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Biogeosciences Discussions

Interactive comment on "Tracking the direct impact of rainfall on groundwater at Mt. Fuji by multiple analyses including microbial DNA" *by* Ayumi Sugiyama et al.

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Reply to Anonymous Referee #1 Received and published: 26 April 2016

Comment 1 (General): For example, the introduction introduces the special case of Mt. Fuji in the middle of a series of paragraph that talk about different methods used to trace groundwater flow times. For the reader, it would be much easier to follow if the authors first introduced the overall problem (using chemical and isotopic properties to separate groundwater and streamflow into sources that have a range of residence times), and why Mt Fuji is a good laboratory to test new methods (including DNA). Then a description of the methods that have been applied to trace groundwater flow and estimate residence times, including their shortcomings (i.e. they only provide averages

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in most cases). Then how extreme events can be used in concert with these methods to indicate the rate and magnitude of response (i.e. the current study).

Reply 1: Thank you for your critical reading of our manuscript and kind suggestions. We herein try to revise the previous Introduction according to your suggestion to be easily understood by readers. (1) Adding one short sentence in between after the second paragraph. (2) Then we explain why Mt. Fuji was selected for this study.

Comment 2 (General): Two issues I found I did not fully understand in this study. First, the stable isotopes ofwater in precipitation will likely differ for high rainfall events from long term averages (i.e. they should be heavier at all elevations). These are given in Table 1, but it was not really clear to me how the conclusions about where the water was coming from were based on actual rainfall measurements during the storm events at the various elevations.

Reply 2: Average discharge height for the examined groundwater at the foot of Mt. Fuji is estimated from 1,500m to 2,000m (Discussion, First paragraph). It was not possible to separately discuss delta18O taken from rainwater for individual groundwater at each event. Thus, delta18O of rainwater was expressed by the values measured for all water sampled at five different sites from R1 (2,364m) to R5 (723m), which almost cover the whole expected precipitation.

Comment 3 (General): Second, Table 1 shows that dissolved O2 levels are still about 80% of saturation in the deep aquifer. Yet the Archea being flushed out are obligate anaerobes. One could take this to mean that there are anaerobic 'pockets' that are not measured under average conditions, and only get flushed out when there is an extreme event. This would be supported if there were a strong decline in DO (and an increase in Na+) in the waters that have high Archea âĔŸAĔĞT is that the case? If not, are there other supporting data indicating that these waters are flushing out cracks?

Reply 3: Thank you very much for your critical reading the data. Yes, you're right and we need to explain the measurement procedure precisely. The physical and chemical properties of the 550m water were not measured directly. The measurement was conducted for the water after it was pumped up. We add this explanation at the bottom of Table 1.

Comment 4 (General): The Wei(Wels?) et al. paper (and others indicating piston flow water residence times/flows) should be introduced first, when discussing the site itself, rather than very late in the paper, so that the reader does not get confused about the flow paths and water sources (at least what was initially though) for G1-G4. What is the source for the arrows indicating flow direction in Figure 1?

Reply 4: Flow direction of water was suggested by the simulation model of ãĂĂGET-FLOWS as was explained briefly in the 2. Materials and methods, 2.1 Study site. See the detail by Kato et al. (2015) in References. Comment 5 (General): It was hard for me as a reader to keep these sites and their difference s straight âËŸAËĞT perhaps the name could be changed to include more information .e.g. G1 could be Spring1-0m, G2 could be Groundwater well 1- and give depth, etc. That would help the reader remember that one is a deep well, one a shallow well, etc.

Reply 5: Thank you for your suggestion. In order to be easily understood the study sites, we change the name of study sites in Fig.1 and others as follows; G1:SP-0m, G2: GW-42m, G3: GW-550m. Revised Fig.1 is attached separately.

Comment 6 (Specific) : Page 2, Line 12. The term “runoff processes of groundwater” seems a little strange (to a non-hydrologist). I think of runoff as a process mostly associated with overland flow (i.e. not groundwater). Perhaps a less confusing expression of the same idea could be“Effect of rainwater on groundwater”

Reply 6: Thank you for your suggestion. We accept your suggestion and change the wording accordingly.

Comment 7 (Specific): Page 2, Line 23. Start of the sentence should read “Our ongoing microbiological study: : :”

Reply 7: Thank you for your suggestion. We change the wording accordingly.

Comment 8 (Specific): Page 2, Line27. " This depth is far below the lava layer that was taken to be a substantial pool of groundwater." I do not understand this sentence. If there is a lava layer closer to the surface, why could that not the source of warmer water and thermophiles? Also, I think what is meant here is that the lava layer formed a barrier to infiltration and thus provided the base for an aquifer? There should be a reference given for the geotherm at Mt. Fuji.

Reply 8: The lava has been considered as a great reservoir of groundwater in Mt. Fuji (Tsuchi 2007;we add this reference), which comprised of rocks with high porosity. The depth of those lava is unveiled by 3-D visualized map (Uesugi, 2009) from 100 to 200m, thus the depth gives 4 to 8 âĎČ higher temperature than the surface water, which is not sufficient for living of thermophilic microbes.

Comment 9 (Specific): Page 3, line 30. "Grouundwater discharge was measured". Does this mean the spring waters sampled? Is the assumption that all groundwater eventually leaves as springs? IN figure 1, it is almost impossible to understand where G1 - G4 are in relation to each other. It is only in the figure caption that we know these are either spring water or shallow wells.

Reply 9: Thank you for your comment. The sentence of p3, line30 is deleted as this is not referred hereafter. Fig.1 is revised to be more clearly indicated the site studied. See revised Fig.1 attached separately.

Comment 10 (Specific): Page 4, lines 9-13. The authors should mention if there was any treatment to remove DOC from water (and if not, what the DOC concentration range was). Organic C has been shown to affect the analysis (see REF).

Reply 10: Thank you for your precise comment on the method employed. The recommended pre-treatment might be necessary, if the water contains significant amount of dissolved organic compound which vaporize when the examined water is treated as extract from plant tissue or beverage. We did not employ such treatment in this study. For further study we'll try to check it to ensure the measurement.

Comment 11 (Specific): Page 5, line 23. " We studied four rainfall events from 2012 to 2014 at the foot of Mt.Fuji." Did the rain only fall at the foot of Mt Fuji, or do you mean that you sampled springs and rivers at the foot of Mt Fuji? Probably precipitation intensity varied with altitude âĖŸAĖĞT were the measurements of rainfall amount given made at the base of Mt. Fuji?

Reply 11: The referred precipitation was represented by the measurement at Shiraito-no-taki station of Japan Weather Association (530m a.s.l.), which is the sole official observation site of rain- and snow-fall in the area studied. The location of Shiraito-no-taki station is shown in the revised Fig. 1.

Comment 12 (Specific): Cn you give an estimate of the volume of groundwater compared tot he volume of rainfall? (Even based on the average residence time and the annual rainfall?) It seems the largest events are flushing a large fraction of the groundwater out âĖŸAĖĞT does that make sense compared to the other estimate based on 'average' conditions?

Reply 12: It's not very possible to calculate precisely the mass balance between rainfall and the amount of groundwater. Daily amount of whole spring water at the foot of Mt. Fuji is estimated to be 2.5 million tons (this sentence is added to the last sentence of the third paragraph at p2, in Introduction).

Comment 13 (Specific): Page 6, line 6. At what elevation was the isotopic composition of rainwater measured? In Table 1 it is indicated that rainwater was measured at several elevations. Obviously it is heavier than the groundwater, so the inference is that most groundwater source is at higher elevation? If the rain water is measured only at the foot of Mt. Fuji, this can tell us about 'local' sources versus groundwater sources, but if there was also a high amount of rainfall at high elevation that would perhaps not be distinguishable?

Reply 13: As was answered to your comment Nr.2, stable isotope signature of rainwater was measured for the water sampled at 5 sites raged from 2,364m to 723m a.s.l. The altitude where the rain water was sampled, thus, higher than the examined spring and groundwater.

Comment 14 (Specific): Figure 2. caption - “the number gives the average value of the var(iable)”. Average over what time scale (most seem outside the range of measurements in the figure?)

Reply 14: Thank you for your comment and we’re sorry that we made misspelling at the end of the legend of Fig. 2. It must be "bar" instead of "var". But, the word must be replaced by an appropriate term as "underline." The numbers shown in Fig. 2 are the mean values of three or four observations indicated by underline.

Comment 15 (Specific): Supplemental material. I think not all readers of Biogeosciences will be familiar with the use of hexadiagrams (they are new to this reviewer). Perhaps the authors could add a brief definition to the figure caption.

Reply 15: Thank you for your comment. We add sentences shown below; Hexadiagram shows major eight ions dissolved in water by their relative abundance.ãĂĂNegative ions are shown in the right side of the figure, while positive ions are in the left. The shape of diagram suggests characteristics of water examined.

Thank you again for your critical reading of our manuscript. We revise related Figures, Table, and Supplemental material as is shown just Fig. 1 below. Kenji Kato, Corresponding author.

Interactive comment on Biogeosciences Discuss., doi:10.5194/bg-2016-78, 2016.

1 Introduction

Groundwater, which comprise about 20% of freshwater in the world, is clearly an essential water resource for human activity. However, the amount of storage in a given hydrologic space and the route of groundwater from recharge to spring is not well understood. Hydrologic studies have attempted to reveal physical properties of groundwater such as (1) locations of groundwater recharge, (2) groundwater residence time, and (3) the route of groundwater in the subsurface environment. Recharge altitude and residence time of groundwater are estimated using environmental tracers such as oxygen and hydrogen stable isotopic ratios (e.g., Mook et al., 1974), and tritium (Gleeson et al., 2016), chlorofluorocarbons (e.g., Dunkle et al., 1993) or ^{36}Cl/Cl (e.g., Milton et al., 2003). A trial estimated the duration of water–rock interaction for a sample by the measurement of ^{87}Sr/^{86}Sr (Nakano et al., 2001). However, the data showed only average values of properties of the groundwater, which had been mixed with water from multiple sources in the subsurface environment. Last, we do not have any appropriate method to estimate the groundwater route.

Runoff processes of groundwater directly affected by rainfall have not been fully explained. However, the runoff process of stream water influenced by rainfall (e.g., Hubert et al., 1969; Tekleab et al., 2014) and runoff peak response time of streams estimated by their flow rate change in Japanese sedimentary and granitic rock basins (Onda et al., 1999; Asai et al., 2001) have been studied. The contribution of rainfall to stream water volume was estimated by those studies, but they did not address the route of groundwater until it affected streamflow.

In order to get indication on the route of groundwater we herein newly applied microbial DNA analysis focusing on heavy rainfall at the foot of Mt. Fuji located in central Japan, which is the largest Quaternary stratovolcano in Japan, with a peak at 3,776 m a.s.l. At the foot of this mountain we previously found that pH of groundwater decreased from 7.29 to 7.02 a few weeks after a typhoon in August and September 2011 (total rainfall was > 800 mm) (Segawa et al. 2015). This decrease of pH was probably influenced by low pH of the rainwater (pH from 4.7 to 6.4; Watanabe et al. 2006). This rapid decrease of pH cannot be explained by piston flow transport of groundwater in which newly supplied water pushes out older water preserved in the subsurface bed (e.g., Bethke and Johnson, 2008). Considering the pH of rainfall at Mt. Fuji, the lowering of groundwater pH suggested that the newly supplied rainwater mixed directly with groundwater over a period of weeks. In addition, our preceding microbiological study of groundwater at the foot of Mt. Fuji furnished a clue to estimate possible groundwater routes by finding thermophilic bacterial DNA in spring water, whose temperature was as low as ~10–15 °C throughout the year (Segawa et al., 2015). Thermophilic prokaryotes are optimally adapted to temperatures > 40 °C. This suggests that at least some of the groundwater source was at a depth of 600 m or greater, based on a temperature gradient of 4 °C/100 m. This depth is far below the lava layer that was taken to be a substantial pool of groundwater (Tsuchi, 2007). Thus, microbial information can help estimate the route of groundwater.

Following the above findings, we tried to estimated the groundwater route with a focus on heavy rainfall, by tracing the signature of direct rainfall impacts. This was done using (i) stable oxygen and hydrogen isotopic analysis to track the movement

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Fig. 1. revised Introduction

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of water molecules, (ii) chemical analysis of silica concentration in groundwater, which indicates its possible dilution by rainwater with low silica concentration, and (iii) microbial analysis including DNA sequencing to estimate the groundwater route, which may include a function extracting microbial particles from the matrix of geologic layers. Whereas stable isotopic and chemical analyses show average values of the water, microbes carried by groundwater suggest the groundwater route prior to examination. To elucidate microbial properties in the studied groundwater, we used total direct counting (TDC) of prokaryotes, catalyzed reporter deposition – fluorescence in situ hybridization (CARD-FISH), 16S rRNA gene-targeted polymerase chain reaction (PCR), denaturing gradient gel electrophoresis (DGGE), and a next-generation sequencing. Here, we first used microbial analysis to reveal the groundwater route in the shallow and deep subsurface environment. Preceding hydrologic studies with microbes only addressed the distribution of anthropogenic pollution in shallow aquifers (e.g., Harvey et al., 2010; Lin et al., 2012).

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Fig. 2. revised Introduction_continue

Table 1. Study site and observed environmental parameters.

Site ID	Site name	Type of water	Altitude (m a.s.l.)	Sampling depth (m)	Observation period	Number of Investigation	Water temperature (℃)	pH	EC (µS cm^{-1})	Eh (Pt) (mV)	DO, degree of saturation (%)
SP-0 m	Shibakawa	Spring water	726		2012/6/15～2014/11/20	n=19	10.1～11.9	5.84～7.35	49.5～128.0	343~497	86.7～92.9
GW-42 m	Yodoshi	Groundwater	150	42	2013/7/2～2014/11/20	n=13	13.8～20.6	6.34～7.24	109.8～161.5	300~478	93.6～100.7
GW-550 m	Aoki	Groundwater	175	550*	2013/7/2～2014/11/20	n=13	13.7～19.8	6.63～8.26	122.7～142.8	246~447	73.0～89.9
R1	Go-gome	Rainwater	2,364		2013/6/17	n=1	22.6	5.30	-	-	-
R2	Kokuyurin	Rainwater	1,431		2013/8/6～2014/10/16	n=8	11.9～22.8	4.04～6.26	-	-	-
R3	Ni-gome	Rainwater	1,081		2013/6/17～2014/10/16	n=11	14.2～23.4	3.94～5.82	-	-	-
R4	Asagiri	Rainwater	850		2013/6/17～2014/10/16	n=15	13.0～31.9	4.12～6.14	-	-	-
R5	Shibakawa	Rainwater	723		2012/10/18～2014/10/16	n=16	10.1～30.8	4.06～6.29	-	-	-

* Measurements were conducted for the water soon after it was pumped up.

Fig. 3. revised Table 1

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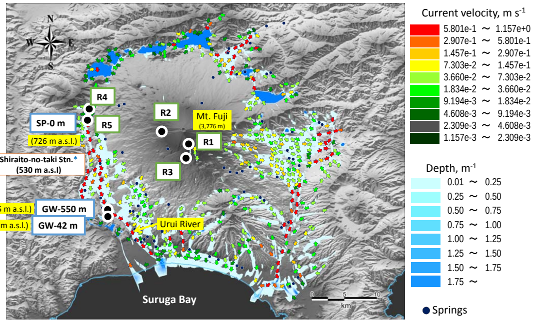

Figure 1. Study sites in western foot of Mt. Fuji. Red arrows indicate main fast flow (GETFLOWS; Kato *et al.*, 2015 partially modified). Precipitation is sampled at R1 to R5. Groundwater is sampled at SP-0 m, GW-42 m and GW-550 m. R1 at 2,364 m a.s.l., R2 at 1,431 m a.s.l., R3 at 1,081 m a.s.l., R4 at 850 m a.s.l. and R5 at 723 m a.s.l.
SP-0 m, Shibakawa at 726 m a.s.l., spring water, GW-42 m, Yodoshi at 150 m a.s.l., shallow well water of 42 m, GW-550 m, Aoki at 175 m a.s.l., deep well water of 550 m. * Amount of precipitation for the studied area was recorded at Shiraito-no-taki Station of Japan Weather Association.

Fig. 4. revised Figure 1