

# Use of Strontium Isotopes to Identify Buried Water Main Leakage Into Groundwater in a Highly Urbanized Coastal Area

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Previous studies indicate that the local aquifer systems in the Mid-Levels, a highly urbanized coastal area in Hong Kong, have commonly been affected by leakage from water mains. The identification of leakage locations was done by conventional water quality parameters including major and trace elements. However, these parameters may lead to ambiguous results and fail to identify leakage locations especially where the leakage is from drinking water mains because the chemical composition of drinking water is similar to that of natural groundwater. In this study, natural groundwater, seepage in the developed spaces, leakage from water mains, and parent aquifer materials were measured for strontium isotope ( $^{87}\text{Sr}/^{86}\text{Sr}$ ) compositions to explore the feasibility of using these ratios to better constrain the seepage sources. The results show that the  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios of natural groundwater and leakage from water mains are distinctly different and thus, they can provide additional information on the sources of seepage in developed spaces. A classification system based on the aqueous  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio is proposed for seepage source identification.

## Introduction

Losses of water in buried distribution networks can reach high percentages of the volume introduced. In European countries, the network losses range from 3% in Germany to over 40% in Slovenia (1). Lerner (2) suggested that rates of 20–25% leakage from water mains are common, and rates up to 50% have been observed, causing large amounts of groundwater recharge. In Hong Kong, where the government spends more than HK\$2,500 million each year for bulk purchase of fresh drinking water in East River (Dongjiang) from Guangdong Province, China (3), about 23% of the fresh water is reported to be lost underground because of leakage from water mains (4). Thus, identification of leakage locations and reduction of the leakage are major goals of the Hong Kong authorities.

The lower slope of the Mid-Levels area in Hong Kong is one of the most heavily urbanized coastal areas in the world (Figure 1). Due to the rugged topography, urbanization has created thousands of cut slopes and retaining walls. Weep-holes and horizontal drains have been installed to lower the high groundwater tables behind the slopes and walls. Seepage from some of the weepholes and horizontal drains is obvious. Because leakage from water mains could lead to localized

high water tables, which may negatively affect the slope stability, the leakage problem has long been a major concern in the study area.

In the late 1970s, the Geotechnical Control Office (GCO), now Geotechnical Engineering Office (GEO), of the Hong Kong Government conducted a groundwater chemical survey aiming to identify the possible leakage locations. The results suggested that substantial amounts of leakage from water mains occurred throughout the study area (5). In that survey, several simple water quality parameters, including electrical conductivity (EC), fluoride ( $\text{F}^-$ ), chloride ( $\text{Cl}^-$ ) and ammonia nitrogen ( $\text{NH}_3\text{-N}$ ) were used for leakage identification. The drawbacks of the above parameters were discussed by GCO (5) and Lerner (2).

Between 2001 and 2004, the Department of Earth Sciences of the University of Hong Kong carried out a comprehensive groundwater chemical study in the Mid-Levels area aiming to investigate the impacts of urbanization on groundwater chemical systems. Based on geochemistry, it was shown that the water mains were still leaking, but not as badly as before (6–9). Leung et al. (7) found that almost all groundwater samples in the developed spaces were of the Na–Cl or Na–Ca–Cl water type and had cation/chloride ratios similar to those of seawater. As seawater has long been used for flushing in the area, the results suggested that even a small amount of leakage from flushing water pipes could significantly change and mask the groundwater chemical signatures. This makes it difficult to identify natural groundwater seepage in the developed spaces based solely on conventional water parameters (8).

Previous studies have demonstrated that certain radiogenic isotopes, including strontium isotopes, can be used as environmental tracers (10–12). In addition, strontium isotopes can also be used to provide information on the mixing of water bodies and to trace groundwater with different sources (13–15). This is possible because the aqueous  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios depend on the  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios of the aquifer materials with which the water interacts. Moreover, natural processes do not fractionate  $^{87}\text{Sr}/^{86}\text{Sr}$  and the measured differences in  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios are caused by mixing of Sr derived from various sources with potentially different  $^{87}\text{Sr}/^{86}\text{Sr}$  compositions (16).

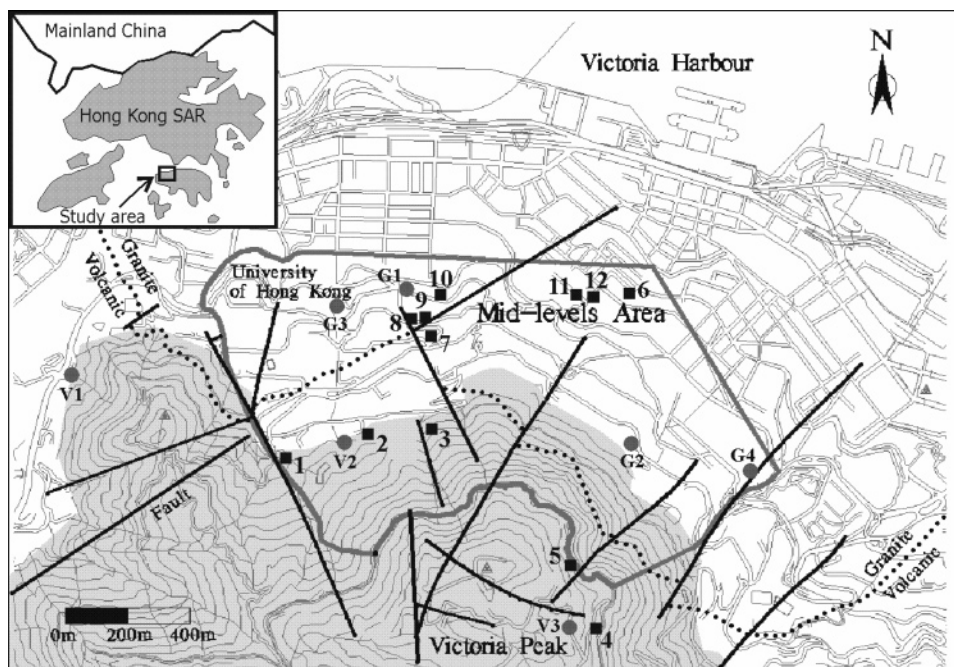
In this study, a number of representative water and parent rock samples were selected, based on the results of previous studies of the Mid-Levels area (6–8), for further  $^{87}\text{Sr}/^{86}\text{Sr}$  analysis. The overall objective was to investigate whether the aqueous  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios can provide additional information to better constrain the seepage sources in the study area. In this paper, “seepage” refers to water running from horizontal drains or weepholes in the developed spaces. “Leakage” refers to water leaked from water mains such as flushing or drinking water pipes.

## Experimental Section

**Geology and Hydrogeology of the Study Area.** The Mid-Levels area, approximately 1.5 km<sup>2</sup> in size, is situated on the northern slope of Victoria Peak (550 m above mean sea level) on Hong Kong Island (Figure 1). The study area can be divided into two parts with significantly different modes of development. The upper part of the area (>170 m above mean sea level) is essentially a natural slope with minimum development. In contrast, the lower part of the area is one of the most heavily urbanized spaces in the world.

The geology and hydrogeology of the area have been described elsewhere (5). In brief, the geology is dominated by two rock types; silicic volcanic rocks and a granitic

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**FIGURE 1.** Overview of the study area. The shaded area represents the natural slopes with minimum development. The area bounded by the gray line is the Mid-levels area. Dotted lines represent the contact between granite and volcanic rocks. Black solid lines represent the locations of normal faults. The black squares represent the water sampling locations. The gray circles represent the parent rock sampling locations.

intrusion. The volcanic rocks have undergone low-grade regional metamorphism and deformation and were affected by contact metamorphism associated with intrusion of the granite. Both lithologies were later intruded by basaltic dykes. In general, colluvium overlies several meters of decomposed rock above the bedrock. GCO (5) has grouped the soil and rock into three aquifer units corresponding to (a) colluvium, (b) decomposed volcanic and granite rocks, and (c) volcanic and granite bedrocks. The colluvium contains transient and permanent perched water tables, whereas, as recently demonstrated by Jiao et al. (17–19), the highly decomposed rocks or saprolite below the colluvium is relatively less permeable due to its high content of clay. The zone along the uppermost part of the bedrock may be fairly permeable with confined groundwater contained within a well-developed fracture network.

**Methodology.** Fourteen representative water samples were selected for analysis of strontium isotopes. These samples are considered to represent natural groundwater, seepages mixed with leakage from water mains to various extents, and leakage from water mains. In addition, 7 fresh bedrock samples, 4 granites and 3 volcanic rocks, were measured for  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios. The bedrock samples were collected from cores drilled for other purposes. The bedrock samples were used to assess the isotopic contribution of the parent rock material to the waters in contact with them. Figure 1 shows the locations of the water and rock samples. Drinking water and flushing water samples were collected from a water tap and flushing water tank, respectively, within the campus of the University of Hong Kong (HKU) for  $^{87}\text{Sr}/^{86}\text{Sr}$  measurements.

The analytical procedures for the strontium isotopic measurements followed standard methods. The rock samples were powdered and then digested in a mixture of concentrated  $\text{HNO}_3$  and HF. Sr was separated from the digested solutions and water samples using an Eichrom Sr-Spec resin column and  $\text{HNO}_3$  solution media. The isotopic ratios of rock samples were measured by a thermal ionization mass spectrometer (TIMS) (model MAT261) made by Finnigan, Germany, with a multi-collector system in the Isotope

Laboratory, Faculty of Earth Sciences at China University of Geosciences, Wuhan, China. The accuracy of the machine was evaluated by measurement of the NBS987 which gave a mean  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio of  $0.710258 \pm 0.000012$  (2 standard deviations) ( $n = 26$ ). The duplication analysis of BCR-2 standard gave a mean  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio of  $0.705018 \pm 0.000008$  (2 standard deviations) ( $n = 10$ ). Measured  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios were corrected for mass fractionation relative to  $^{86}\text{Sr}/^{88}\text{Sr}$  ratio of 0.11940. The isotopic ratios of water samples were measured using an IsoProbe multi-collector inductively coupled plasma mass spectrometer (MC-ICPMS) (GV Instruments Ltd., UK) at the Guangzhou Institute of Geochemistry of the Chinese Academy of Sciences. The mass bias (fractionation) of  $^{87}\text{Sr}/^{86}\text{Sr}$  during the measurement was internally calibrated by exponential law using the  $^{86}\text{Sr}/^{88}\text{Sr}$  ratio of 0.11940. Errors are quoted throughout as 2 standard deviations from the measured or calculated values. During the course of the isotopic analysis, 15 measurements of NBS987 were obtained, yielding an average  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio of  $0.710305 \pm 0.000034$  (2 standard deviations). The total chemical blank was less than 0.1 ng for Sr. The water samples were also measured for physiochemical parameters during collection at the sampling sites and were then sent to HKU for analysis of major, trace, and rare earth element (REE) concentrations, using the standard methods of AWWA (20). The detailed analytical procedures and results were described by Leung (6). In this study, only the concentrations of relevant elements are presented.

## Results and Discussion

### Strontium Isotope Ratios of the Host Aquifer Materials.

Table 1 shows the  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios obtained for the granite and volcanic rock samples. The granite samples have ratios ranging from 0.720263 to 0.764090 with an average of 0.733230, whereas the volcanic rocks have ratios ranging from 0.713150 to 0.744028 with an average of 0.723791. Thus, the granites have somewhat higher  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios than the volcanic rocks. Generally, the  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios of the host aquifer materials are well above 0.713000.

**TABLE 1. Summary of Parent Rock Samples in the Mid-Levels Area (mbs = Meters Below Surface)**

sample	rock type	depth (mbs)	<sup>87</sup> Sr/ <sup>86</sup> Sr	± 2 σ SE
G1	medium grained granite	35.58	0.727309	0.000013
G2	medium grained granite	12.00	0.721256	0.000014
G3	medium grained granite	11.00	0.764090	0.000011
G4	medium grained granite	37.70	0.720263	0.000017
V1	fine ash crystal tuff	10.36	0.744028	0.000016
V2	fine ash crystal tuff	unknown	0.714196	0.000010
V3	course ash crystal tuff	30.70	0.713150	0.000013

**Strontium Isotope Ratios and Chemical Compositions of Water Leaking from Water Mains.** It has been shown by Leung et al. (7) and Leung and Jiao (9) that in the study area the amount of leakage from sewage pipes and stormwater drains is insignificant compared to that from flushing water and drinking water pipes. Samples of leakage from flushing water and drinking water pipes were collected and analyzed for <sup>87</sup>Sr/<sup>86</sup>Sr compositions. Because seawater has long been used for flushing in the study area, the major element concentrations of the flushing water sample are generally close to seawater values, which are significantly higher than those of natural groundwater. The Na/Cl and Mg/Cl ratios of the sample are 0.55 and 0.07, respectively, and the <sup>87</sup>Sr/<sup>86</sup>Sr ratio of the sample is 0.709003, similar to that of modern seawater (0.70916) (21).

As shown in Table 2, the concentrations of strontium and major ions, including Na, Mg, and Cl ions, in the drinking water sample are within the range of the groundwater samples in the natural slopes. However, the two types of water can be easily distinguished by their <sup>87</sup>Sr/<sup>86</sup>Sr ratios. The drinking water sample has a ratio of 0.709341, which is significantly lower than that of the natural groundwater (higher than 0.711000, to be discussed in the next section).

On the basis of these data, seepage samples from weep-holes and horizontal drains that originated mainly from nearby leaking water mains should have <sup>87</sup>Sr/<sup>86</sup>Sr ratios close to the range of 0.709003 to 0.709341, due to minimal water-rock interactions over a short flow path. This assumption is proved to be valid as discussed in the latter section.

**Strontium Isotope Ratios and Chemical Compositions of Groundwater in the Natural Slopes.** Five groundwater samples were collected from the natural slopes for analysis of Sr isotopes. The <sup>87</sup>Sr/<sup>86</sup>Sr ratios and the relevant compositions of these samples are summarized in Table 2. Groundwater in the natural slopes has a wide variety of water types

and contains Na/Cl and Mg/Cl ratios significantly higher than those of water from leaking water mains. Most of the samples have low total dissolved solids (TDS). The <sup>87</sup>Sr/<sup>86</sup>Sr ratios of the natural groundwater samples range from 0.711579 to 0.716691 with an average of 0.713403. These values are in reasonable agreement with the values of the host volcanic rock, suggesting that the natural groundwater developed <sup>87</sup>Sr/<sup>86</sup>Sr signatures similar to those of the host aquifer by water-rock interactions. The <sup>87</sup>Sr/<sup>86</sup>Sr ratios of natural groundwater and water leaking from water mains are distinctly different. The natural groundwater has ratios generally higher than 0.711000, whereas, leaking flushing water and drinking water have ratios of 0.709003 and 0.709341, respectively. Thus, these values can be used to identify potential natural groundwater outcrops in the developed spaces.

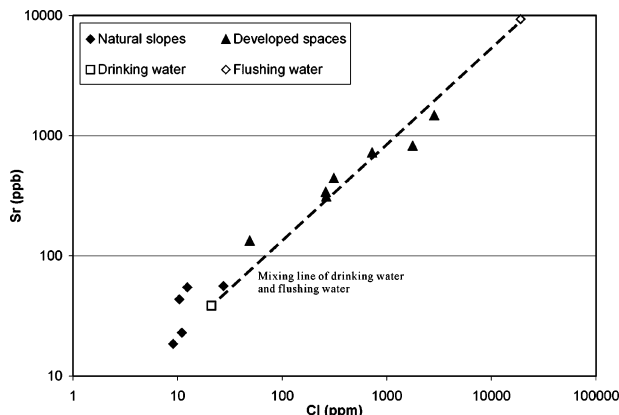
**Strontium Isotope Ratios and Chemical Compositions of Seepage in the Developed Spaces.** Seven representative samples of seepage water were collected in the developed spaces for additional <sup>87</sup>Sr/<sup>86</sup>Sr isotopic determinations. Table 2 summarizes the isotopic ratios and the key chemical features of these samples. See Leung (6) and Leung et al. (7) for detailed chemical data on the seepage waters.

Seepage samples in the developed spaces are dominated by Na-Cl and Na-Ca-Cl water types and contain TDS ranging from 275 to 5331 ppm. The concentrations of major ions and strontium are significantly higher than those in waters from the natural slopes. The samples have Na/Cl and Mg/Cl ratios close to those of the leaking flushing and drinking waters as shown in Table 2. As can be seen in Figure 2, Sr is strongly correlated with Cl in all the seepage samples in the developed spaces (*R* = 0.966), indicating that seawater (flushing water) imposes a significant influence on the distribution of aqueous strontium in the developed spaces. These findings suggest that all of the 7 seepage samples were composed of leaking flushing water in various extents. As mentioned earlier, seepage samples in the developed spaces are extremely sensitive to small fractions of flushing water. Although some of the seepage samples may mainly come from natural groundwater, it is difficult to identify these locations solely based on the major and trace element compositions. However, the aqueous <sup>87</sup>Sr/<sup>86</sup>Sr ratios appear to be able to provide some insights on this.

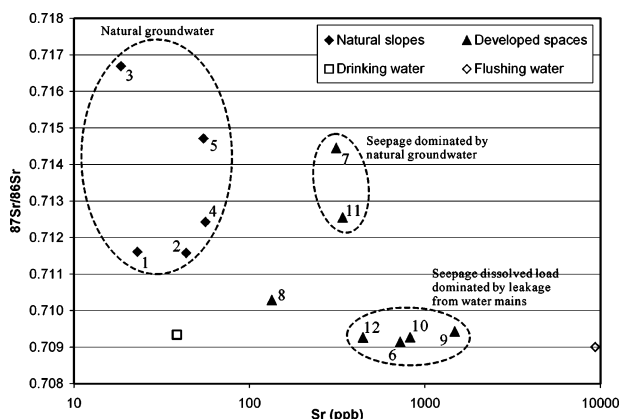
As can be seen in Table 2, seepage samples at locations 7 and 11 have Na/Cl and Mg/Cl ratios very close to those of water leaking from water mains, but their <sup>87</sup>Sr/<sup>86</sup>Sr ratios of 0.714442 and 0.712548, respectively, are similar to the values

**TABLE 2. Summary of Selected Chemicals in Water Samples in the Study Area (Concentrations Are Expressed in ppm Unless Otherwise Specified; n.d. = Not Determined)**

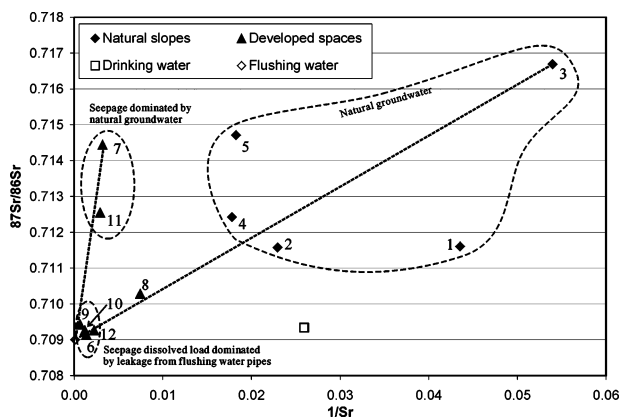
sample	water type	F <sup>-</sup>	Na <sup>+</sup>	Mg <sup>2+</sup>	Cl <sup>-</sup>	NO <sub>3</sub> <sup>-</sup>	Sr (ppb)	Na/Cl	Mg/Cl	TDS	<sup>87</sup> Sr/ <sup>86</sup> Sr	± 2 σ SE
<b>Natural Slopes</b>												
1	Na-HCO <sub>3</sub> -Cl	0.16	10.18	1.10	10.99	1.84	22.97	0.93	0.10	66.62	0.711608	0.000041
2	Na-Ca-HCO <sub>3</sub> -Cl	0.26	10.92	1.05	10.41	1.87	43.57	1.05	0.10	91.19	0.711579	0.000055
3	Na-Ca-Cl-HCO <sub>3</sub> -SO <sub>4</sub>	0.10	7.59	1.09	9.06	2.79	18.53	0.84	0.12	57.70	0.716691	0.000053
4	Ca-Na-Cl-HCO <sub>3</sub> -SO <sub>4</sub>	0.42	21.85	2.92	27.43	19.44	56.19	0.80	0.11	173.61	0.712427	0.000045
5	Ca-Na-HCO <sub>3</sub> -SO <sub>4</sub>	0.35	15.06	4.08	12.41	8.43	54.75	1.21	0.33	145.42	0.714712	0.000031
<b>Developed Spaces</b>												
6	Na-Cl	n.d.	396.91	46.75	722.80	28.15	722.90	0.55	0.06	1673.30	0.709138	0.000018
7	Na-Ca-Cl	0.14	137.09	13.84	263.85	11.00	312.00	0.52	0.05	593.03	0.714442	0.000021
8	Na-Ca-Cl-HCO <sub>3</sub>	0.28	38.01	3.79	48.96	21.12	134.10	0.78	0.08	275.39	0.710283	0.000018
9	Na-Cl	n.d.	1448.45	186.88	2844.39	9.00	1477.00	0.51	0.07	5331.71	0.709423	0.000045
10	Na-Cl	0.30	894.38	100.76	1766.42	11.90	823.10	0.51	0.06	3353.81	0.709264	0.000042
11	Na-Ca-Cl	0.33	140.11	16.21	260.61	17.18	340.00	0.54	0.06	664.80	0.712548	0.000037
12	Na-Ca-Cl	0.20	189.10	20.83	312.42	12.18	443.20	0.61	0.07	799.07	0.709263	0.000022
<b>Drinking Water</b>												
not numbered	Ca-Na-HCO <sub>3</sub> -Cl	0.48	11.00	1.61	21.07	6.76	38.55	0.52	0.08	124.57	0.709341	0.000058
<b>Flushing Water</b>												
not numbered	Na-Cl	1.30	10500	1350	19000	0.50	9334.00	0.55	0.07	32585	0.709003	0.000027



**FIGURE 2.** Plot of strontium concentrations versus chloride concentrations of water samples in the Mid-Levels area.



**FIGURE 3.** Plot of  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios versus strontium concentrations of water samples in the Mid-Levels area.



**FIGURE 4.** Plot of  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios versus  $1/\text{Sr}$  of water samples in the Mid-Levels area. The two dotted lines represent two mixing lines, one between the “natural groundwater” field and the “flushing water” end member, the other between the “seepage dominated by natural groundwater” field and the “flushing water” end member.

of natural groundwater. Thus, the seepage samples at these two sites probably contain significant quantities of natural groundwater (Figure 3). And, it is likely that the seepage dissolved loads at these two locations are mainly derived from water–rock interactions. This interpretation is further supported by the fact that location 11 is a permanent spring. The high TDS and Na/Cl and Mg/Cl ratios of these samples may also imply that the natural groundwater at these sites was slightly contaminated by least amounts of leakage of saline flushing water. Furthermore, according to Figure 4, it seems that the seepages at locations 7 and 11 are mixing toward a different natural groundwater end member than

**TABLE 3.** Possible Range of  $^{87}\text{Sr}/^{86}\text{Sr}$  Ratios of Seepage Samples with Different Mixing Degrees of Leakage from Water Mains in the Mid-Levels Area in Hong Kong

	$^{87}\text{Sr}/^{86}\text{Sr}$ ratio
seepage samples mainly derived from natural groundwater with least amounts of leakage from water mains	$>0.711000$
seepage samples mixed with a small amount of leakage from water mains	$0.709500\text{--}0.711000$
seepage dissolved load dominated by leakage from water mains	$<0.709500$

represented by the upslope samples. The results appear to suggest that the end member has the same or slightly less radiogenic Sr isotopic composition and a higher Sr concentration than the upslope natural groundwater end member (for example, sample at location 3). The difference in the Sr isotopic characteristics between the two natural groundwater end members could be related to the source geology, weathering phenomenon, or cation exchange (22–24). In this preliminary investigation, only 14 water samples were collected for Sr analysis. Further studies are required to better explain the possible causes of the Sr isotopic differences observed.

The other four seepage samples, collected at locations 6, 9, 10, and 12, have significantly lower  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios ranging from 0.709138 to 0.709423, values very close to those of water from leaking water mains. This further demonstrates that the Sr isotopic composition of leakage waters would not be changed because of the minimal water–rock interactions over a short flow path. In other words, seepage waters that have more radiogenic Sr isotopic compositions would likely be dominated by natural groundwater. It is further suggested in Figure 4 that seepage samples at locations 6, 9, 10, and 12 are mixed with flushing water to a larger extent than samples at locations 7 and 11, with relatively little natural water. Although the seepage sample at location 12 has a TDS level and chemical composition similar to those at locations 7 and 11, its distinctly lower  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio indicates that its dissolved load is mainly derived from a different source, most likely from leaking flushing water mains. The seepage sample from location 8 has an intermediate  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio of 0.710283, suggesting that it is moderately mixed with leakage from water mains with the amount smaller than that at locations 6, 9, 10, and 12, but larger than that at locations 7 and 11.

Our results suggest that in the Mid-Levels area of Hong Kong, seepage samples in developed spaces that mainly derived from natural groundwater should have  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios higher than 0.711000, whereas those, in terms of seepage dissolved loads, dominated by leakage from water mains should have ratios lower than 0.709500. Seepage with intermediate  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios, in the range of 0.709500 and 0.711000, probably suggests that it is mixed with some leakage waters, although not in a significant amount. Further studies on the stable isotopes of  $\text{H}_2\text{O}\text{--}\delta^{18}\text{O}$  and  $\text{H}_2\text{O}\text{--}\delta^2\text{H}$  would be helpful to estimate the proportions of leakage waters in the seepage samples in the developed spaces. Table 3 summarizes the possible range of  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios of seepage samples in the developed spaces with different mixing degrees of leaked water. Although limited in the number of samples, our results appear to suggest that there are at least two natural groundwater end members in the study area. These results should assist local government authorities, such as the Water Supplies and Drainage Services Departments, to identify locations with potential leakage. We recommended that all seepage samples in the developed spaces studied in this project be

analyzed for their  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios and stable isotopes of water ( $^{18}\text{O}$  and  $^2\text{H}$ ) in order to better evaluate the hydrogeological setting and the variable degrees of leakage.

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