

Natural radon concentrations as indicators for mountain karst aquifer hydrodynamics and seismicity: the case study of Bossea Cave (Piedmont, Italy).

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SUPPLEMENTARY MATERIAL A

This section reports the time series of radon concentrations acquired by the instrumentation inside the *Bossea* Cave: measurements have been conducted in atmosphere since 2003 and in water since 2010. Sampling frequency of each instrument is indicated in Supplementary Table A1 and A2; its position in Supplementary Figure A1. Daily rainfalls and external average temperatures are available on ARPA Piemonte database (<https://www.geoportale.piemonte.it/>). Abbreviations: AG - *AlphaGuard*; SN - *Sun Nuclear*; ATM - atmosphere; AQ - water; ST - *Sala del Tempio*; POR - *Porfiroidi*; UL - Upper Laboratory; TM - *Mora* creek; PA - *Polla delle Anatre*; Ext T. - average external daily temperatures.

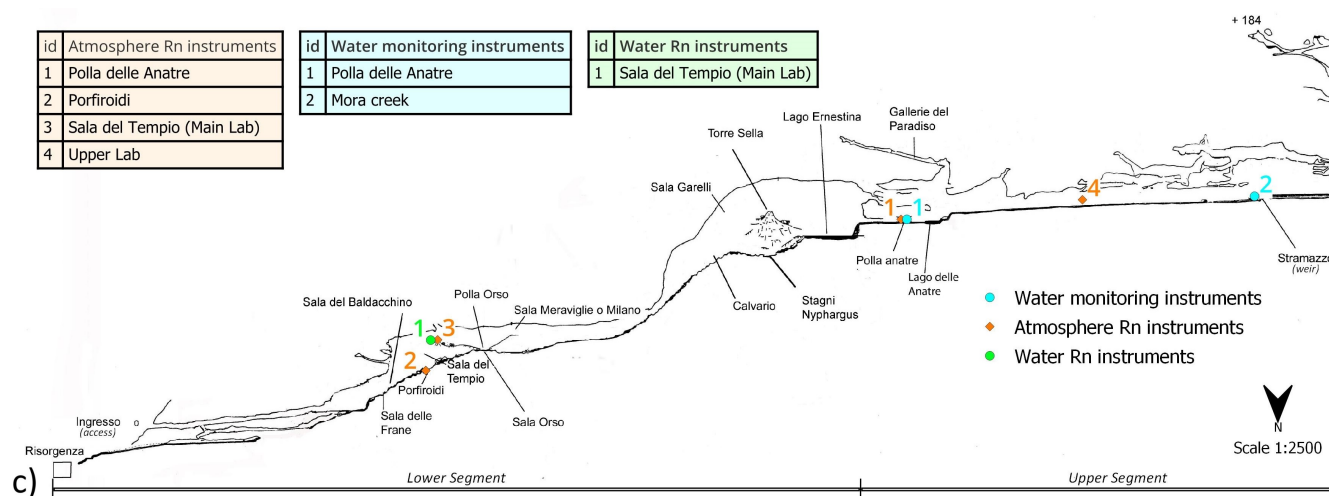
Instrument	Time window	Sampling frequency
<i>AlphaGuard AG1450</i> Sala del Tempio (Main Lab)	Jul 2004 → <i>working</i>	10'
<i>AlphaGuard AG0156</i> Upper Laboratory	Mar 2019 → Mar 2021	From 10' to 60'
<i>AlphaGuard AG0283</i> Upper Laboratory	Mar 2021 → Oct 2022	From 10' to 60'
<i>Sun Nuclear</i> Sala del Tempio (Main Lab)	Jul 2004 → Dec 2014 (*)	24 h
<i>Sun Nuclear</i> Porfiriodi	Jul 2004 → Dec 2021 (*)	24 h
<i>Sun Nuclear</i> Upper Laboratory	Dec 2004 → Aug 2020 (*)	24 h

(*): dataset with significant gaps during the indicated period

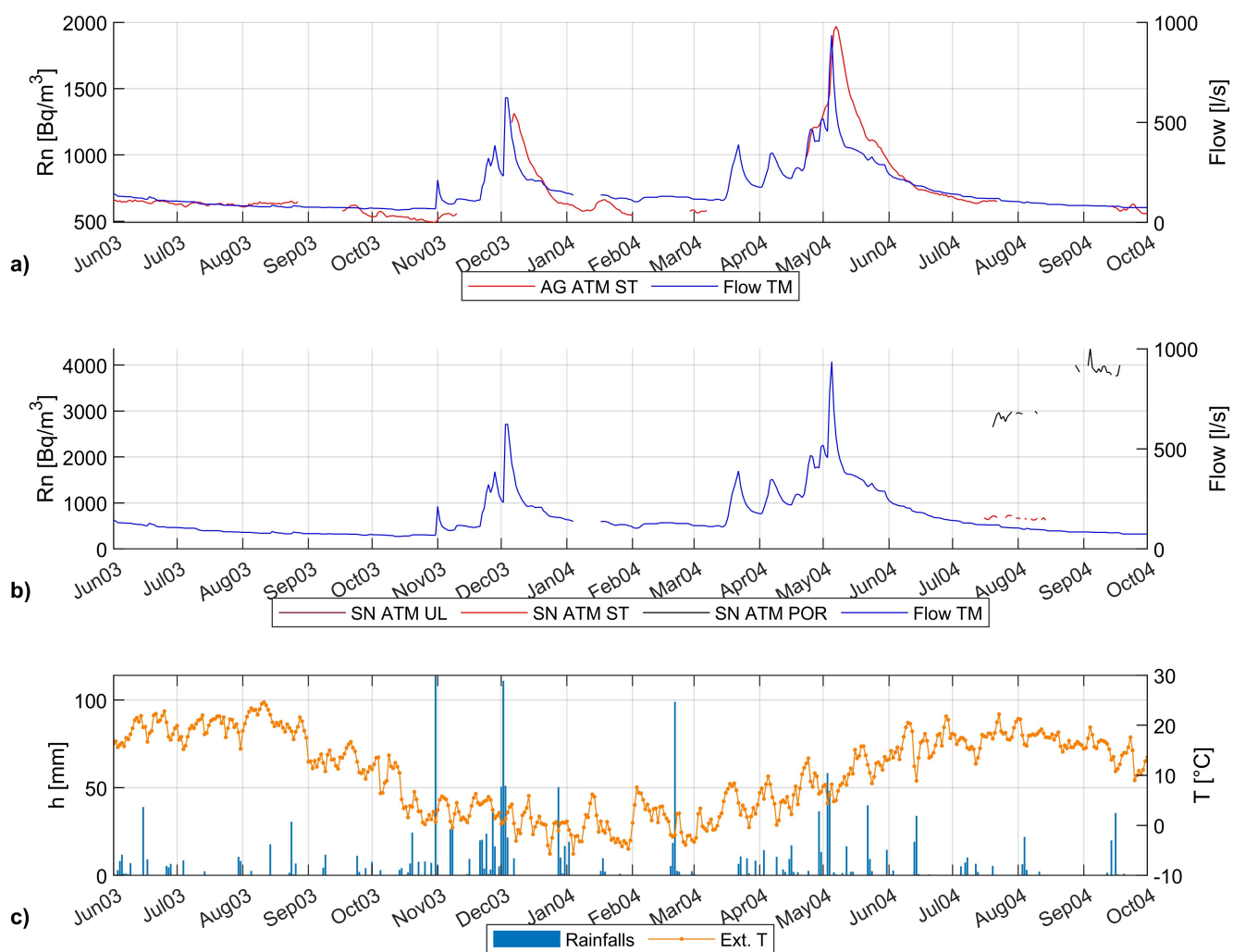
Supplementary Table A 1. Working period of instruments for continuous measurement of Rn in cave atmosphere.

	TORRENTE MORA (<i>Mora creek</i>)		POLLA ANATRE	
Instrument	Time window	Sampling frequency	Time window	Sampling frequency
<i>AlphaGuard AG1889</i>	Oct 2010 → Dec 2020	From 10' to 60'	-	-
<i>AlphaGuard AG0332</i>	Dec 2020 → <i>working</i>	10'	Jul 2011 → Dec 2014	10'
<i>AlphaGuard AG2141</i>	-	-	Jan 2015 → Oct 2017	10'

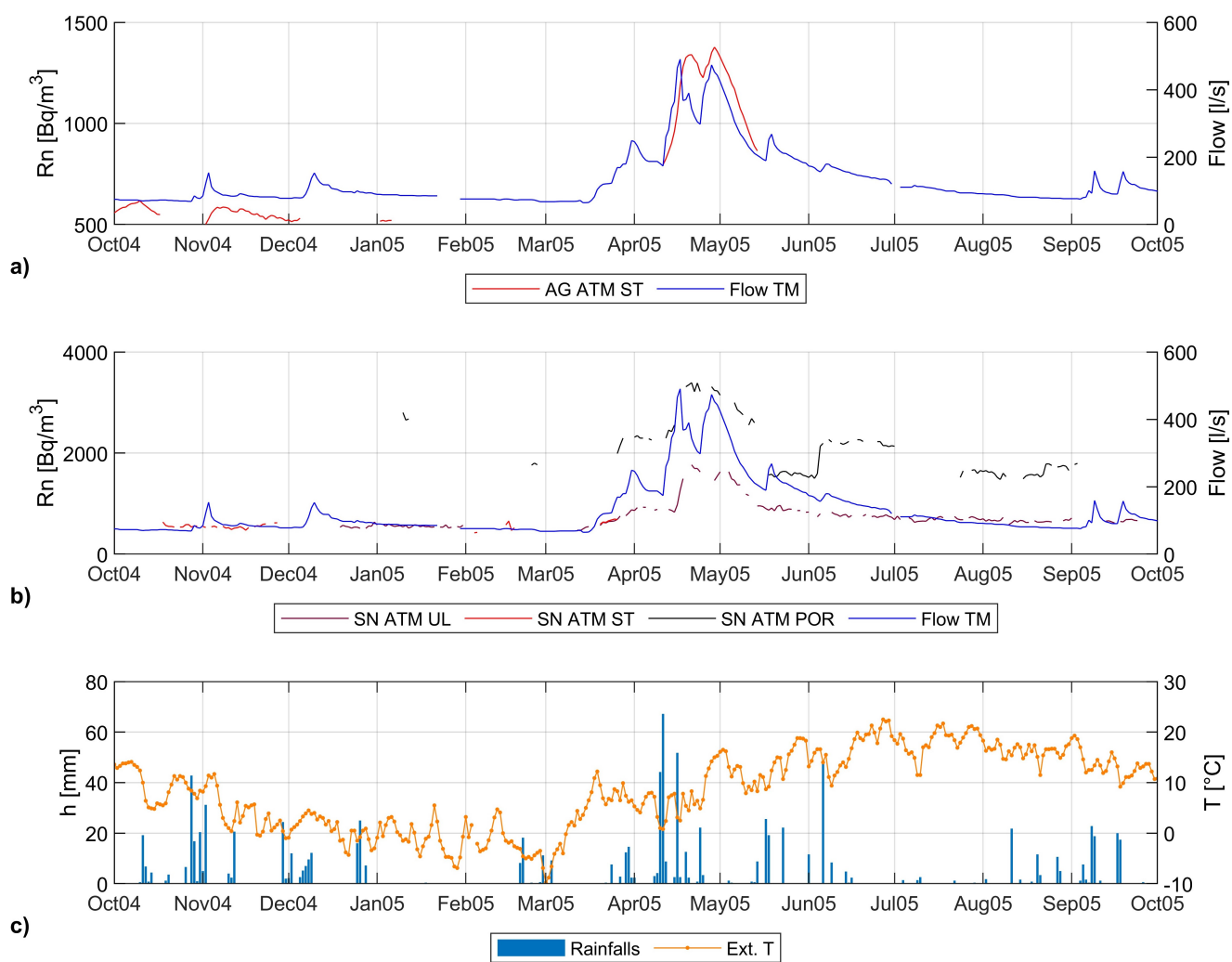
Supplementary Table A 2. Working period of instruments (located in the Main Laboratory) for continuous measurement of Rn in water



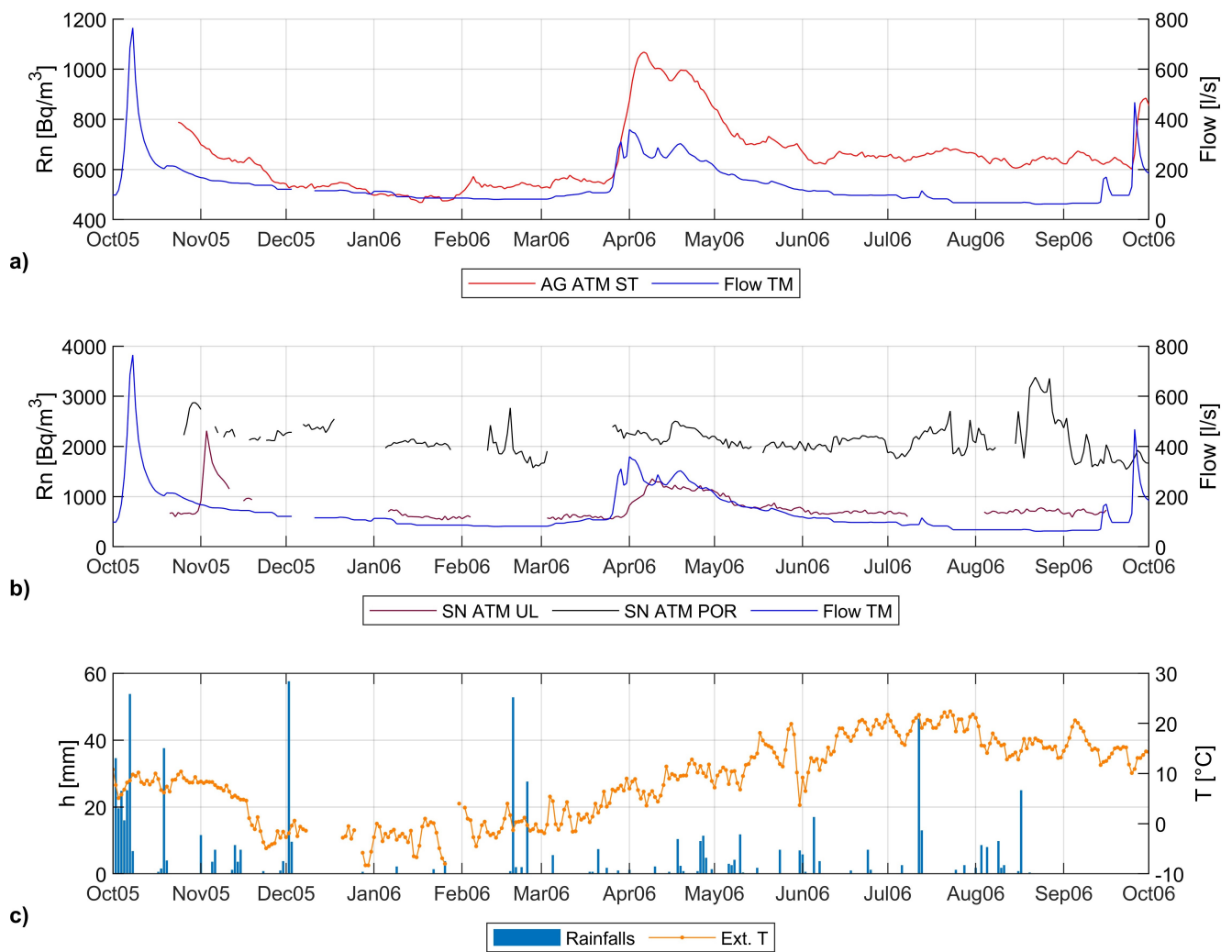
Supplementary Figure A 1. Positions of radon measuring instruments in air and water within *Bossea* cave, as well as those acquiring water flow rates and geochemical parameters data (electrical conductivity and water temperature). Modified from vertical long profile East-West traced by *Club Alpino Italiano, S.O. Bossea*.



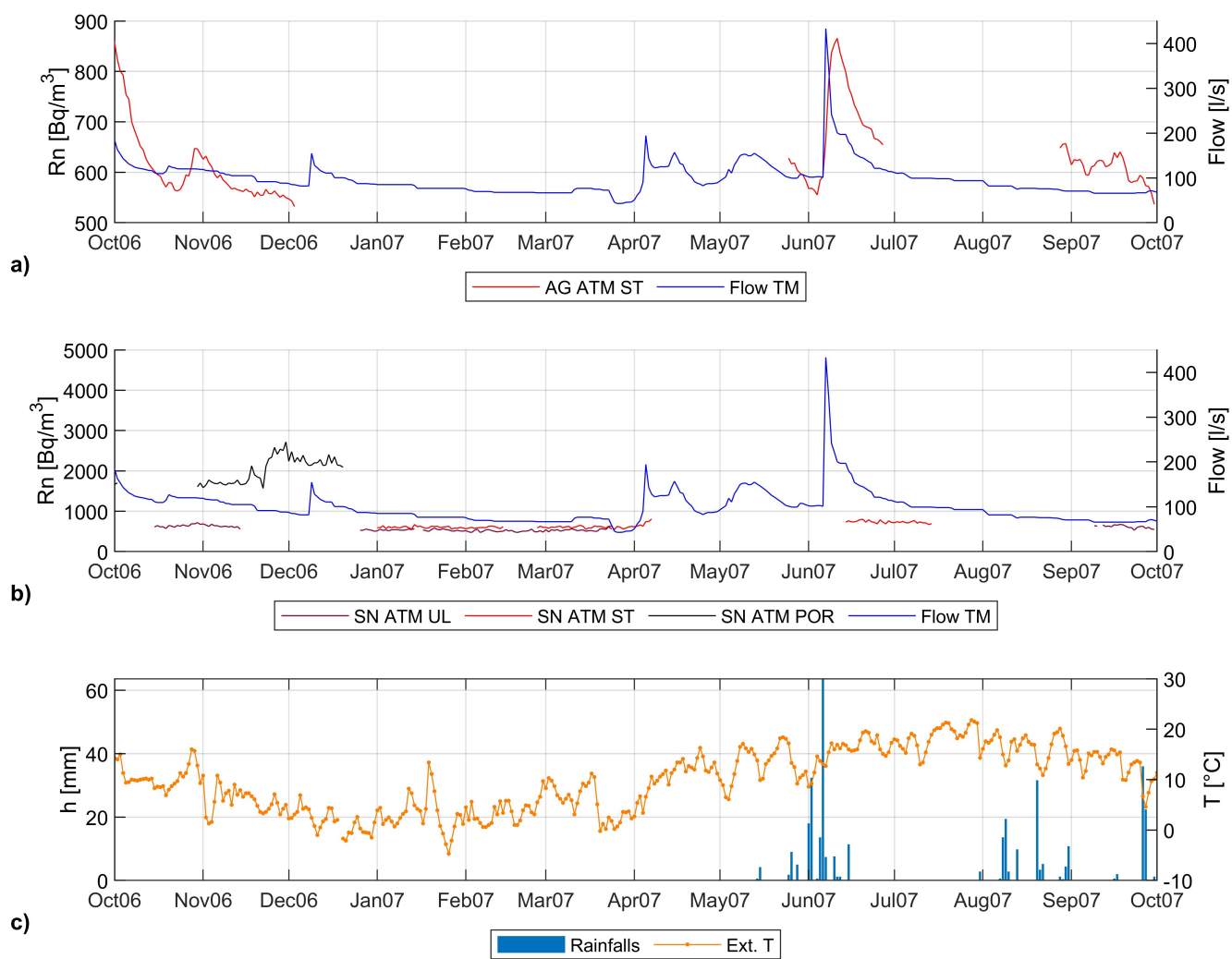
Supplementary Figure A 2. Hydrological year 2003-2004 **a)** Rn monitored by *AlphaGuard* instruments and *Mora* creek flows. **b)** Rn monitored by *Sun Nuclear* instruments and *Mora* creek flows. **c)** Daily rainfalls and external average temperatures.



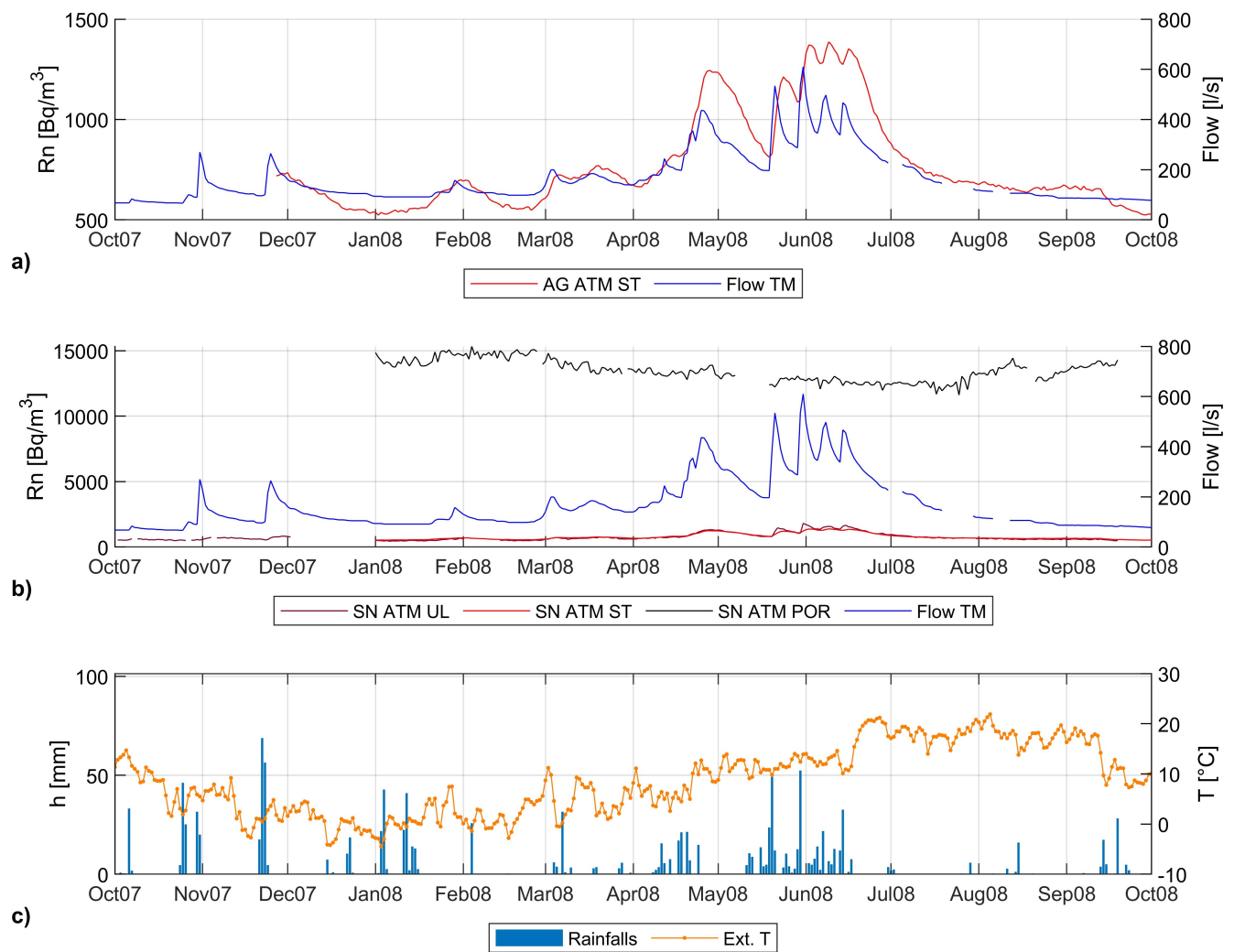
Supplementary Figure A 3. Hydrological year 2004-2005 **a)** Rn monitored by *AlphaGuard* instruments and *Mora* creek flows. **b)** Rn monitored by *Sun Nuclear* instruments and *Mora* creek flows. **c)** Daily rainfalls and external average temperatures.



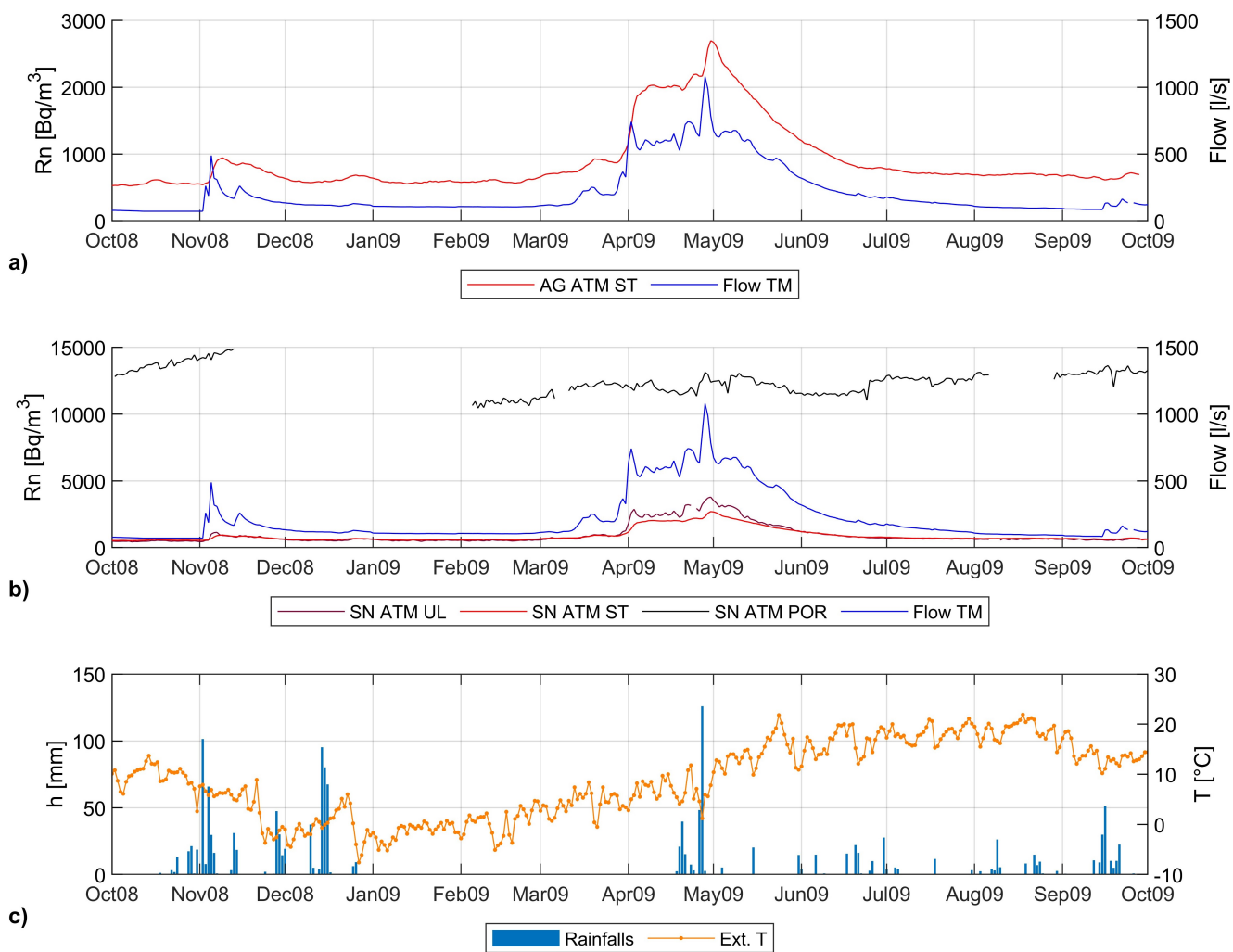
Supplementary Figure A 4. Hydrological year 2005-2006 **a)** Rn monitored by *AlphaGuard* instruments and *Mora* creek flows. **b)** Rn monitored by *Sun Nuclear* instruments and *Mora* creek flows. **c)** Daily rainfalls and external average temperatures.



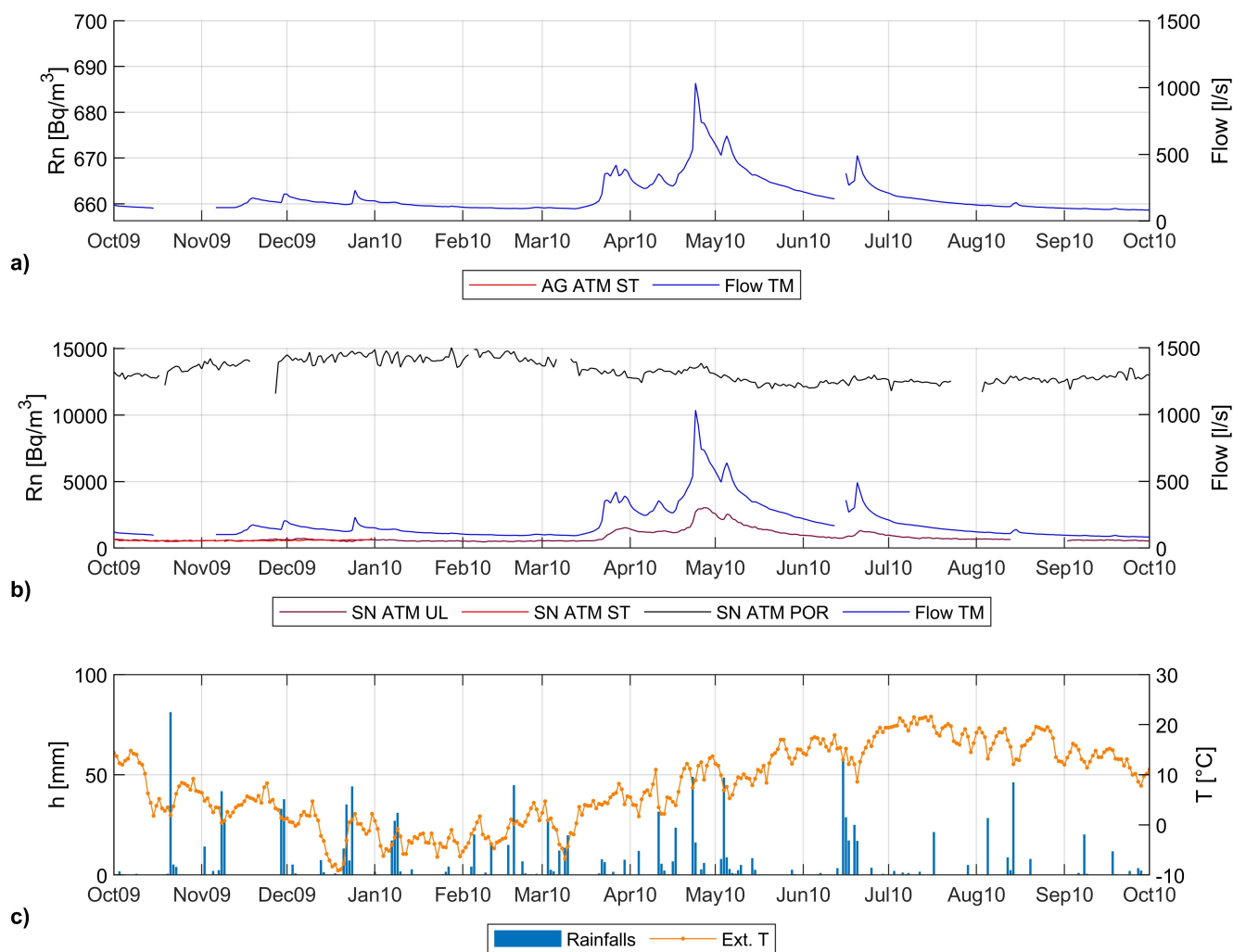
Supplementary Figure A 5. Hydrological year 2006-2007 **a)** Rn monitored by *AlphaGuard* instruments and *Mora* creek flows. **b)** Rn monitored by *Sun Nuclear* instruments and *Mora* creek flows. **c)** Daily rainfalls and external average temperatures.



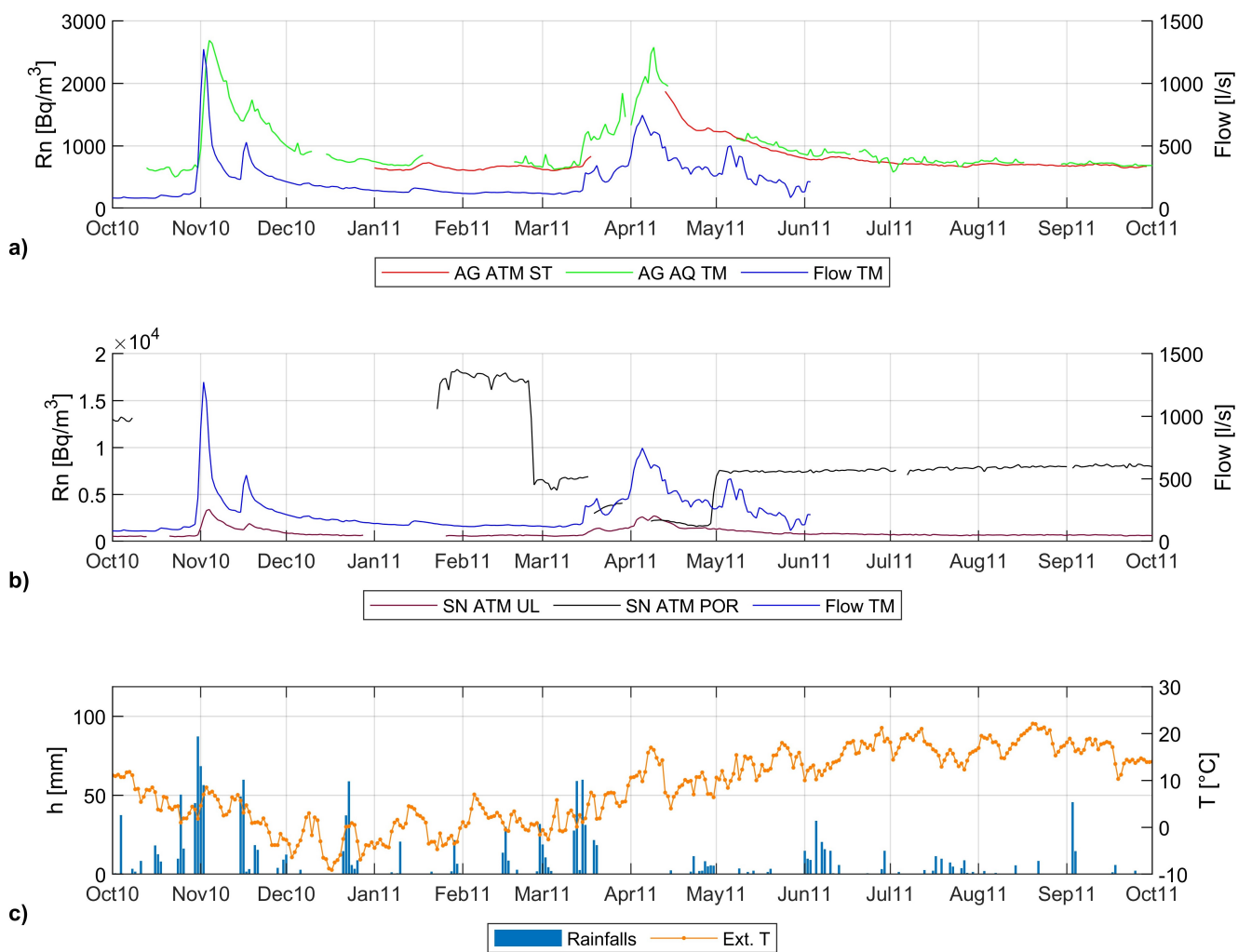
Supplementary Figure A 6. Hydrological year 2007-2008 **a)** Rn monitored by *AlphaGuard* instruments and *Mora* creek flows. **b)** Rn monitored by *Sun Nuclear* instruments and *Mora* creek flows. **c)** Daily rainfalls and external average temperatures.



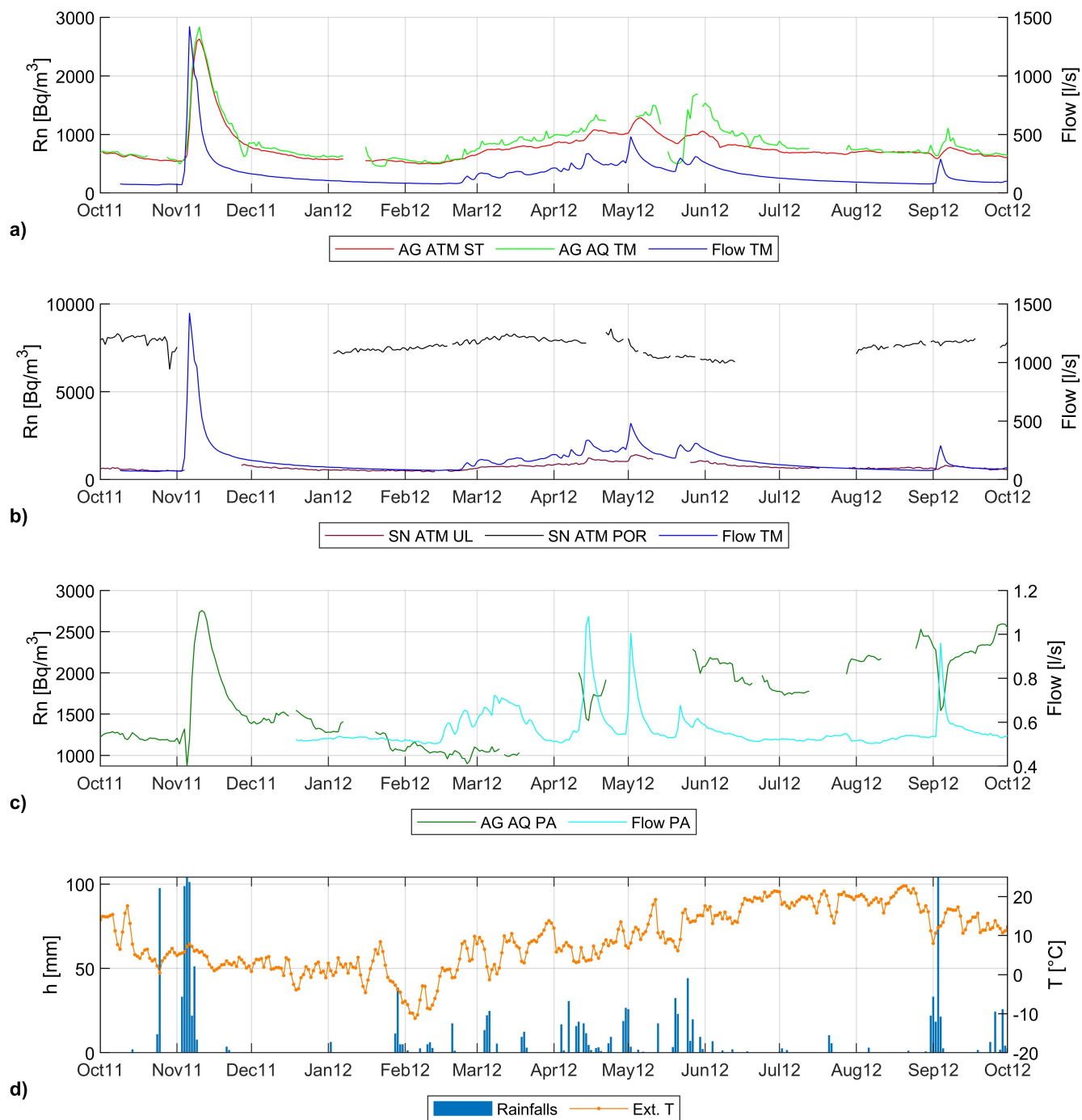
Supplementary Figure A 7. Hydrological year 2008-2009 **a)** Rn monitored by *AlphaGuard* instruments and *Mora* creek flows. **b)** Rn monitored by *Sun Nuclear* instruments and *Mora* creek flows. **c)** Daily rainfalls and external average temperatures.



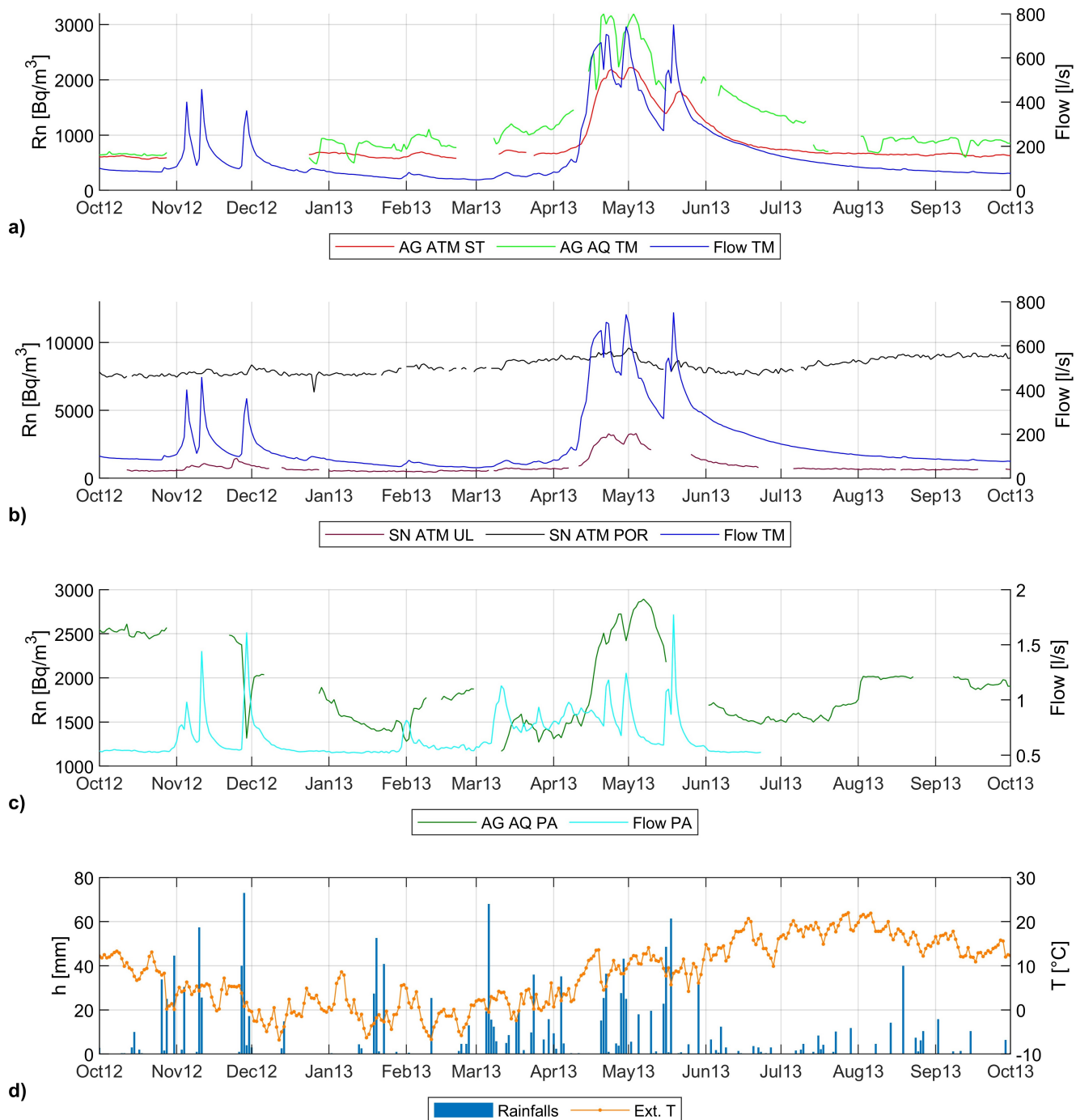
Supplementary Figure A 8. Hydrological year 2009-2010 **a)** Rn monitored by *AlphaGuard* instruments and *Mora* creek flows. **b)** Rn monitored by *Sun Nuclear* instruments and *Mora* creek flows. **c)** Daily rainfalls and external average temperatures.



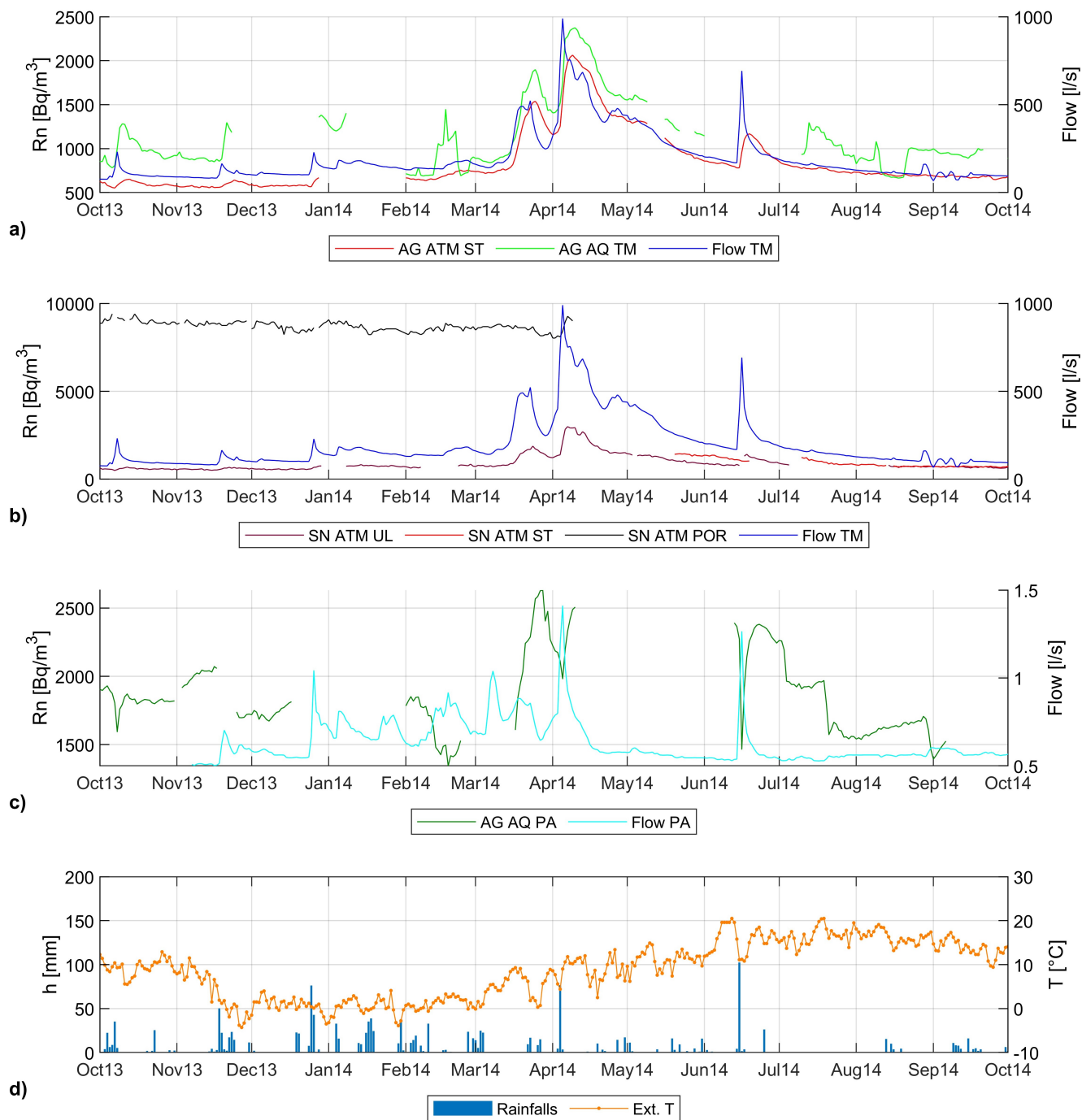
Supplementary Figure A 9. Hydrological year 2010-2011 **a)** Rn monitored by *AlphaGuard* instruments and *Mora* creek flows. **b)** Rn monitored by *Sun Nuclear* instruments and *Mora* creek flows. **c)** Daily rainfalls and external average temperatures.



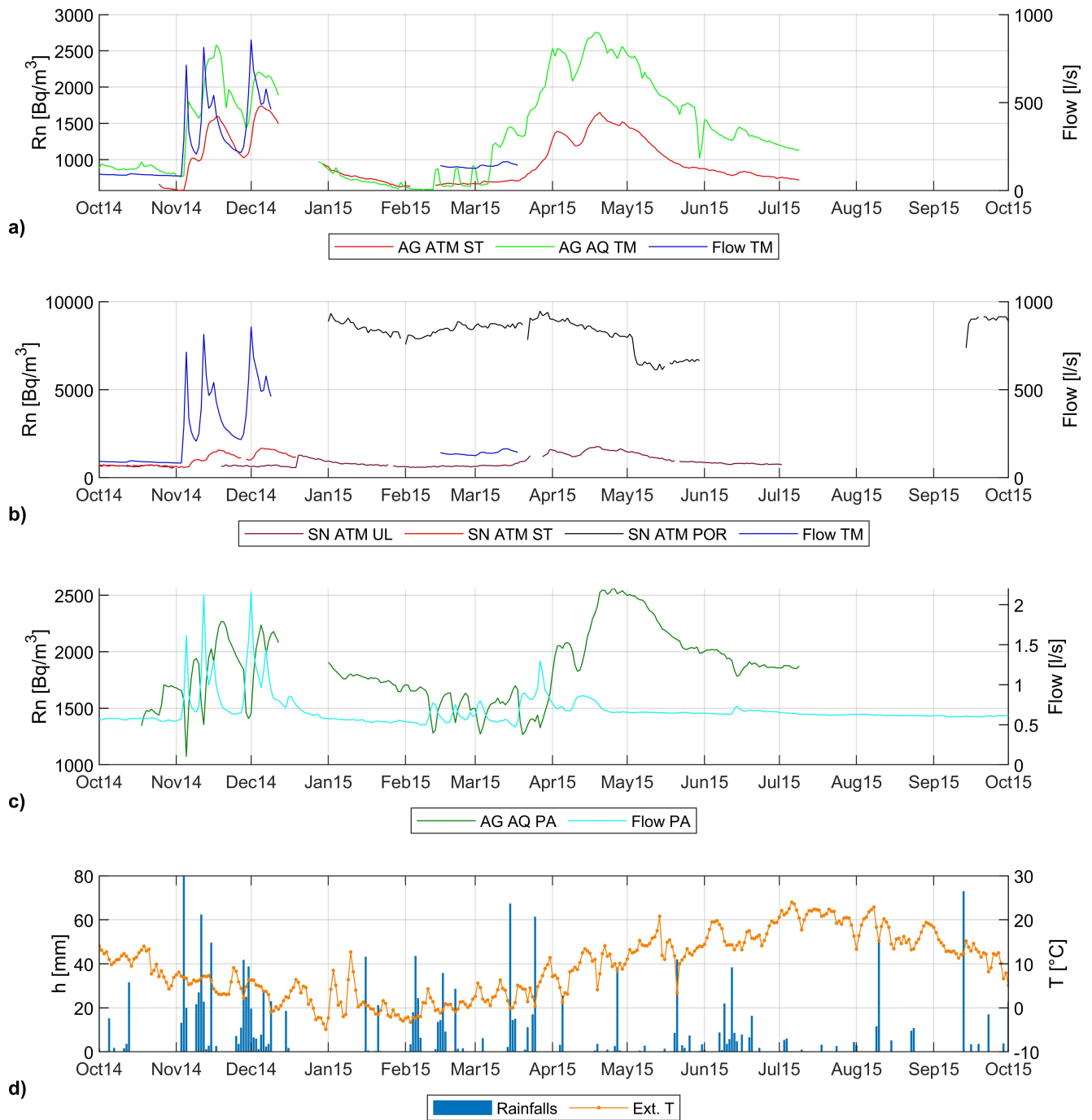
Supplementary Figure A 10. Hydrological year 2011-2012 **a)** Rn monitored by *AlphaGuard* instruments and *Mora* creek flows. **b)** Rn monitored by *Sun Nuclear* instruments and *Mora* creek flows. **c)** Rn monitored by *AlphaGuard* instruments and *Polla delle Anatre* flows. **d)** Daily rainfalls and external average temperatures.



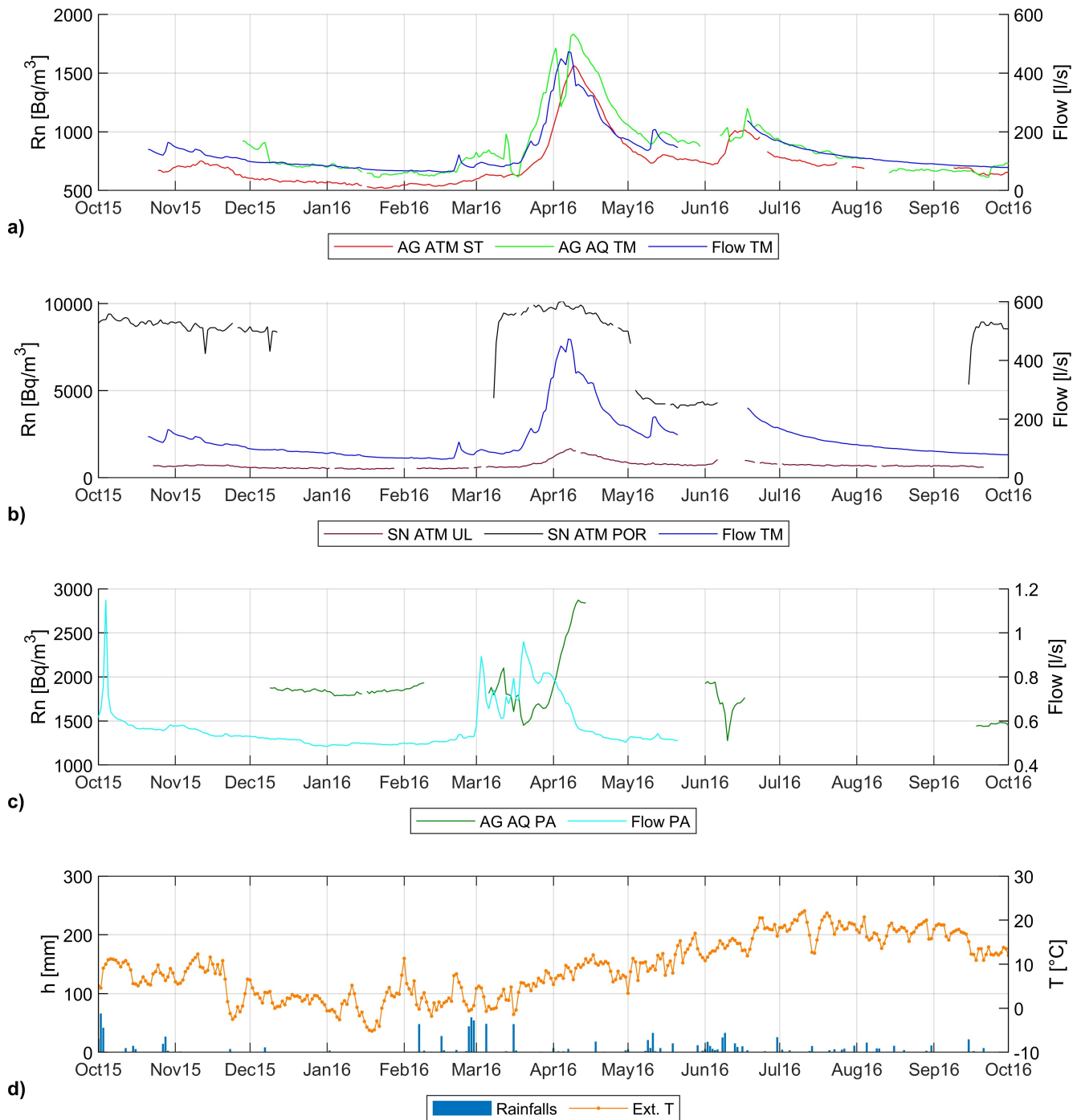
Supplementary Figure A 11. Hydrological year 2012-2013 **a)** Rn monitored by *AlphaGuard* instruments and *Mora* creek flows. **b)** Rn monitored by *Sun Nuclear* instruments and *Mora* creek flows. **c)** Rn monitored by *AlphaGuard* instruments and *Polla delle Anatre* flows. **d)** Daily rainfalls and external average temperatures.



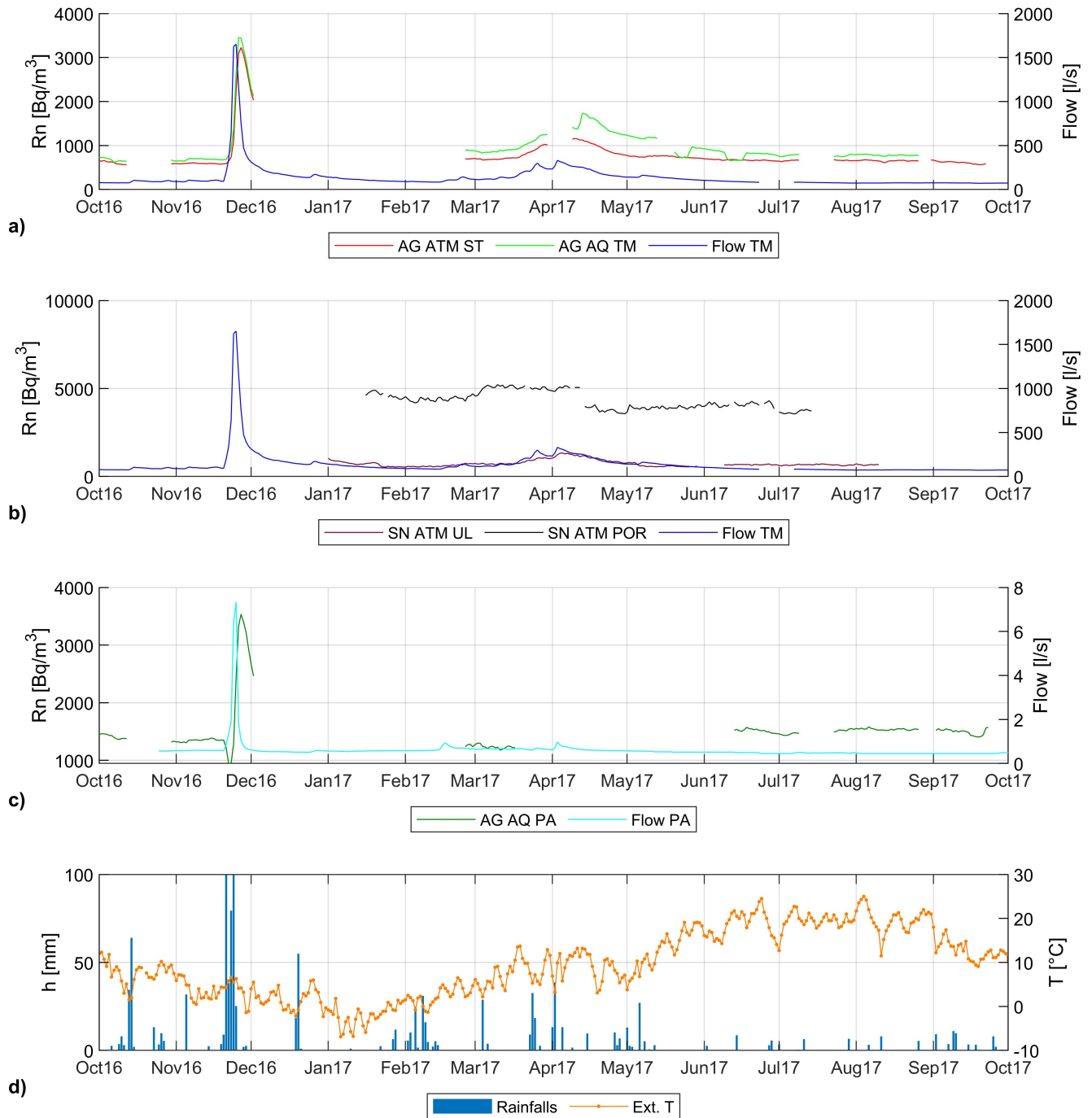
Supplementary Figure A 12. Hydrological year 2013-2014 **a)** Rn monitored by *AlphaGuard* instruments and *Mora* creek flows. **b)** Rn monitored by *Sun Nuclear* instruments and *Mora* creek flows. **c)** Rn monitored by *AlphaGuard* instruments and *Polla delle Anatre* flows. **d)** Daily rainfalls and external average temperatures.



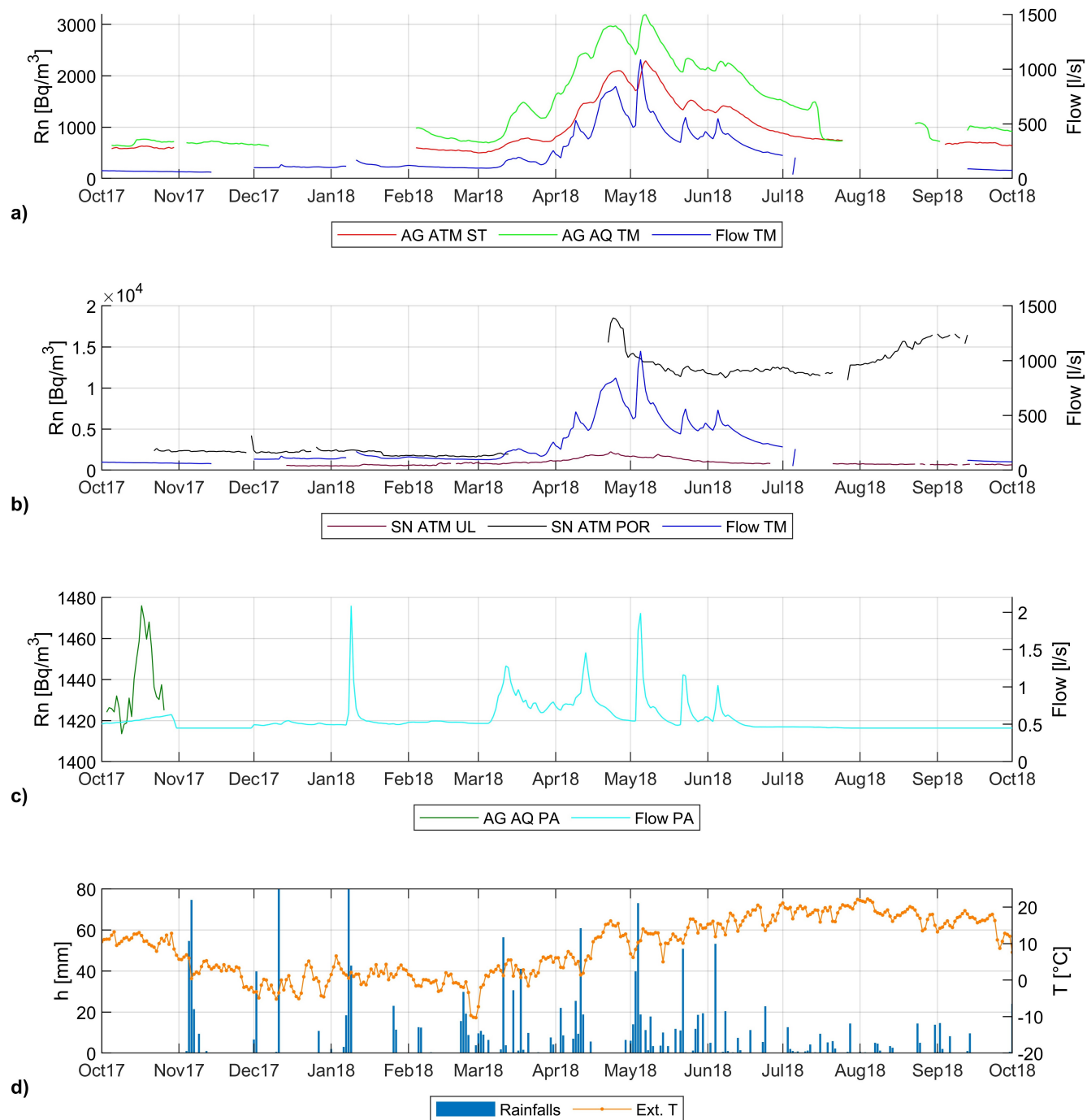
Supplementary Figure A 13. Hydrological year 2014-2015 **a)** Rn monitored by *AlphaGuard* instruments and *Mora* creek flows. **b)** Rn monitored by *Sun Nuclear* instruments and *Mora* creek flows. **c)** Rn monitored by *AlphaGuard* instruments and *Polla delle Anatre* flows. **d)** Daily rainfalls and external average temperatures.



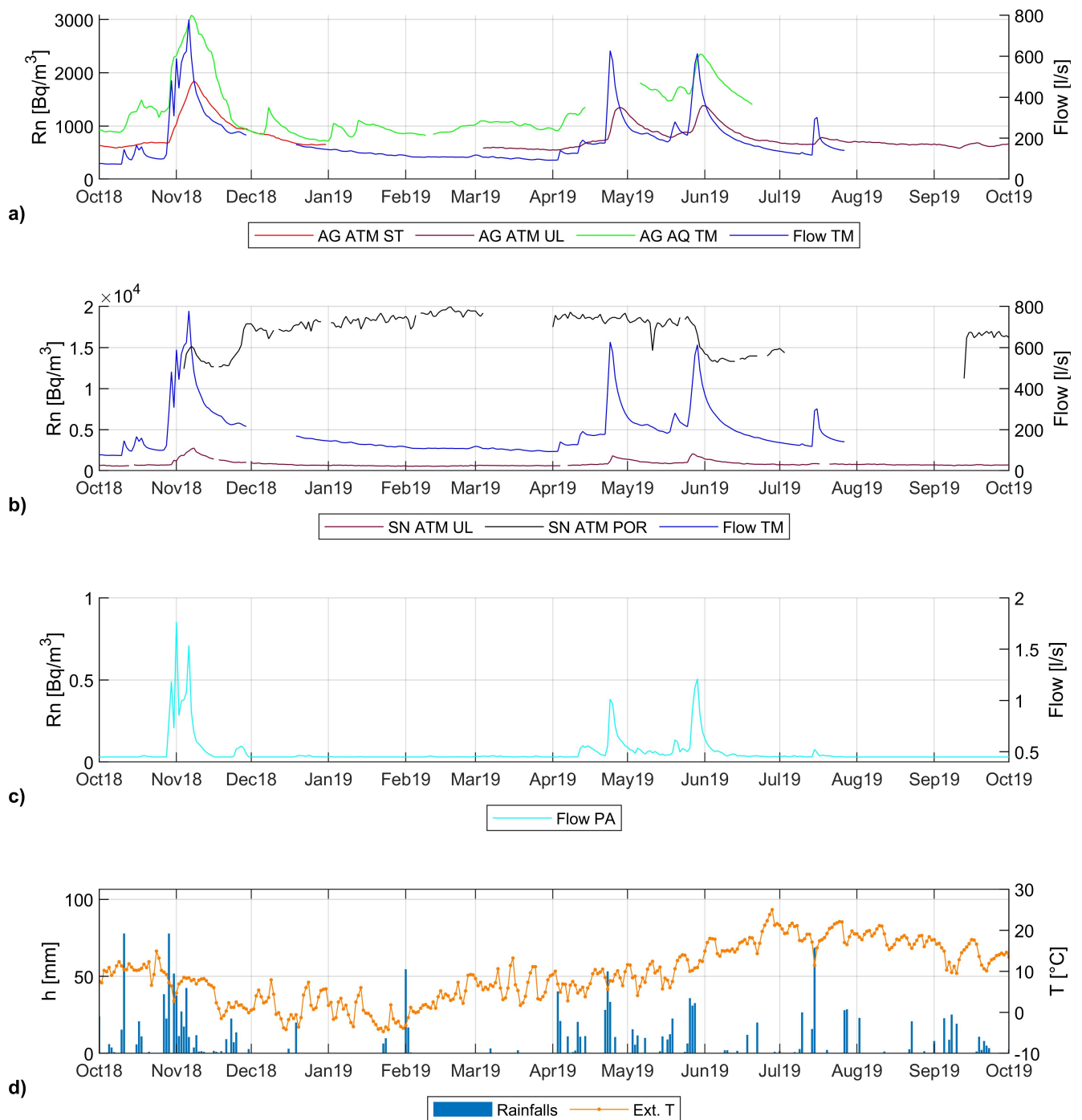
Supplementary Figure A 14. Hydrological year 2015-2016 **a)** Rn monitored by *AlphaGuard* instruments and *Mora* creek flows. **b)** Rn monitored by *Sun Nuclear* instruments and *Mora* creek flows. **c)** Rn monitored by *AlphaGuard* instruments and *Polla delle Anatre* flows. **d)** Daily rainfalls and external average temperatures.



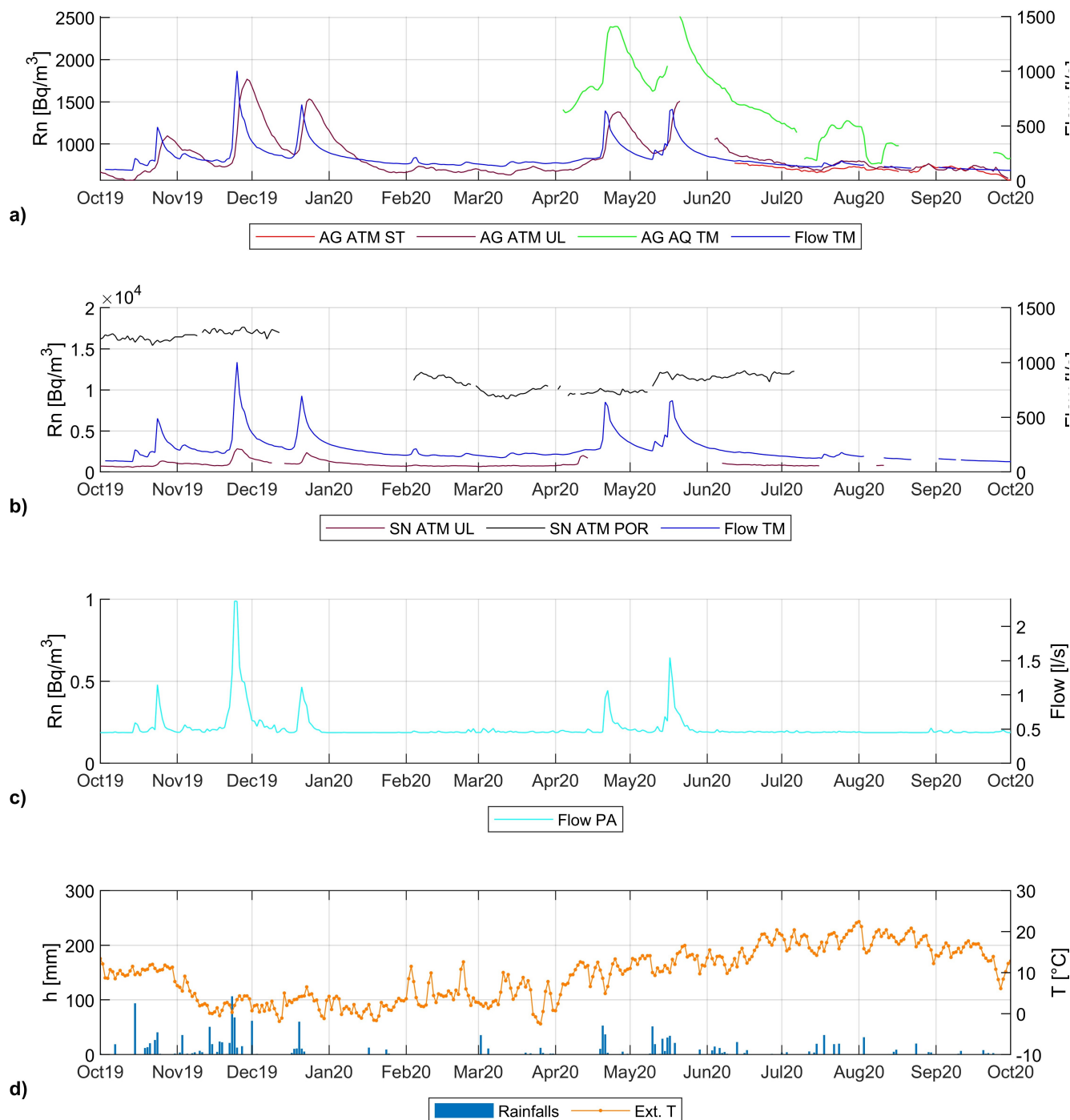
Supplementary Figure A 15. Hydrological year 2016-2017 **a)** Rn monitored by *AlphaGuard* instruments and *Mora* creek flows. **b)** Rn monitored by *Sun Nuclear* instruments and *Mora* creek flows. **c)** Rn monitored by *AlphaGuard* instruments and *Polla delle Anatre* flows. **d)** Daily rainfalls and external average temperatures.



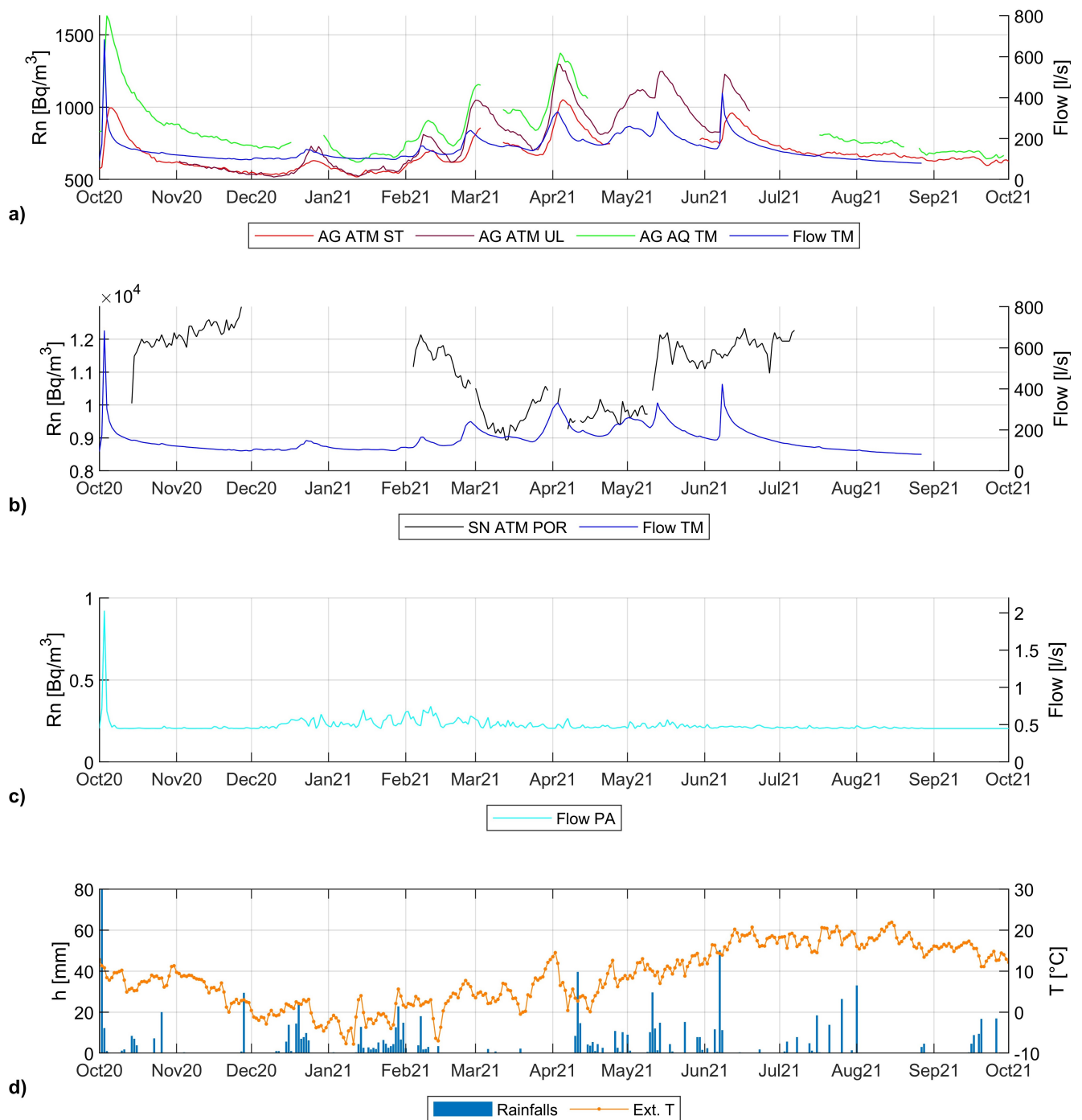
Supplementary Figure A 16. Hydrological year 2017-2018 **a)** Rn monitored by *AlphaGuard* instruments and *Mora* creek flows. **b)** Rn monitored by *Sun Nuclear* instruments and *Mora* creek flows. **c)** Rn monitored by *AlphaGuard* instruments and *Polla delle Anatre* flows. **d)** Daily rainfalls and external average temperatures.



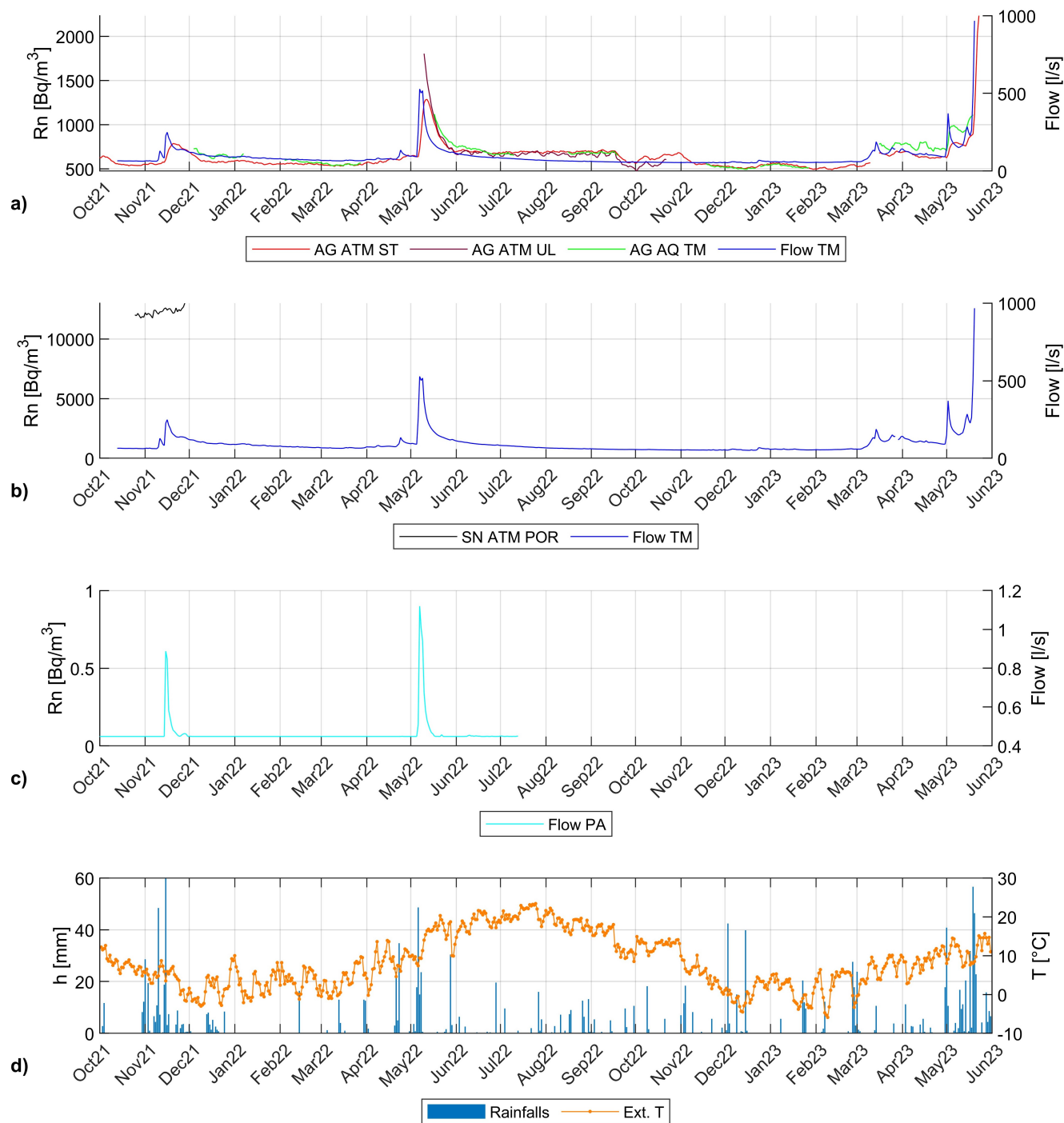
Supplementary Figure A 17. Hydrological year 2018-2019 **a)** Rn monitored by *AlphaGuard* instruments and *Mora* creek flows. **b)** Rn monitored by *Sun Nuclear* instruments and *Mora* creek flows. **c)** Rn monitored by *AlphaGuard* instruments and *Polla delle Anatre* flows. **d)** Daily rainfalls and external average temperatures.



Supplementary Figure A 18. Hydrological year 2019-2020 **a)** Rn monitored by *AlphaGuard* instruments and *Mora* creek flows. **b)** Rn monitored by *Sun Nuclear* instruments and *Mora* creek flows. **c)** Rn monitored by *AlphaGuard* instruments and *Polla delle Anatre* flows. **d)** Daily rainfalls and external average temperatures.



Supplementary Figure A 19. Hydrological year 2020-2021 **a)** Rn monitored by *AlphaGuard* instruments and *Mora* creek flows. **b)** Rn monitored by *Sun Nuclear* instruments and *Mora* creek flows. **c)** Rn monitored by *AlphaGuard* instruments and *Polla delle Anatre* flows. **d)** Daily rainfalls and external average temperatures.

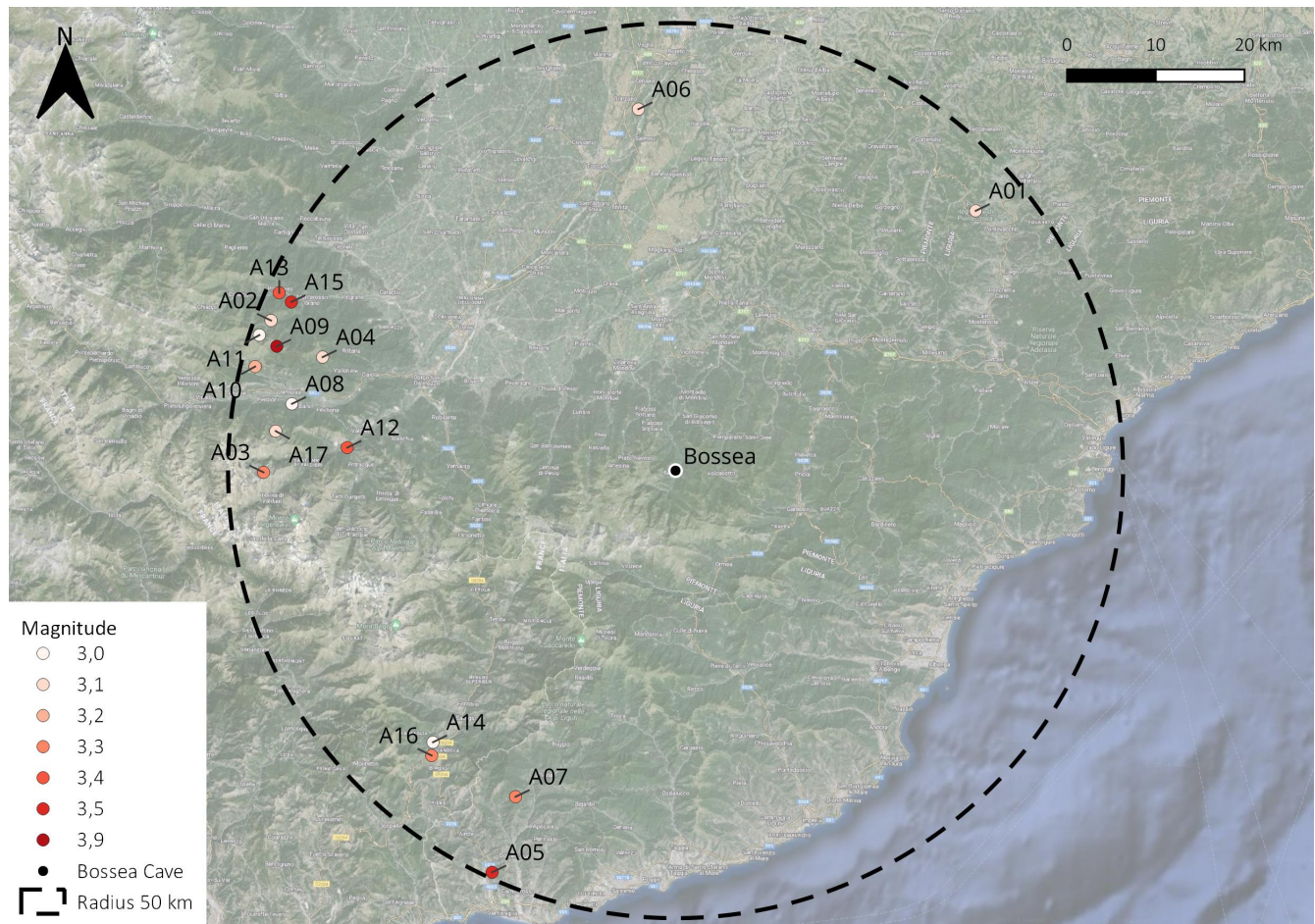


Supplementary Figure A 20. Hydrological year 2021-2022 and part of 2022-2023 **a)** Rn monitored by *AlphaGuard* instruments and *Mora* creek flows. **b)** Rn monitored by *Sun Nuclear* instruments and *Mora* creek flows. **c)** Rn monitored by *AlphaGuard* instruments and *Polla delle Anatre* flows. **d)** Daily rainfalls and external average temperatures.

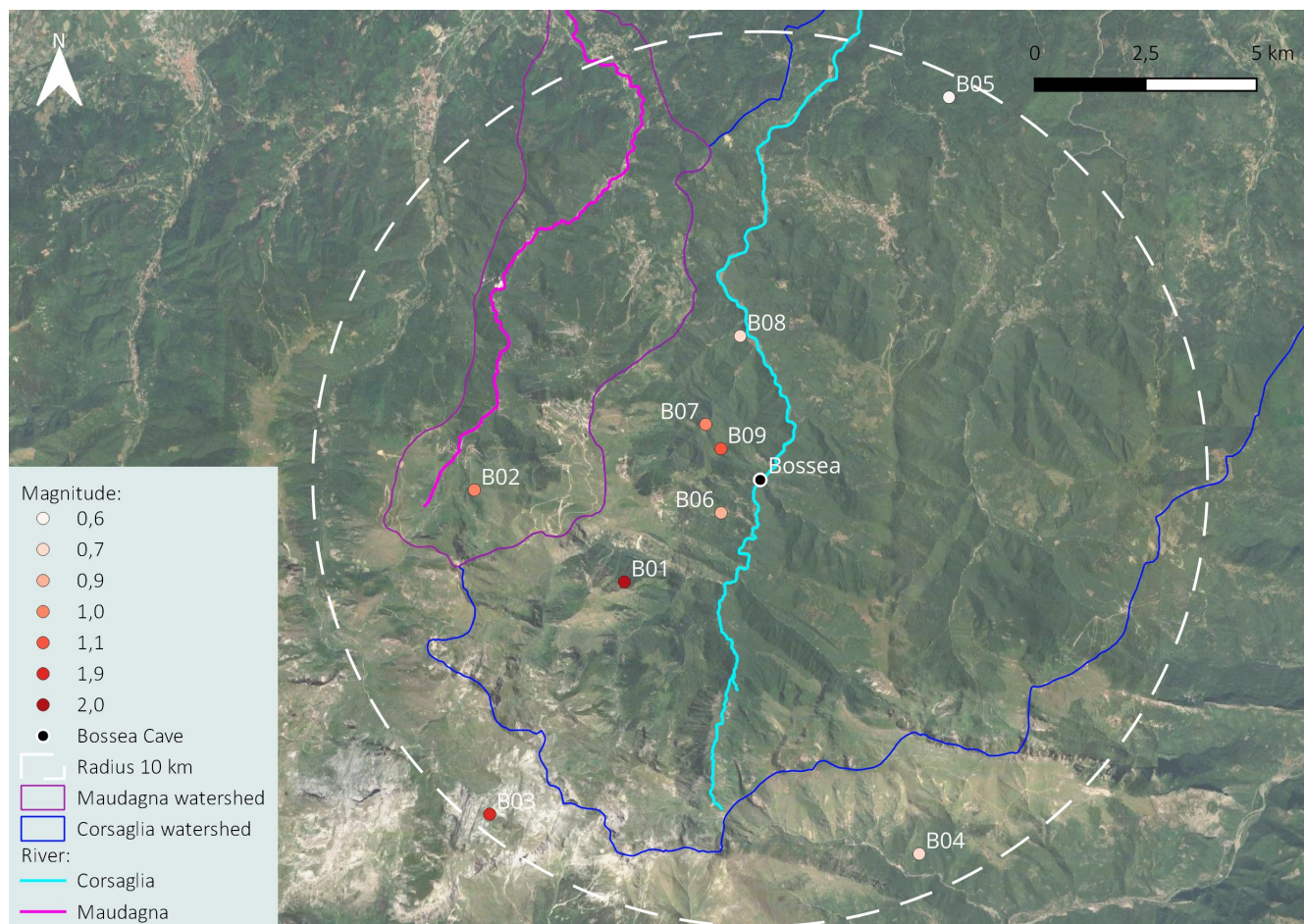
SUPPLEMENTARY MATERIAL B

This section includes the list of earthquakes which are considered for the application of the *residual method* (Supplementary Table B1). Earthquakes from 01 January 2003 with epicentral distance from *Bossea* cave < 50 km are indicated in Supplementary Figure B1, while those from 01 Jan 2003 with epicentral distance < 10 km are in Supplementary Figure B2.

Statistical parameters obtained through the application of the *residual method* are reported in Supplementary Table B1. Poor correlations with seismic events emerge: absence of significant peaks beyond the conventional anomaly threshold of $+2\sigma$ is reported in all cases, except for earthquake B03 (atmospheric residuals). In this case, it is necessary to consider that the limited extent of the time series does not allow for accurate estimation, as highlighted by the residual mean exceeding $1 \cdot 10^{-3}$ (Supplementary Figure B6d); meantime, no relevant peaks are appreciated in water (see also Supplementary Figure B6f).



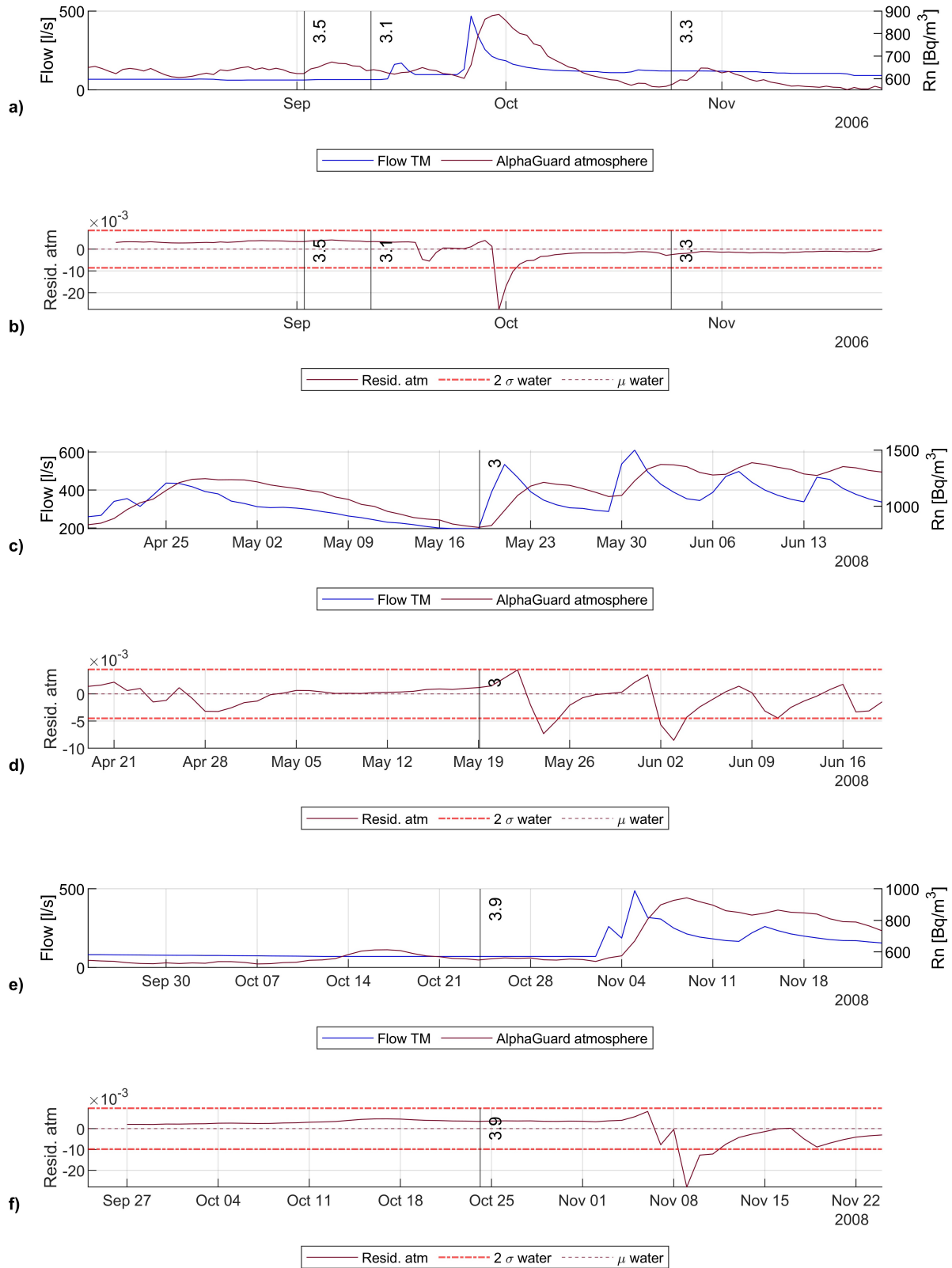
Supplementary Figure B 1. Earthquakes from 01 Jan 2003 with epicentral distance < 50 km from Bossea Cave and $M > 3.0$. The spatial distribution of epicenters in the study area shows strong heterogeneities: several clusters can be observed near the Argentera massif with events of magnitude greater than 4.0, as well as along the extent of the Ligurian Alpine arc, with intensification as it transitions towards the Maritime Alps domain. Indeed, the territory straddling the Italian-French border stretches between the Argentera and the topographic depression of the Ligurian oceanic basin, whose current configuration is linked to the convergence between the Eurasian and African plates began 100 million years ago¹. Concurrently, the opening of the Ligurian-Provençal basin (between 28 and 18 million years ago) resulted in a constant compressive action, as measured by the convergent displacements between the two plates of $6,2 \pm 0,5$ mm/yr in the direction of $163N \pm 9^\circ$ ². Seismic database: INGV web portal (<https://www.ingv.it/>).



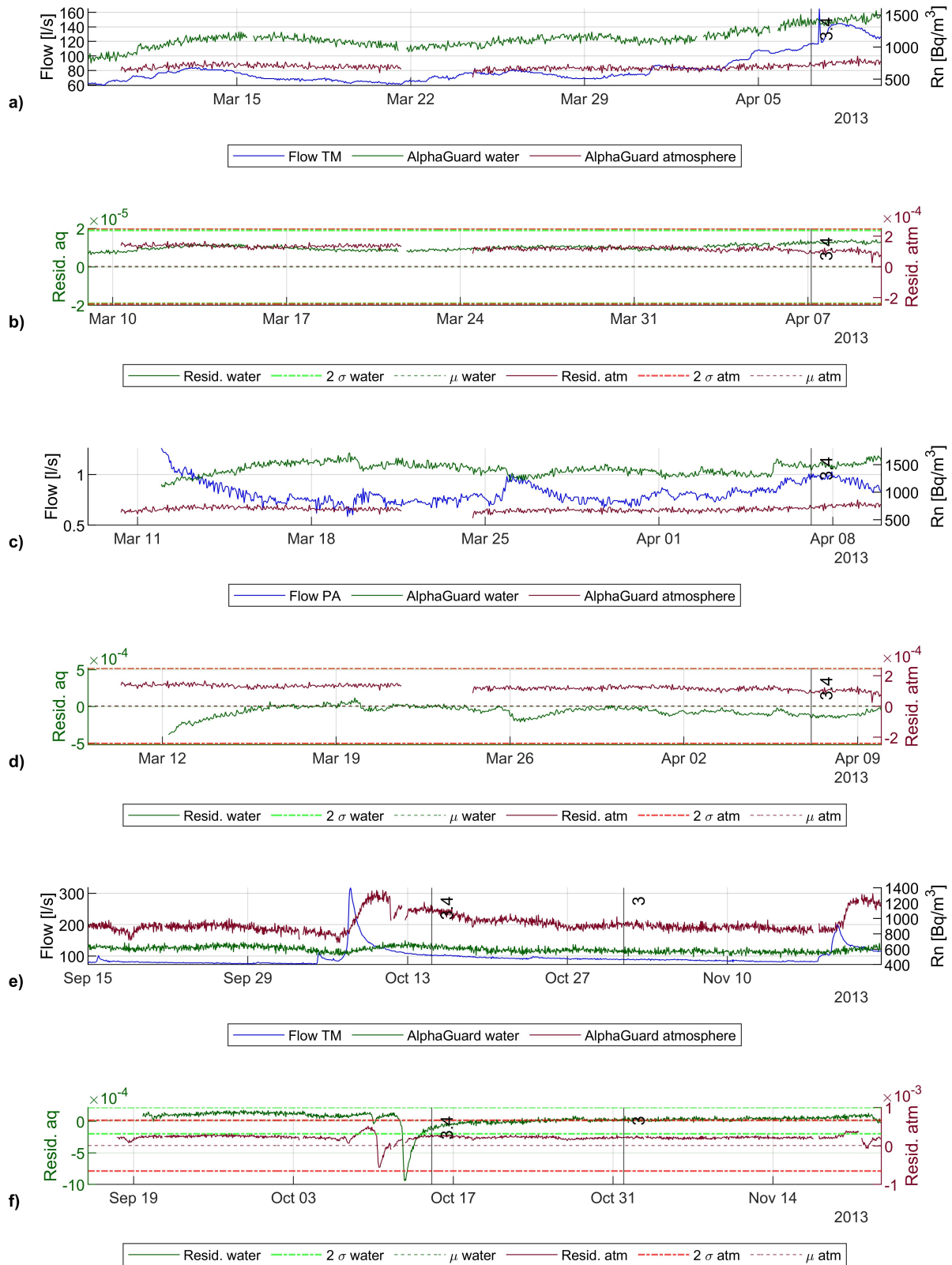
Supplementary Figure B 2. Earthquakes from 01 Jan 2003 with epicentral distance < 10 km from Bossea Cave. Seismic database: INGV web portal (<https://www.ingv.it/>).

				Water Rn		Atmosphere Rn	
ID	Date	M	Radius [km]	$\mu_{res} (\cdot 10^{-8})$	$\sigma_{res} (\cdot 10^{-4})$	$\mu_{res} (\cdot 10^{-6})$	$\sigma_{res} (\cdot 10^{-4})$
A05	02/09/2006 01:21	3.5	49.7	–	–	14.3	43.0
A06	11/09/2006 15:04	3.1	41.0	–	–	14.3	43.0
A07	24/10/2006 17:31	3.3	40.9	–	–	-0.8	64.0
A08	19/05/2008 01:53	3	43.7	–	–	7.6	23.0
A09	24/10/2008 03:06	3.9	46.9	–	–	13.9	49.0
A12	07/04/2013 03:13	3.4	36.9	9.2(*) 6.6(+)	2.6(*) 3.0(+)	3.25	1.2
A13	15/10/2013 02:47	3.4	48.8	-5.7 (+)	3.2 (+)	11.3	1.0
A14	31/10/2013 22:09	3	40.9	-5.7 (+)	3.2 (+)	11.3	1.0
A15	14/03/2016 13:36	3.5	47.1	197.1(*) 110.6(+)	3.7(*) 1.1(+)	0.7	1.7
A16	03/09/2016 19:14	3.3	42.1	-1.2(+)	0.7(+)	–	–
B01	08/04/2009 05:13	2.0	3.7	–	–	14.2	14.0
B03	22/10/2017 14:15	1.9	9.6	24.8(*) 49.7(+)	7.5(*) 5.2(+)	1000	0.9
B06	03/09/2021 03:46	0.9	1.1	2521.1(+)	0.4(°) 3.7(+)	-0.022	0.7
B07	04/09/2021 19:41	1	1.7	2521.1(+)	0.4(°) 3.7(+)	-0.022	0.7
B08	09/09/2021 09:17	0.7	3.3	2521.1(+)	0.4(°) 3.7(+)	-0.022	0.7
B09	24/09/2021 12:58	1.1	1.1	2521.1(+)	0.4(°) 3.7(+)	-0.022	0.7
(*): Water Rn AlphaGuard Polla delle Anatre; (+): Water Rn AlphaGuard Torrente Mora; (°) Water Rn MIAM Torrente Mora							

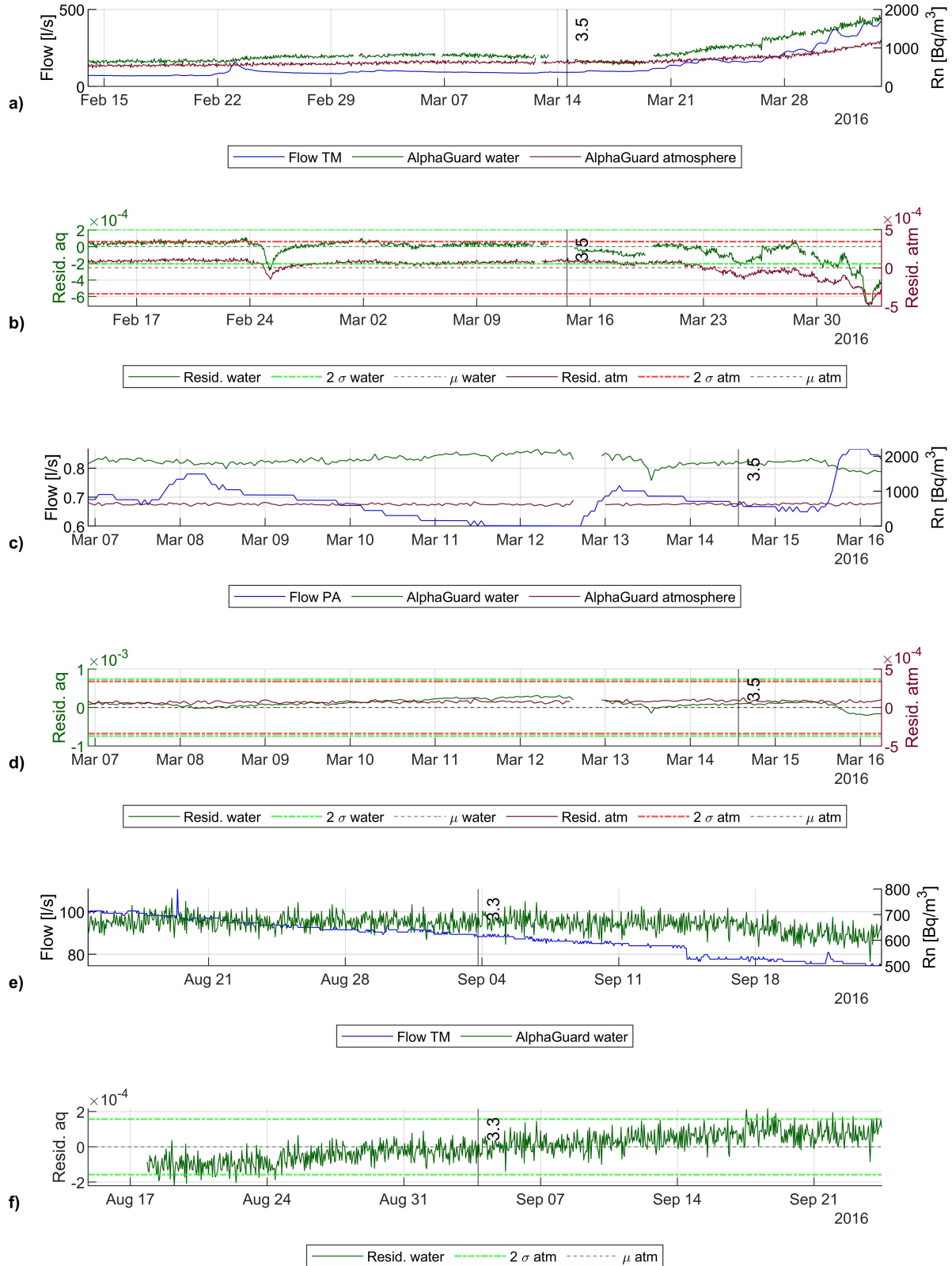
Supplementary Table B 1. Statistical parameters obtained from the application of the *residual method*. Earthquakes for which no Rn data were available are not listed.



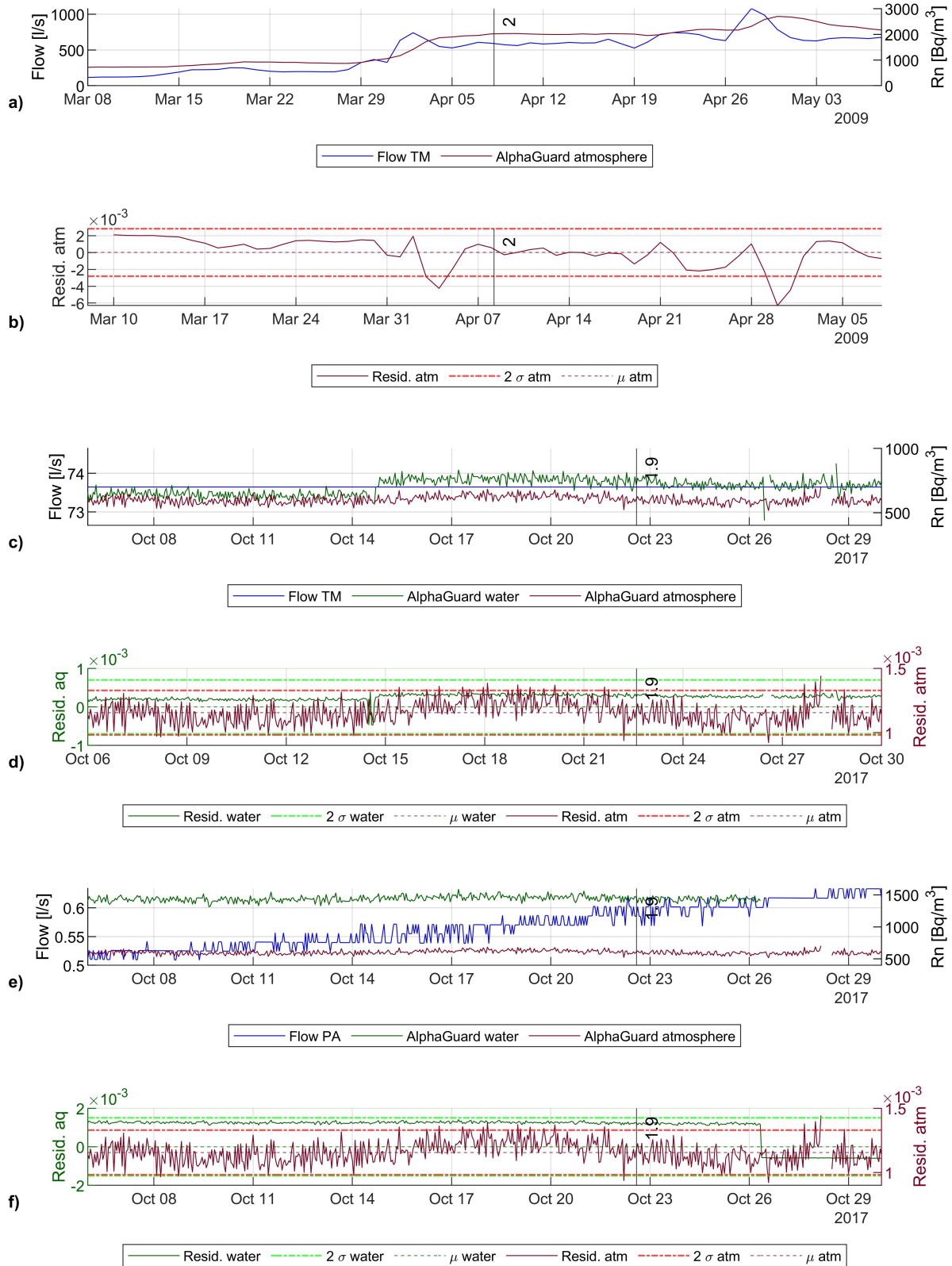
Supplementary Figure B 3. a) Earthquakes A05, A06 and A07. b) Residuals in atmosphere. c) Earthquake A08 and its residuals (d) in atmosphere. e) Earthquake A09 and its residuals in atmosphere (f). Abbreviations: TM - Mora creek.



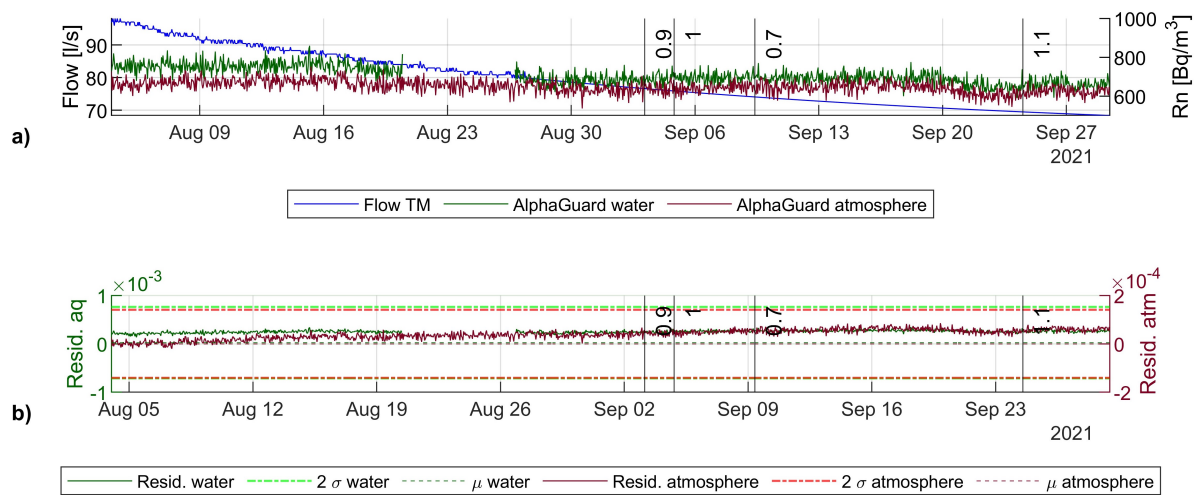
Supplementary Figure B 4. a) Earthquake A12 and its residuals (b) in water (TM) and atmosphere. c) Earthquake A12. (d) Residuals in water (PA) and atmosphere. e) Earthquakes A13 and A14. f) Residuals in water (TM) and atmosphere. Abbreviations: TM - *Mora creek*; PA - *Polla delle Anatre*.



Supplementary Figure B 5. **a)** Earthquake **A15**. **(b)** Residuals in water (TM) and atmosphere. **Earthquake A15**. **(d)** Residuals in water (PA) and atmosphere. **e)** Earthquake **A16** and its residuals **(f)** in water (TM). Abbreviations: TM - *Mora* creek; PA - *Polla delle Anatre*.



Supplementary Figure B 6. a) Earthquake **B01** and its residuals (b) in atmosphere. c) Earthquake **B03**. (d)) Residuals in water (TM) and atmosphere. e) Earthquake **B03**. f) Residuals in water (PA) and atmosphere. Abbreviations: TM - *Mora creek*; PA - *Polla delle Anatre*.



Supplementary Figure B 7. a) Earthquakes **B06**, **B07**, **B08** and **B09**. b) Residuals in water (TM) and atmosphere. Abbreviations: TM - *Mora* creek.

References

1. Larroque, C. *et al.* Active and recent deformation at the southern alps-ligurian basin junction. *Geol. en Mijnbouw* **80**, 255–272 (2001).
2. DeMets, C., Gordon, R. G., Argus, D. F. & Stein, S. Effect of recent revisions to the geomagnetic reversal time scale on estimates of current plate motions. *Geophys. research letters* **21**, 2191–2194 (1994).