



Massive arrival of desalinated seawater in a regional urban water cycle: A multi-isotope study (B, S, O, H)



W. Kloppmann^{a,*}, Ido Negev^b, Joseph Guttman^b, Orly Goren^c, Ittai Gavrieli^c, Catherine Guerrot^a, Christine Flehoc^a, Marie Pettenati^a, Avihu Burg^c

^a BRGM, French Geological Survey, 3. av. C. Guillemin, BP 36009 F-45060 Orléans Cedex 2, France

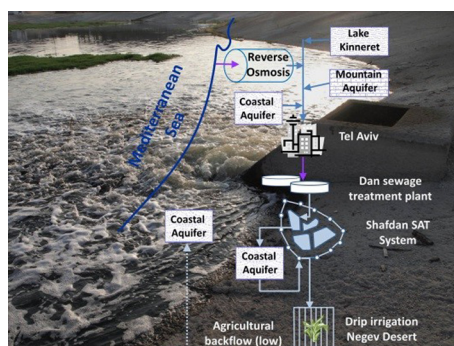
^b Mekorot National Water Company, 9 Lincoln Street, P.O. Box 20128, Tel-Aviv 6713402, Israel

^c Geological Survey of Israel, 30 Malkhe Israel St., Jerusalem 95501, Israel

HIGHLIGHTS

- Unconventional water types dominate a regional urbanized water cycle.
- RO-derived freshwater and wastewater can be traced through isotopes.
- Specific fingerprints are found throughout the urban water cycle.
- Desalinated seawater is massively introduced into the environment.

GRAPHICAL ABSTRACT



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ABSTRACT

“Man-made” or unconventional freshwater, like desalinated seawater or reclaimed effluents, is increasingly introduced into regional water cycles in arid or semi-arid countries. We show that the breakthrough of reverse osmosis-derived freshwater in the largely engineered water cycle of the greater Tel Aviv region (Dan Region) has profoundly changed previous isotope fingerprints. This new component can be traced throughout the system, from the drinking water supply, through sewage, treated effluents, and artificially recharged groundwater at the largest Soil-Aquifer Treatment system in the Middle East (Shafdan) collecting all the Dan region sewage. The arrival of the new water type (desalinated seawater) in 2007 and its predominance since 2010 constitutes an unplanned, large-scale, long-term tracer test and the monitoring of the breakthrough of desalination-specific fingerprints in the aquifer system of Shafdan allowed to get new insights on the water and solute flow and behavior in engineered groundwater systems. Our approach provides an investigation tool for the urban water cycle, allowing estimating the contribution of diverse freshwater sources, and an environmental tracing method for better constraining the long-term behavior and confinement of aquifer systems with managed recharge.

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1. Introduction

Unconventional water resources such as desalinated water and treated sewage play today a key role in the water balance of many developed countries in arid and semi-arid regions. The main drivers are

* Corresponding author.

E-mail address: w.kloppmann@brgm.fr (W. Kloppmann).

climatic and economic: water resources are under double pressure, from decreasing availability at global and regional scale due to climate changes (Li and Urban, 2016) and from increasing demand in regions with strong demographic, agricultural and industrial development (AghaKouchak et al., 2015). Pressure is further enhanced by poor water management structures (Montenegro et al., 2006). Urbanization in coastal regions of industrialized (semi-) arid countries creates particularly “hot spots” of water stress in terms of both available quantity and quality (Fletcher et al., 2013; McGrane, 2016). Groundwater salinization is an enhancing factor of water quality degradation in such contexts (Cary et al., 2015; Chatton et al., 2016).

Only relatively recently, desalination has become a predominating factor in the freshwater supply of many water-scarce industrialized regions. Between 2004 and 2012, the global desalination capacity has more than doubled (Zotalis et al., 2014), reaching presently 29 Bm³/year. In Israel, desalination provided in 2017 around 85% of the domestic urban water consumption and 40% of the total national water consumption (Marin et al., 2017) with a total desalination capacity of 585 Mm³/year. This massive arrival of desalinated seawater in the water cycle is expected to have an impact on the geochemical and isotope characteristics of surface-derived and groundwater that can be used for monitoring purposes. A pilot study in 2008 (Kloppmann et al., 2008b) has shown the highly specific isotope signatures of reverse osmosis (RO) desalinated seawater, notably $\delta^{18}\text{O}$ and $\delta^2\text{H}$ values close to the original seawater and a significant enrichment in ^{11}B , that can be used to identify and quantify the increasing contribution of RO-derived freshwater in regional water balances.

Another important component of “man-made” water cycles is groundwater; aquifers being increasingly used as storage and purification media via Managed Aquifer Recharge (MAR) and Soil Aquifer Treatment (SAT) (Kazner et al., 2012). Major challenges in the study of such systems are to assess and monitor the penetration of artificially recharged waters into the natural groundwater system and to provide reliable estimations of mixing proportions. Those allow estimating transfer and residence times, crucial factors for the cleaning performance of the system, and provide proof of the confinement of MAR or SAT systems with respect to the surrounding groundwater resources.

Here, again, the use of environmental isotopes as tracers has major assets: (1) a much more sensitive and reliable calculation of mixing than chemical tracers alone due to their low detection limits (Quast et al., 2006), their non-linear mixing behavior (see section “Materials and Methods: Isotope mixing models” in the Supplementary Material), and constancy of source-specific signatures over time, (2) no artificial tracers are introduced in the system, and (3) the use of such intrinsic tracers is not limited in time like artificial tracer tests but allows for long-term integration over time. Though, the main condition of their use is a contrast of the injected and background isotopic fingerprints. Distinguishing injected water from baseline conditions may thus need a multi-isotope approach (Kloppmann et al., 2008a) or a combination of isotopes with persistent chemical tracers (Cary et al., 2013) to get unambiguous results. Compared to artificial tracing (Quast, 2003; Quast et al., 2006), the long term input function of isotope signatures is also often less well known. Some of the isotope signatures can also be altered by (bio)geochemical reactions within the aquifer, e.g. boron (Kloppmann et al., 2015), but such reactions can also alter the concentrations of chemical tracers, especially organic substances (Quast et al., 2006).

Certain isotopes have been widely used to specifically trace waste water in groundwater systems, in particular the stable isotopes of the water molecule, behaving conservatively (Kracht et al., 2007; Negev et al., 2017) and boron isotopes (Cary et al., 2013; Vengosh, 1998; Vengosh et al., 1994; Verstraeten et al., 2005); boron being present in significant concentrations in domestic wastewater with a specific isotope signature (Barth, 1998; Barth, 2000; Gabler and Bahr, 1999; Vengosh et al., 1999).

In the greater Tel-Aviv area (Dan region), the focal point of our study, most of the previously mentioned factors of water stress

superimpose (Thi Hoang Duong et al., 2011), notably low availability of natural freshwater, urbanization of catchments, industrialization, demographic rise, as well as being situated on coastal aquifers, affected by salinization. Yet, to cope with regional water stress, Israel looks back to 60 years of experience of integrated water resources management combining water sources development, water transport, rain harvesting, waste water reuse and desalination with conservation measures (Tal, 2006). Large-scale water transport, relying on the National Water Carrier (NWC) as its backbone, water recycling and artificial recharge of groundwater reservoirs as well as desalination as major freshwater source resulted in a largely engineered water cycle at national and regional scales. Our current study investigates the breakthrough of desalinated seawater within the regional urban and largely engineered water cycle of the greater Tel Aviv (Fig. 1), following its isotope signatures through the drinking water supply, via waste water collection, treatment and recharge (SAT), and the groundwater reservoir of the Coastal aquifer. The objective of this study is thus to monitor conventional (surface and groundwater) and unconventional (treated sewage, desalinated seawater) components of freshwater supply throughout the water supply system and in the aquifer by using intrinsic tracers (isotopes of B, H + O_{H2O}, S + O_{SO4}) and to investigate their usefulness for the regional water management in urbanized water cycles. A particular focus lies on the SAT site of Shafdan, the largest system of this type in the Middle East (Goren et al., 2014; Goren et al., 2012; Oren et al., 2007), recharging the Coastal aquifer, itself pumped for irrigation in the Negev desert. Here, a crucial point of water management is to control the confinement of the infiltration-recovery system with respect to the surrounding natural unaffected groundwater.

2. Materials and methods

2.1. Site description

The Dan Region Metropolitan water supply is assured by the NWC and by private groundwater wells (Fig. 1, Fig. S1, Supporting Information). Before the desalination era, the sources of the water supply were from the two major groundwater resources of Israel, the sandy Coastal aquifer and the carbonate Mountain aquifer, as well as from the fresh Lake Kinneret (Sea of Galilee). In the last few years, the percentage of freshwater from seawater desalination in the total water supply to the NWC has increased to about 70% while the rest is from local wells pumping from the Coastal and Mountain aquifers (Negev et al., 2017). The contribution of water from Lake Kinneret in the past (before the establishment of the Mediterranean desalination plants) was quite high but since 2005 it has been reduced in parallel to the gradual increase in the volume of the desalinated water. Today the contribution of Lake Kinneret to the NWC is <15%, and is almost negligible in the Dan Region.

The Dan Region Reclamation Plant (Shafdan) reclaims approximately 135 Mm³/y of secondary treated wastewater from the Dan Metropolitan area and several other neighboring municipalities. The influent wastewater, collected from the Dan Region, consists of $\approx 90\%$ domestic sewage and $\approx 10\%$ industrial sewage. After secondary treatment, effluents are recharged for tertiary treatment into a sandy aquifer via a SAT system with a retention time that has been estimated at 0.5 to 60 months (Negev et al., 2017). The plant consists of 6 infiltration basins (each comprising a number of operation ponds), covering a total area of ~ 110 ha. Retention times for individual pumping wells strongly depend on the position of the wells with respect to the associated infiltration ponds and on the local flow regime.

The Shafdan infiltration basins are located 3–4 km inland of the coastline, above the Coastal aquifer of Israel. This aquifer stretches over a length of about 100 km with a width of 15 to 30 km along the Mediterranean coast, comprising Quaternary alternating units of sand, calcareous sandstones, loams and clays. In the Shafdan area, the aquifer is topped by a thick unsaturated zone (20–40 m) which plays a decisive

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