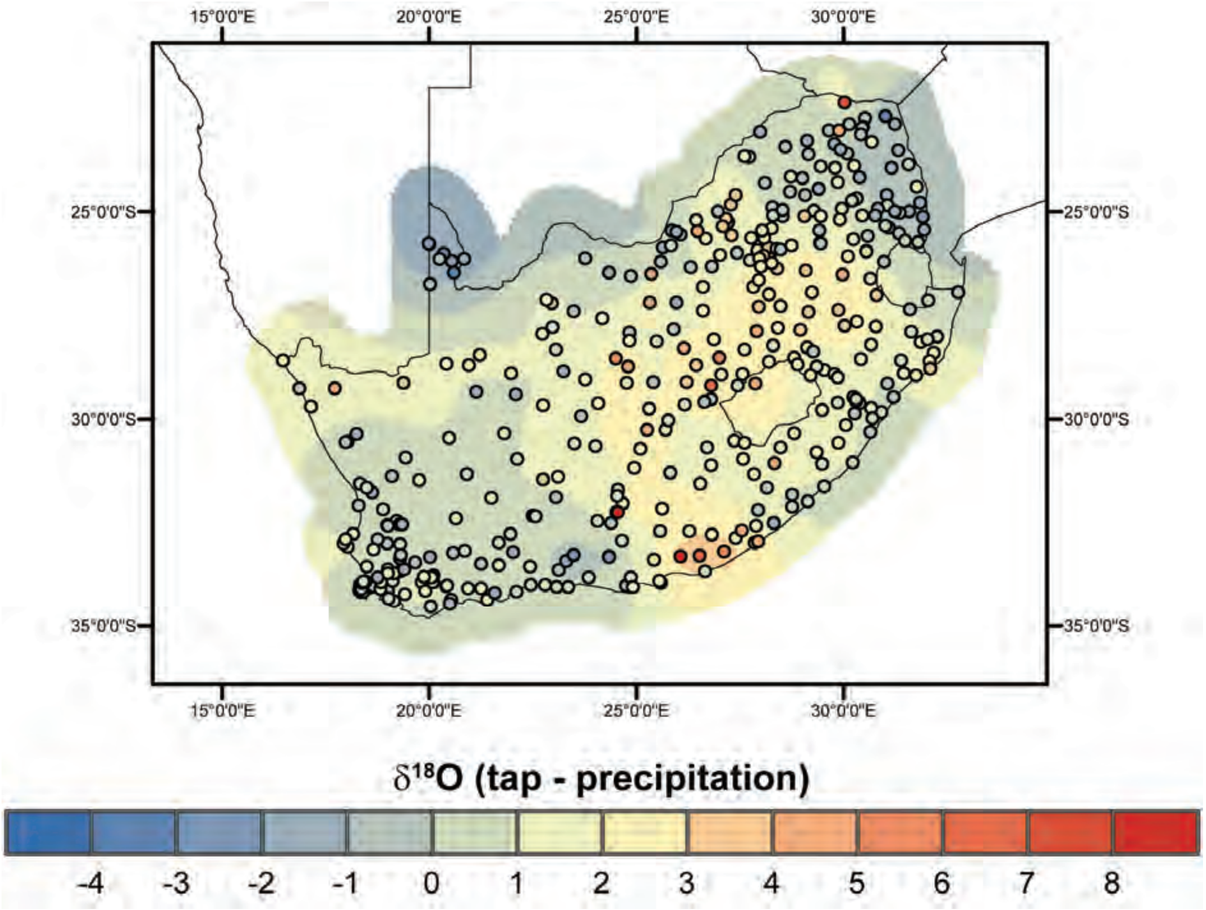

Figure 11: Tap water residuals (tap δ^{18}O – precipitation δ^{18}O) plotted as point and interpolated values.

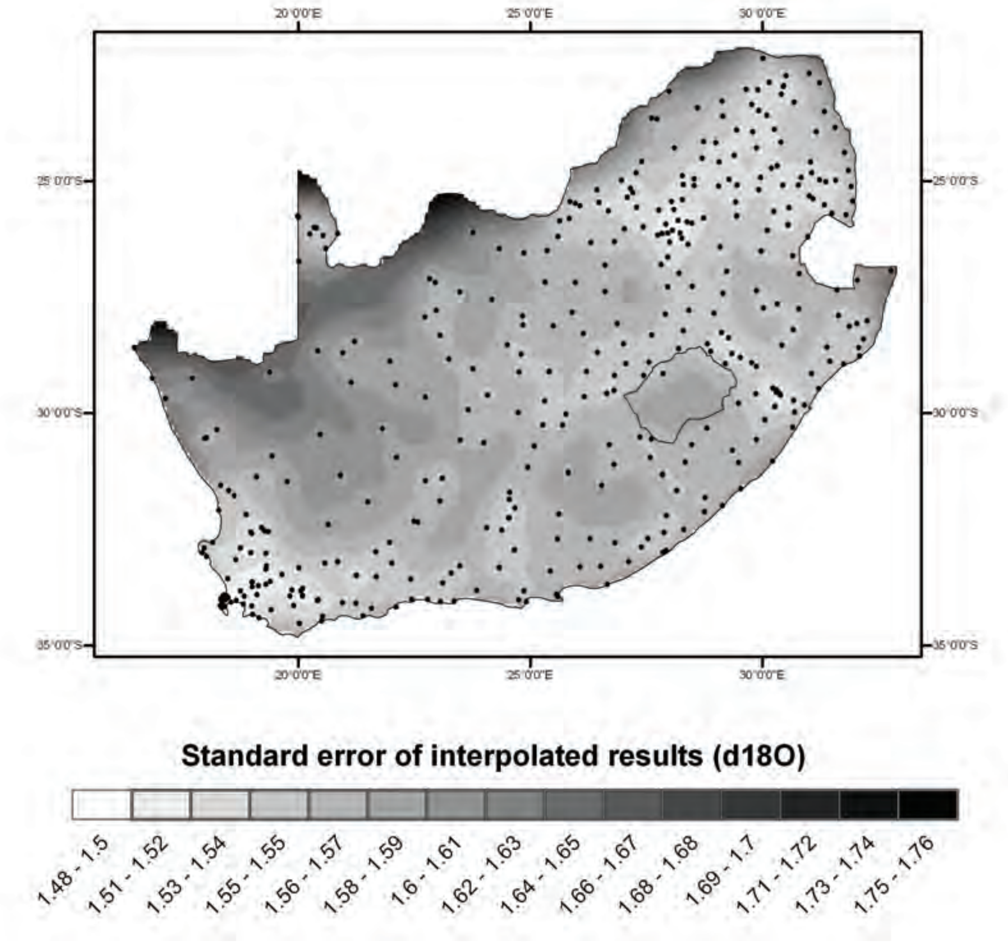

Figure 12: Standard error of the interpolated δ^{18}O tap water results

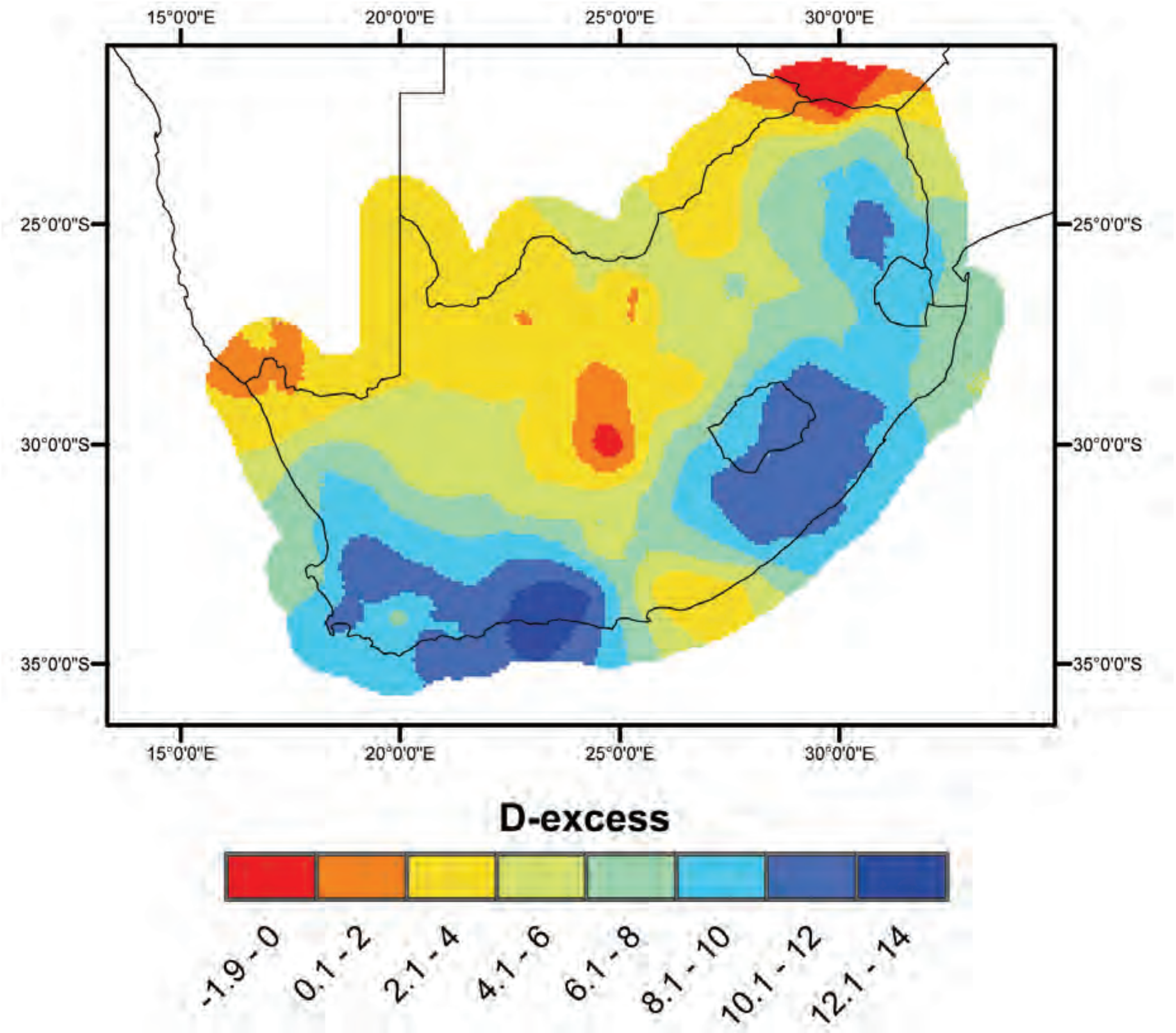

Figure 13: Prediction map of tap water *d* across South Africa

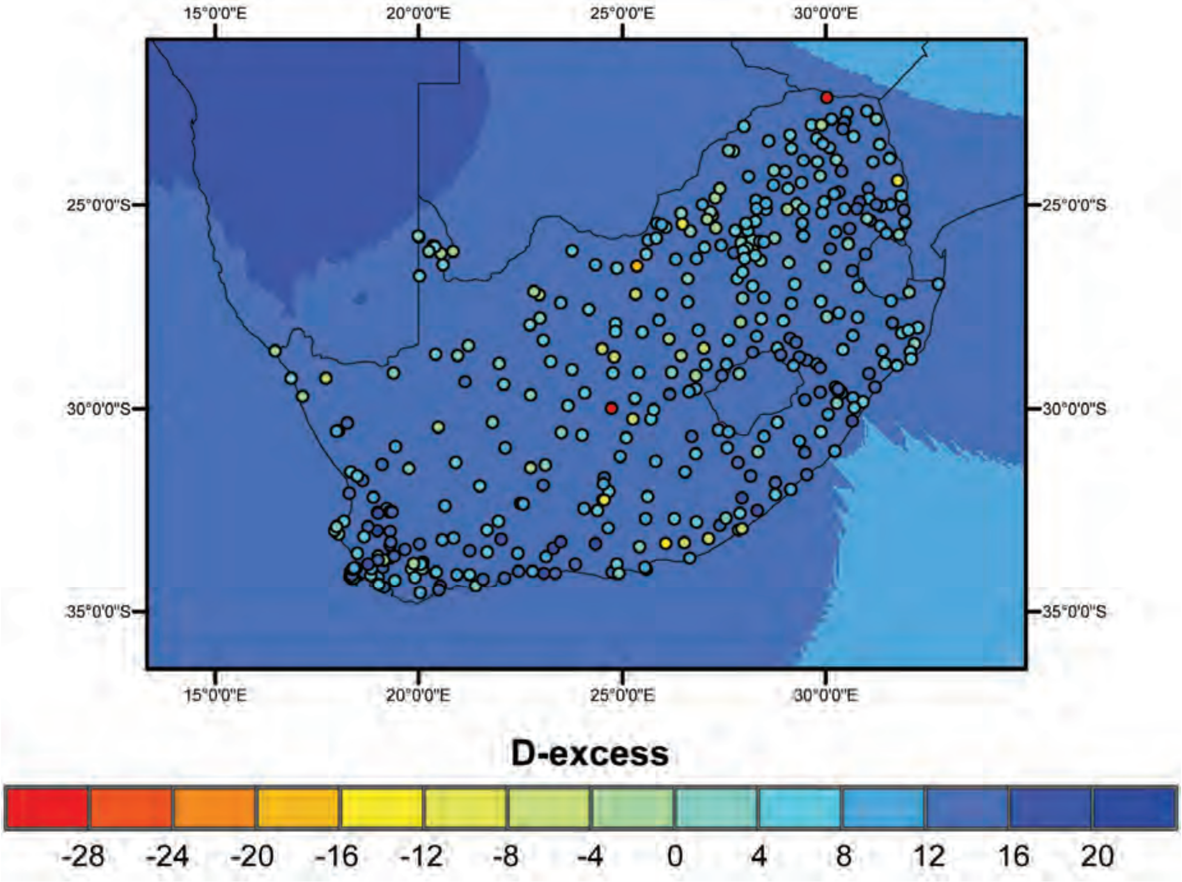

Figure 14: Actual tap observations *d* plotted over the modeled precipitation field

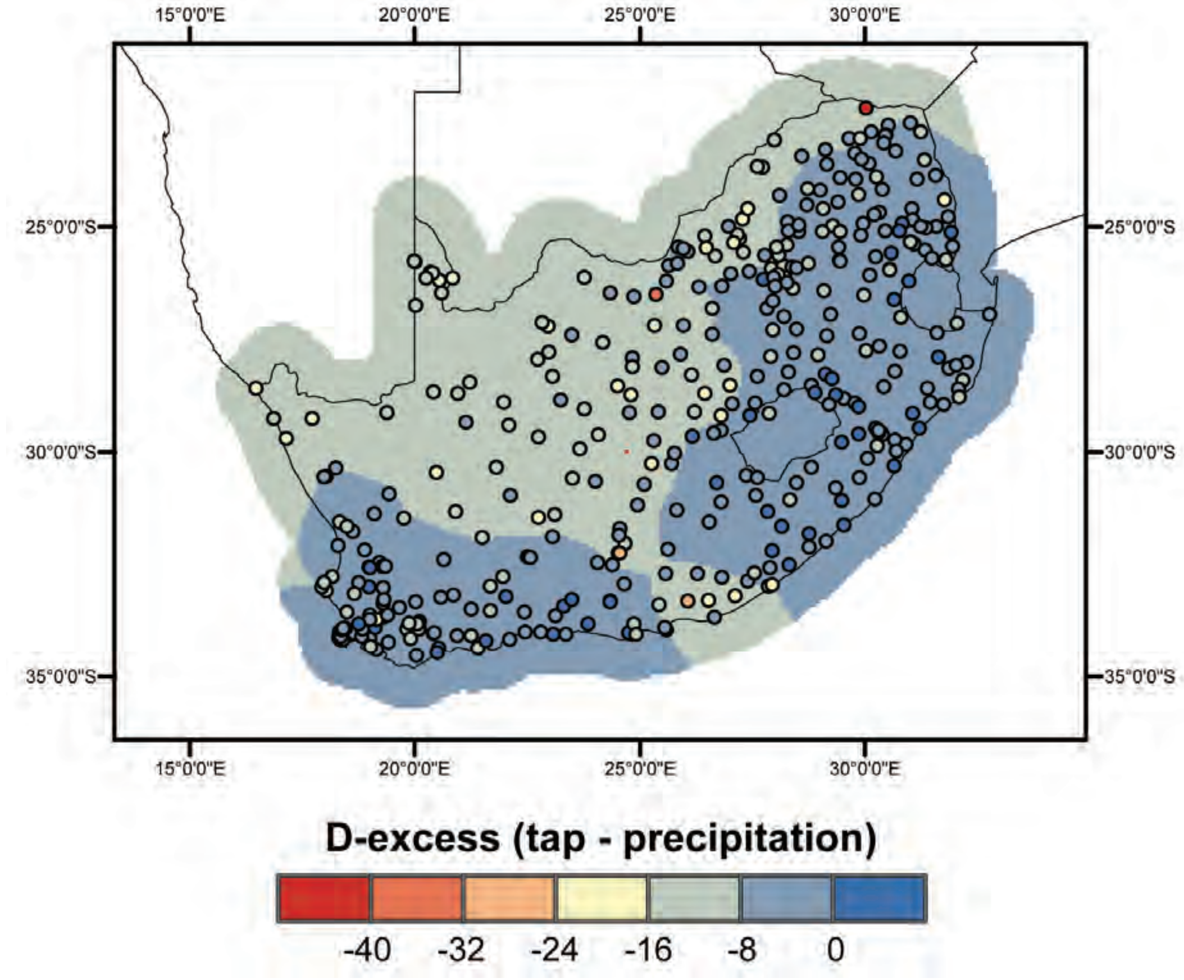

Figure 15: Tap water residuals (tap *d* – precipitation *d*) plotted as point and interpolated values.

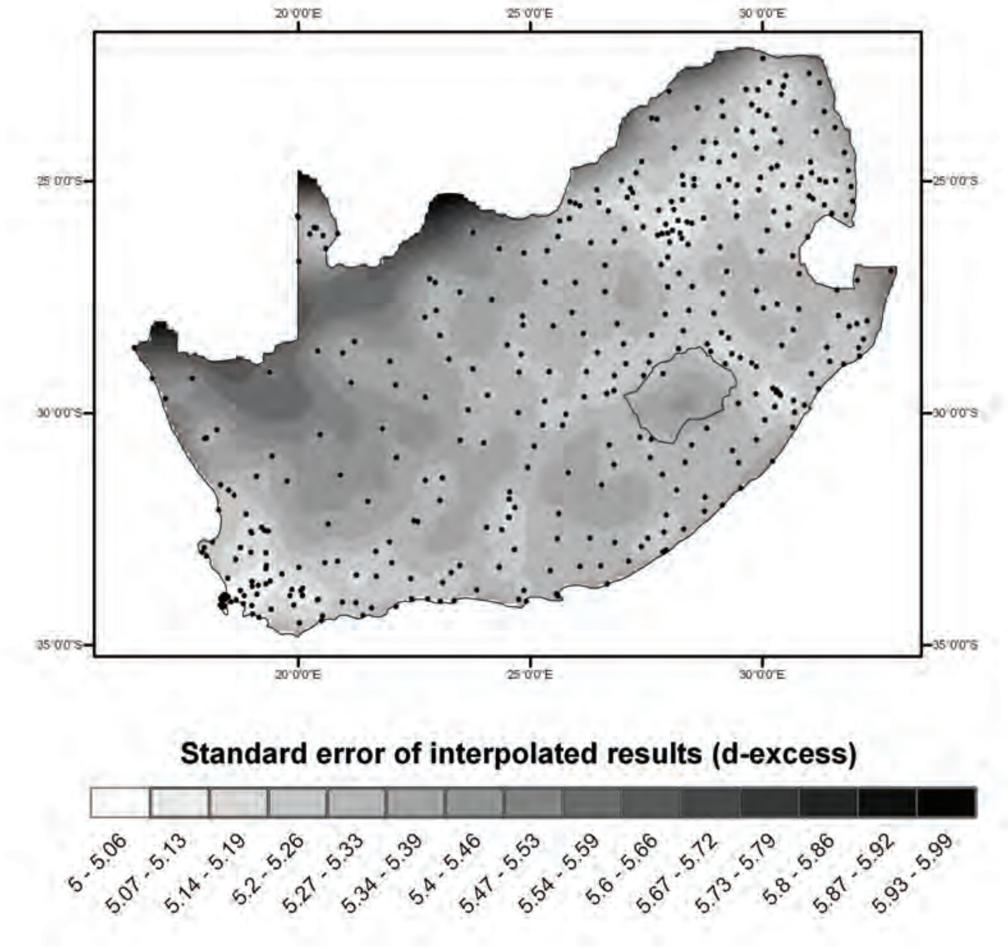

Figure 16: Standard error of the interpolated *d* tap water results

Discussion

Our goal was to produce a map of tap water isotope ratios across South Africa to add to the growing scientific effort in producing “isoscapes” (West et al., 2010b). The maps we have produced show a coherent spatial distribution that makes intuitive sense based on our understanding of stable isotope distribution in natural waters (Criss, 1999). For example, much of the spatial structure in the tap water data is closely related to that of the precipitation distribution (Figures 6, 10, 14). This can be seen from the extensive areas with small residuals from the precipitation layers (Figures 7, 11, 15). For areas where there is a large discrepancy between tap water and precipitation values, there is potential to explore the mechanisms for this decoupling. For example, where tap water is isotopically more negative than precipitation (a negative residual), this may be indicative of borehole or groundwater use. Ground waters are often only recharged by large precipitation events that characteristically have a relatively negative isotopic composition. Extremely negative differentials may reflect the use of fossil water; water that was not produced under current climatic conditions. Where tap water is more positive than precipitation (a positive residual), this may be indicative of a tap water source open to evaporation such as dam water. This is supported in areas where the *d*-excess is low as this is a characteristic sign of evapoconcentration of surface waters. Another potential use of this discrepancy between the isotopic composition of tap water and precipitation is for leak detection of urban mains water. As demand for municipal water resources increases, possibly in concert with decreases in recharge due to climate change (New, 2002), an important step in maintaining water supply is to reduce unnecessary losses from the system. As has been shown in previous studies (Harris et al., 1999; Verhagen and Butler, 2001), the stable isotopic composition of tap water can be used as a potential tracer of mains water leakage. This is only the case where tap water presents an isotopic signal distinct from other water sources in the area (e.g. precipitation, ground water). As can be seen from our maps, there are many areas where the isotopic signature of tap water differs considerably from the modeled precipitation value (Figures 7, 11, 15). In these areas, the stable isotopic composition of water may provide a useful tracer for detecting mains water leakages.

However, the real power in maps like these comes from monitoring over time. Environmental change can be reflected in the isotope ratios of water. Documenting seasonal and annual changes in isotopic composition of tap water (Kennedy et al., 2011) will allow resource managers to identify hotspots of change, potentially detecting areas prone to increased losses from evaporation, or changes in seasonality of rainfall.

Lastly, our isotope maps may be of interest to other research communities outside of the water resources sector. For example, maps of stable isotope of drinking water can be used to predict the isotopic composition of biological tissues such as hair (Ehleringer et al., 2008) and bird feathers (Hobson and Wassenaar, 1997). As such, these maps may aid novel research in ecological and forensic studies that ultimately relate to the spatiotemporal variation in water resources (Kennedy et al., 2011). In order to facilitate future research, all GIS data are freely available for download and use from http://waterisotopes.org.

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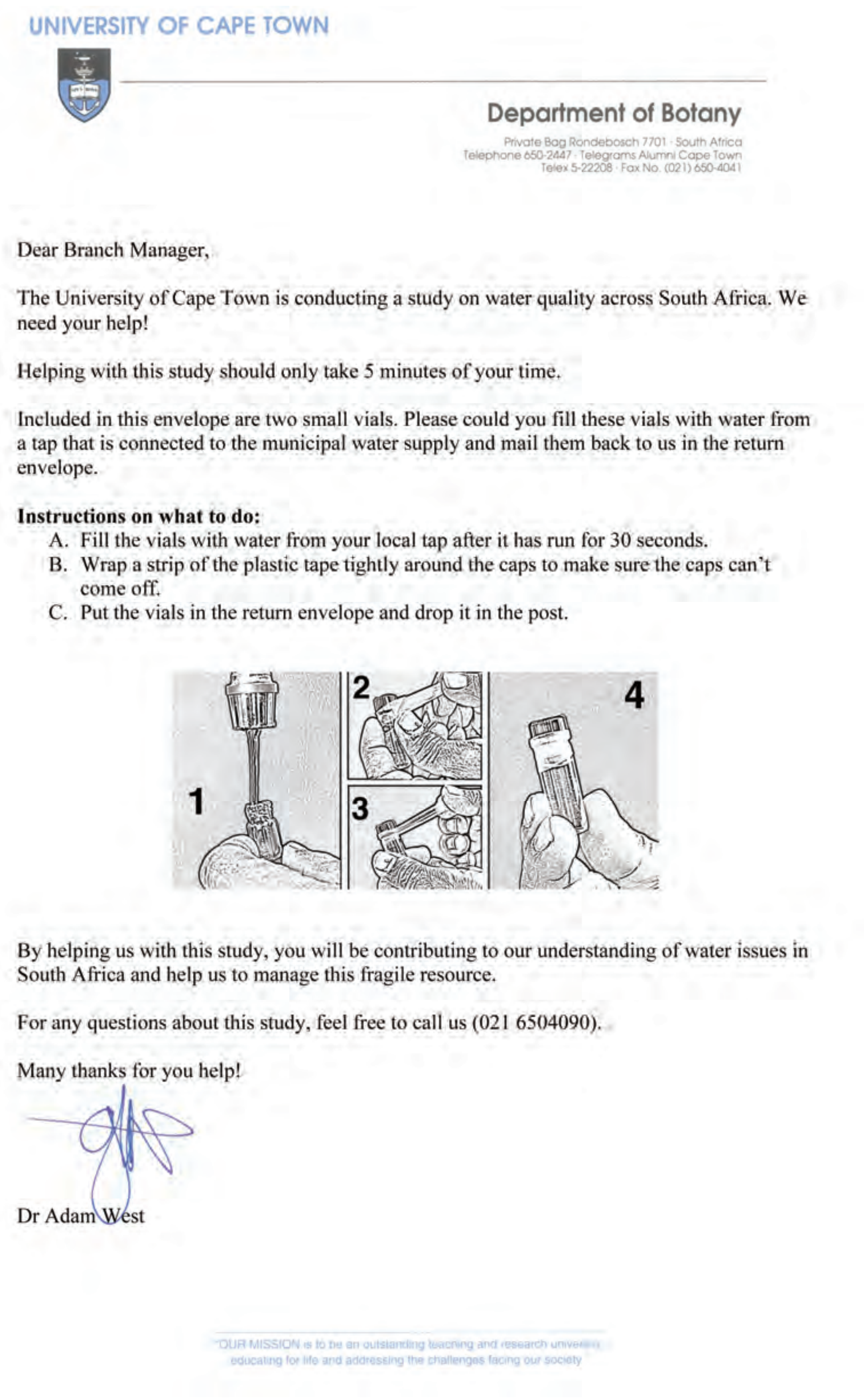
List of Electronic supplements:

1) Isotope data: “tap water data final.csv”
2) Chemcorrect™ analysis file: “chemcorrect_inst avg_orgeval_06.csv”
3) GIS layer files (described in Appendix 2): “GIS files.zip”

Appendix 1: An example of the mailer sent to post-offices around the country.



UNIVERSITY OF CAPE TOWN

Department of Botany
Private Bag Rondebosch 7701 · South Africa
Telephone 650-2447 · Telegrams Alumni Cape Town
Telex 5-22208 · Fax No. (021) 650-4041

Dear Branch Manager,

The University of Cape Town is conducting a study on water quality across South Africa. We need your help!

Helping with this study should only take 5 minutes of your time.

Included in this envelope are two small vials. Please could you fill these vials with water from a tap that is connected to the municipal water supply and mail them back to us in the return envelope.

Instructions on what to do:

A. Fill the vials with water from your local tap after it has run for 30 seconds.
B. Wrap a strip of the plastic tape tightly around the caps to make sure the caps can't come off.
C. Put the vials in the return envelope and drop it in the post.

By helping us with this study, you will be contributing to our understanding of water issues in South Africa and help us to manage this fragile resource.

For any questions about this study, feel free to call us (021 6504090).

Many thanks for you help!

Dr Adam West

"OUR MISSION is to be an outstanding teaching and research university, educating for life and addressing the challenges facing our society"

Appendix 2: GIS files for analysis and mapping of South African tap water isotope ratios

Analysis by G.J. Bowen, 6/1/2011
All files created or analyzed in ArcGIS Desktop for Windows 9.3 and Adobe Illustrator CS2

Tabular Data

110520tapwater.csv
Raw data file containing isotope ratio data for monitoring sites

Raster Data (ArcASCII format)

dexres
D-excess residuals (measured tap – modeled precip) interpolated using ordinary kriging

dexres_msk
Version of *dexres* masked to *ZA_Buffer*

dexres_se
Standard error of interpolated predictions for D-excess residual values

hres
δ^2H residuals (measured tap – modeled precip) interpolated using ordinary kriging

hres_msk
Version of *hres* masked to *ZA_Buffer*

hres_se
Standard error of interpolated predictions for δ^2H residual values

ores
δ^{18}O residuals (measured tap – modeled precip) interpolated using ordinary kriging

ores_msk
Version of *ores* masked to *ZA_Buffer*

ores_se
Standard error of interpolated predictions for δ^{18}O residual values

tapdex
Prediction map for tap water D-excess, calculated as precipitation D-excess + *dexres*

taph
Prediction map for tap water δ^2H, calculated as precipitation δ^2H + *hres*

tapo
Prediction map for tap water δ^{18}O, calculated as precipitation δ^{18}O + *ores*

Vector Data

sites_110601
Shapefile containing monitoring station data

ZA
Shapefile of South Africa country border

ZA_Buffer
Shapefile of 100 buffer zone around South African boarder

List of Illustrator Figure Files

d2H, d18O, dex
Tap observations plotted as points over the OIPC precipitation field

d2H_res, d18O_res, dex_res
Tap water residuals (tap – precipitation) plotted as point and interpolated values

d2H_map, d18O_map, dex_map
Prediction maps for tap water isotope values