

## Supplementary Materials for

### **Agricultural lime disturbs natural strontium isotope variations: Implications for provenance and migration studies**

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Fig. S4. Comparison of strontium concentrations measured by quadrupole ICP-MS and multicollector ICP-MS.

Reference (44)

## Supplementary materials

**Table S1. Strontium isotopic composition and concentration of surface water and reference samples.** For each water sample, we show sample locality, sample number, sampling date, geographic coordinates (decimal Grade System),  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio, strontium concentration, 1/Sr conc. and distance from pristine water body to farmland. External reproducibility is determined from repeated analyses of Holocene Okinawa foraminiferal standards.

Locality	Sample no.	Date of collection	Latitude	Longitude	$^{87}\text{Sr}/^{86}\text{Sr}$	Sr (mg/L)	1/Sr	Distance to farming (m)	Code*
<b>1. Kompedal Plantage-Vallerbæk Stream-Karup River, (Fig. 3, S1A)</b>									
Vallerbæk Stream	K-5	18-10-2017	56.241283	9.283874	0.71350	0.006	154.42	1200	WP
-	K-3	31-08-2017	56.261781	9.252835	0.71317	0.007	134.82	420	WP
-	K-1	31-08-2017	56.264756	9.244798	0.71307	0.010	95.36	490	WP
-	K-2	31-08-2017	56.267736	9.237932	0.71268	0.010	100.80	370	WP
-	K-14	08-06-2018	56.269419	9.231289	0.71305	0.025	39.80	18§	WP
-	K-13	08-06-2018	56.269858	9.229301	0.71245	0.036	28.00	5§	WP
-	K-6	22-11-2017	56.270895	9.224567	0.71099	0.048	20.95		WF
-	K-6a	08-06-2018	56.270895	9.224567	0.71153	0.038	26.60		WF
-	K-4	31-08-2017	56.275278	9.207502	0.70993	0.069	14.40		WP
Karup River	KAS	09-11-2017	56.290014	9.182303	0.70968	0.084	11.97		WF
-	KAM	09-11-2017	56.303477	9.159860	0.70970	0.090	11.15		WF
-	KAN	09-11-2017	56.313539	9.142920	0.70973	0.103	9.70		WF
Karup R at Hagebro	KAHA	09-11-2017	56.406313	9.008283	0.70954	0.110	9.06		WF
Karup R at Hagebro	KAHA-2	13-02-2018	56.377812	9.074645	0.70961	0.109	9.17		WF
Karup R at Resen	KARE	13-02-2018	56.377812	9.074645	0.70970	0.111	9.02		WF
Karup R at Trevad	KATR	13-02-2018	56.461869	8.984471	0.70946	0.115	8.72		WF
Karup R at Bærs	KABA	13-02-2018	56.514108	8.982449	0.70944	0.119	8.44		WF
Knudstrup (Pond)	K-7	29-10-2017	56.269251	9.312944	0.70804	0.045	22.35		CF
Groundwater†	KG-2	07-06-2018	56.231453	9.280806	0.71391	0.048	20.90	1180	WP
Groundwater‡	KG-3	07-06-2018	56.219763	9.248550	0.71308	0.054	18.60	190	WP
<b>2. Gludsted Plantage (Fig. S1C)</b>									
-	G-2b	02-09-2017	56.087317	9.363807	0.71183	0.003	342.13	680	WP
-	G-2a	02-09-2017	56.087075	9.363065	0.71183	0.002	527.37	620	WP
-	G-4	01-09-2017	56.086048	9.354797	0.71277	0.006	173.23	240	WP
-	G-1	01-09-2017	56.085255	9.349334	0.71147	0.009	114.29	210	WP
-	G-5	19-09-2017	56.089903	9.324909	0.71039	0.006	159.89	810	WP
Storå river	G-3	02-09-2017	56.101352	9.274616	0.70905	0.110	9.08		WF
<b>3. Nørlund Plantage-Harrild Heath (Fig. S1D)</b>									
Hallund Brook	NP-1	31-08-2017	56.053104	9.197114	0.71097	0.011	93.04	1550	WP
-	NP-2	31-08-2017	56.052532	9.194030	0.71195	0.006	176.77	1410	WP
-	NP-3	01-08-2017	56.050135	9.189121	0.71142	0.037	27.13	440	WP
-	NP-4	31-08-2017	56.056118	9.139240	0.71011	0.095	10.53		WF
-	NP-5	03-10-2017	56.059590	9.111094	0.70980	0.065	15.34		WF
Harrild Heath	HAR1	19-09-2017	56.014125	9.183813	0.71338	0.005	197.40	880	WP
<b>4. Frederikshåb Plantage (Fig. S1B)</b>									
N. Voldborg Lake	FPNS-1	03-10-2017	55.687061	9.161826	0.71161	0.023	43.47	550	WP
-	FPNS-2	03-10-2017	55.687061	9.161826	0.71229	0.025	40.20	550	WP
-	FPNS-3	03-10-2017	55.687061	9.161826	0.71219	0.025	40.11	550	WP
S. Voldborg Lake	FPVS-1	15-08-2017	55.685132	9.163733	0.71227	0.015	64.93	820	WP
-	FPVS-2	07-12-2017	55.685132	9.163733	0.71218	0.017	57.31	820	WP
Seven Year Lakes	FPSS-1	15-08-2017	55.678481	9.176481	0.71362	0.017	59.10	500	WP
-	FPSS-2	07-12-2017	55.678481	9.176481	0.71103	0.038	26.28	500	WP
Hestdalen	FPH-1	03-10-2017	55.674149	9.224173	0.71431	0.009	113.37	710	WP
-	FPH-2	07-12-2017	55.673964	9.232968	0.71495	0.020	51.25	575	WP
-	FHP-3	12-12-2017	55.674052	9.233284	0.71372	0.022	44.76	525	WP
<b>5. Randbøl Heath (Fig. S1B)</b>									
-	RH-1	15-08-2017	55.664033	9.150261	0.71138	0.005	190.48	160	WP
-	RH-2	01-09-2017	55.670165	9.167517	0.71079	0.002	426.14	620	WP
-	RH-2a	07-12-2017	55.670165	9.167517	0.71078	0.003	300.31	620	WP
-	RH-3	01-09-2017	55.670549	9.167251	0.71246	0.004	257.07	630	WP
-	RH-4	03-10-2017	55.658232	9.164567	0.71240	0.002	463.37	540	WP
-	RH-5	06-10-2017	55.663273	9.151096	0.71146	0.005	214.99	250	WP

-	RH-6	11-02-2018	55.655292	9.134861	0.71278	0.009	108.02	530	WP
Sluggård Plantage	SLA-1	11-02-2018	55.641518	9.118307	0.71210	0.004	229.93	480	WP
<b>6. Hærvejen (Fig. S1B)</b>									
-	HÆR-1	06-10-2017	55.645679	9.21044	0.71110	0.004	236.38	180	CP
Raised Bog	HÆR-2	09-01-2018	55.645222	9.213184	0.70952	0.016	61.17	320	CP
	OEL-1	09-01-2018	55.634112	9.235347	0.71156	0.019	51.45	260	CP
Natural spring	OEL-3	09-01-2018	55.632508	9.233643	0.71191	0.064	15.74	170	CP
	FIT-1	07-12-2017	55.620457	9.160824	0.70929	0.087	11.50		WF
<b>7. Gyttegård Plantage (Fig. S1E)</b>									
-	GYT-2	07-12-2017	55.728742	9.028517	0.71484	0.016	62.97	350	WP
<b>8. Bevtøft Plantage (Fig. S1J)</b>									
-	BEV-1	0-3-10-2017	55.205788	9.242873	0.71147	0.029	35.05	400	WP
-	BEV-2	03-10-2017	55.203302	9.245468	0.71414	0.010	101.70	180	WP
-	BEV-2a	12-12-2017	55.203302	9.245468	0.71414	0.010	97.79	180	WP
-	BEV-3	03-10-2017	55.210800	9.261471	0.70977	0.133	7.54		WF
<b>9. Ruggård Plantage (Fig. S1G)</b>									
-	RUG-1	12-12-2017	55.114907	9.317624	0.71115	0.006	179.55	350	WP
<b>10. Sepstrup Sande-Salten River (Fig. S1F)</b>									
-	SEPS-2	29-10-2017	56.087291	9.400274	0.71356	0.005	186.81	590	CP
-	SEPS 1	19-09-2017	56.082517	9.422146	0.71239	0.028	35.11	1030	CP
Salten River	SEPS-3	29-10-2017	56.056101	9.458571	0.70918	0.089	11.18		CF
<b>11. Addit</b>									
-	ADD-1	01-01-2018	56.059889	9.644173	0.71075	0.001	694.57	620	CP
Natural spring	ADD-3	01-01-2018	56.056287	9.617739	0.71011	0.051	19.63	310	CP
<b>12. Vejle Å (Fig. S1B)</b>									
Rosdam spring	RD-1	06-10-2017	55.658691	9.248123	0.70930	0.084	11.94	450	CP
Vejle River	VE-1	07-12-2017	55.650397	9.307452	0.70896	0.146	6.83		CF
<b>13. Egtved (Fig. S1H)</b>									
Fuglsang Forest	EGS-1	09-01-2018	55.596524	9.310741	0.71139	0.009	105.71	300	CP
Egtved Forest	EGS-2a	11-02-2018	55.596003	9.291486	0.71175	0.023	42.85	210	CP
Bølling Stream	BOE-1	11-02-2018	55.585392	9.344317	0.70945	0.133	7.51		CF
<b>14. Mejsling Forest (Fig. S1)</b>									
-	MEJ-1	11-02-2018	55.634461	9.454002	0.71300	0.019	51.65	250	CP
<b>15. Othillias Spring, Fillerup</b>									
-	OTH-1	10-11-2017	55.963641	10.119033	0.70936	0.232	4.31		EF
<b>16. Vejle Fjord Fig. S1K)</b>									
Lerbæk stream	LER-1	08-10-2017	55.678270	9.806090	0.70917	0.303	3.31	380	EP
Rosenvold River	ROS-1	08-10-2017	55.678064	9.808310	0.70908	0.400	2.50		EF
<b>17. Trelde Næs (Fig. S1L)</b>									
-	TN-1	16-10-2017	55.620047	9.836078	0.70847	0.323	3.10	400	EP
-	TN-2	16-10-2017	55.608214	9.823985	0.70897	0.270	3.70	850	EP
-	TN-3	16-10-2017	55.610126	9.825847	0.70869	0.316	3.16	500	EP
<b>18. Stenderup (Fig. S1M)</b>									
Stenderup Nørre Forest	SN-1	16-10-2017	55.489128	9.6444371	0.70944	0.196	5.10	400	EP
-	SN-2	16-10-2017	55.484884	9.649937	0.70949	0.232	4.31	850	EP
Solkær River	SOL-1	16-10-2017	55.459499	9.607834	0.70959	0.230	4.34		EF
<b>19. Pamhule Forest (Fig. S1I)</b>									
Sophies Spring	PS-1	03-10-2017	55.229121	9.436083	0.70886	0.338	2.96	500	EP
Usholt Brook	PS-2	03-10-2017	55.229984	9.437467	0.70953	0.168	5.94		EF
Pond	PS-3	12-12-2017	55.229067	9.387822	0.70949	0.163	6.13	360	EP
Pond	PS-4	12-12-2017	55.217326	9.402244	0.71016	0.062	16.07	425	EP
<b>20. Hytterkobbel</b>									
Hytterkobbel Forest	HYT-1	12-12-2017	55.161993	9.406865	0.71042	0.057	17.52	390	EP
<b>Reference samples</b>									
Rainwater, Odder		10-04-2017	55.972679	10.145713	0.708	0.00008	12177		
Holocene Foraminifera., Okinawa		17-10-2017			0.70918				
Holocene Foraminifera., Okinawa		16-01-2018			0.70918				
Holocene Foraminifera., Okinawa		16-01-2018			0.70918				
Holocene Foraminifera., Okinawa		28-09-2017			0.70917				
Holocene Foraminifera., Okinawa		28-02-2018			0.70918				
Holocene Foraminifera., Okinawa		14-06-2018			0.70918				

\*Code: W, Zone West Jutland; C, Zone Central Jutland; E, Zone East Jutland P, pristine sample; F, farmland sample

†76.1007, ‡76.240, number in National well-database GEUS-Jupiter database.

§Two samples were collected to illustrate the transition from pristine areas to farmland.

**Table S2. Basic statistical data on strontium isotopic composition ( $^{87}\text{Sr}/^{86}\text{Sr}$ ) and strontium concentration of pristine and farmland samples from West, Central and East Jutland.**

Strontium isotopic composition						
	West Jutland		Central Jutland		East Jutland	
	Pristine	Farmland	Pristine	Farmland	Pristine	Farmland
Number	37	15	12	4	10	4
Minimum	0.70905	0.70905	0.70930	0.70804	0.70847	0.70908
Maximum	0.71495	0.71099	0.71356	0.70945	0.71042	0.70959
Mean	0.71231	0.70972	0.71136	0.70890	0.70931	0.70939
Stand. Dev.	0.00130	0.00043	0.00130	0.00061	0.00062	0.00022

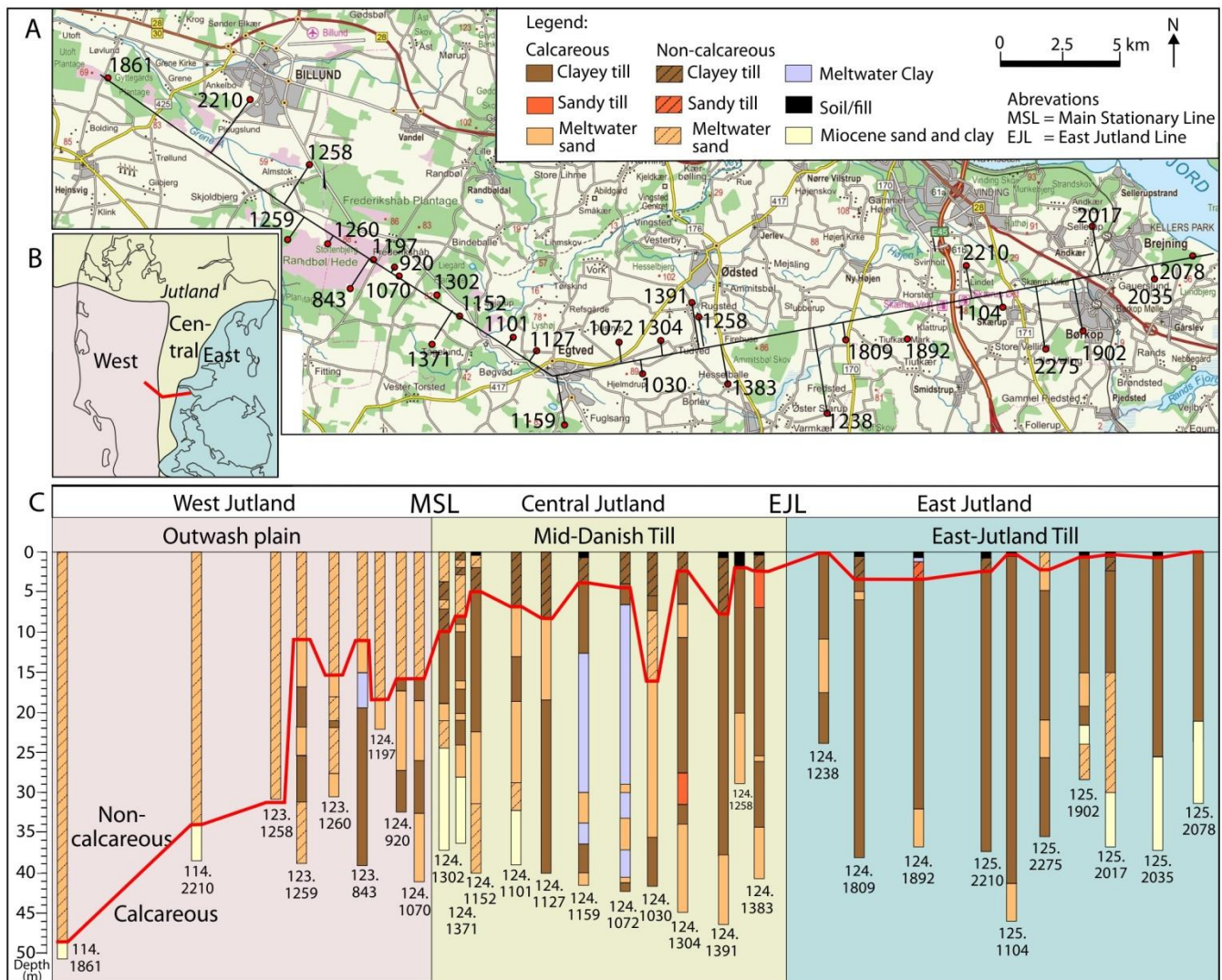
Strontium concentration						
	West Jutland		Central Jutland		East Jutland	
	Pristine	Farmland	Pristine	Farmland	Pristine	Farmland
Number	37	15	12	4	10	4
Minimum	0.002	0.048	0.001	0.045	0.057	0.168
Maximum	0.110	0.133	0.084	0.146	0.338	0.400
Mean	0.015	0.096	0.027	0.103	0.226	0.258
Stand. Dev.	0.019	0.023	0.026	0.046	0.104	0.010

**Table S3. Strontium composition of lime products, fertilizers and animal feed.**

Product	$^{87}\text{Sr}/^{86}\text{Sr}$	Sr conc. (ppm)
<b>Lime and lime products</b>		
Chalk, Dania (U. Maastrichtian)	0.70784	797
Chalk Gudumholm (U. Maastrichtian)	0.70786	1055
Mag. Limestone, England (Permian)	0.70753	58
2.5 % Mg-Chalk, Dankalk, Aggersund	0.70785	599
<b>Fertilizers (chemical)</b>		
NS 27-4	0.70741	214
NP 20-9	0.70344	38.4
Kali 49	0.71475	7.3
<b>Fertilizer (organic) (AKK)</b>		
Protamylasse (potato-based)	0.70773	0.1
<b>Local plant production</b>		
Maize	0.70821	11.3
Rolled Barley	0.70769	4.8
Barley, whole	0.70773	22.3
Concentrated feed (Import)	0.70973	34.5
<b>Imported animal feed</b>		
Soy meal Argentina A, (dlg)	0.70730	18.6
Soy meal Argentina B, (dlg)	0.70722	21.8
<b>Other soy products</b>		
Soy bean China A	0.70994	7.1
Soy bean China B	0.71012	5.1
Soy Austria A	0.71227	3.7
Soy Austria B	0.71230	3.9

**Table S4. Calculated strontium isotope composition and concentration for two representative farms in the Vallerbæk area, and for the strontium composition of manure.**

<b>Farm 1 - 80 hectares</b>						
Component	kg/ha/year	Sr (ppm)	g Sr/ha/year	$^{87}\text{Sr}/^{86}\text{Sr}$	% of total Sr	
NS 27-4 (Fertilizer)	450	214	96.3	0.70741	10.7	
Kali 49 (Fertilizer)	90	7.3	0.7	0.71475	0.1	
Agricultural Lime	1000	800	800	0.70785	89.2	
<b>Total</b>			<b>897</b>	<b>0.70781</b>		
<b>Farm 2 - 100 hectares</b>						
Component	kg/ha/year	Sr (ppm)	g Sr/ha/year	$^{87}\text{Sr}/^{86}\text{Sr}$	% of total Sr	
NP 20-9	50	38.4	1.9	0.70344	0.3	
NS 27-4	130	214	27.8	0.70741	3.7	
Kali 49	66	7.3	0.5	0.71475	0.1	
Protamylasse (K fertilizer)	2000	0.1	0.2	0.70773	0.0	
Manure	35000	2.2	76.9	0.70829	10.3	
Agricultural Lime	800	800	640	0.70785	85.6	
<b>Total</b>			<b>747</b>	<b>0.70787</b>		
<b>Manure</b>		$^{87}\text{Sr}/^{86}\text{Sr}$	Sr (ppm)	kg/day/cow	g Sr/day/cow	% of total Sr
Rolled Barley		0.70769	4.8	11	0.05	13.9
Whole Cut Grass		0.70773	22.3	11	0.24	64.1
Soy Pellets		0.70973	34.5	2	0.07	18.0
Water		0.71300	0.1	150	0.02	3.9
<b>Average Manure</b>		<b>0.70829</b>	<b>2.20</b>	<b>174</b>	<b>0.382</b>	



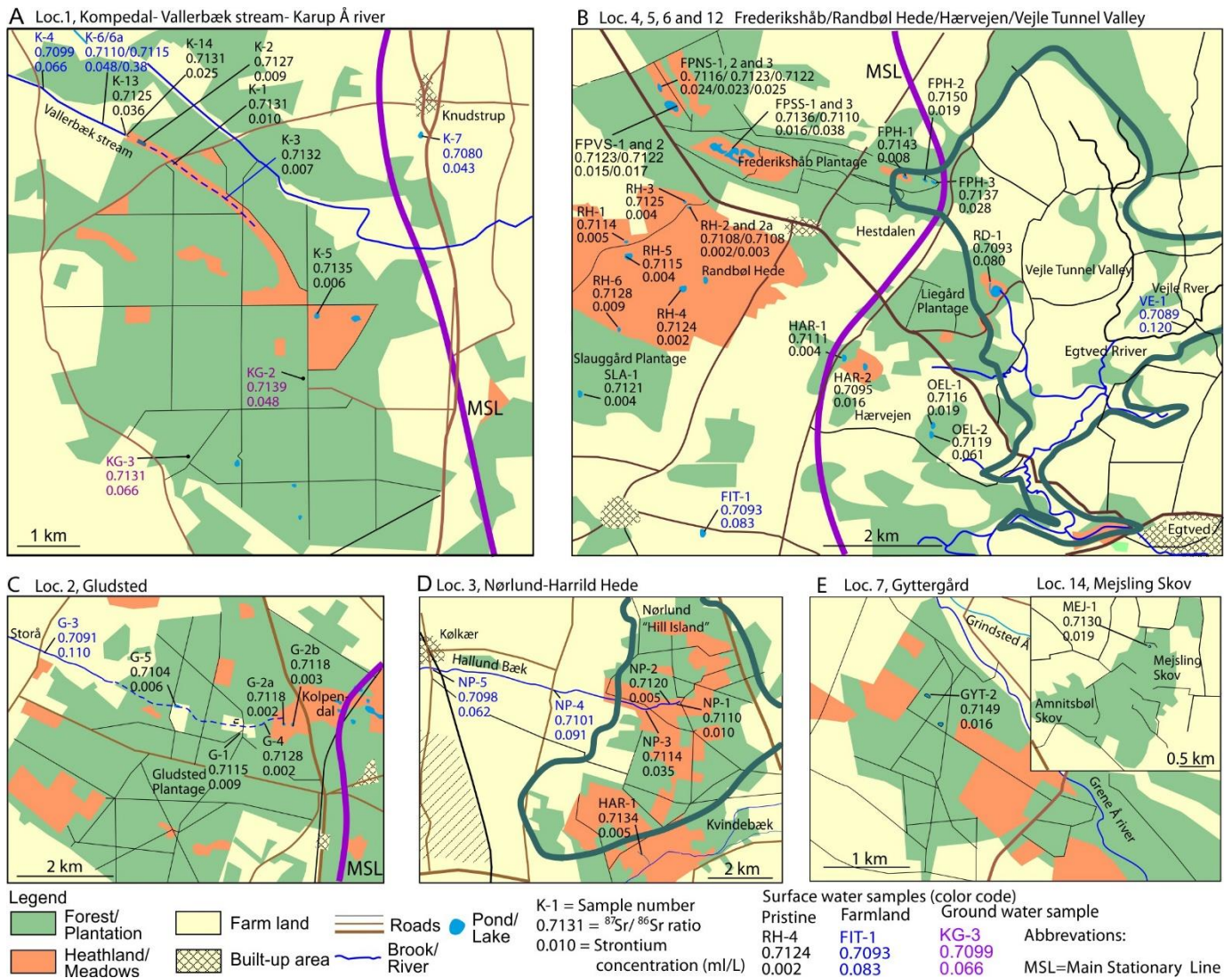
**Fig. S1. Calcium carbonate distribution in glaciogenic deposits in an east-west geological profile across middle Jutland from Bjerring to Billund.** (A) Map showing the location of examined wells. (B) Map showing the position of the geological cross section relative to the geological subdivision of the study area into West, Central and East Jutland (see main text Fig. 1). (C) Geological profile showing the thickness of the non-calcareous zone and the distribution of main lithological units. The cross section is based on data from the GEUS-Jupiter database of water wells and shallow borings in Denmark, maintained by the Geological Survey of Denmark and Greenland (GEUS). Maps in (A) and (B) are based on data from "Styrelsen for Dataforsyning og Effektivisering, skærkortet, WMS-tjeneste".

**fig. S1. Supplementary text.**

The glacial deposits of the study area are subdivided into three geological zones, termed West, Central and East (fig. S2). One of the most important differences between the three zones is the

content of calcium carbonate. Calcium carbonate in surface deposits is often dissolved due to leaching. However, the thickness of the leached zone varies. It is thickest in West and Central Jutland and thinnest in East Jutland (18), but the thickness of the non-calcareous zone is poorly known. The tills of East Jutland are clearly more calcareous than the deposits of West and Central Jutland. In 5\*5 km grid investigation, 3% and 5% of the samples from West and Central Jutland, respectively, contained calcium carbonate, compared to 50% of the samples from East Jutland (44).

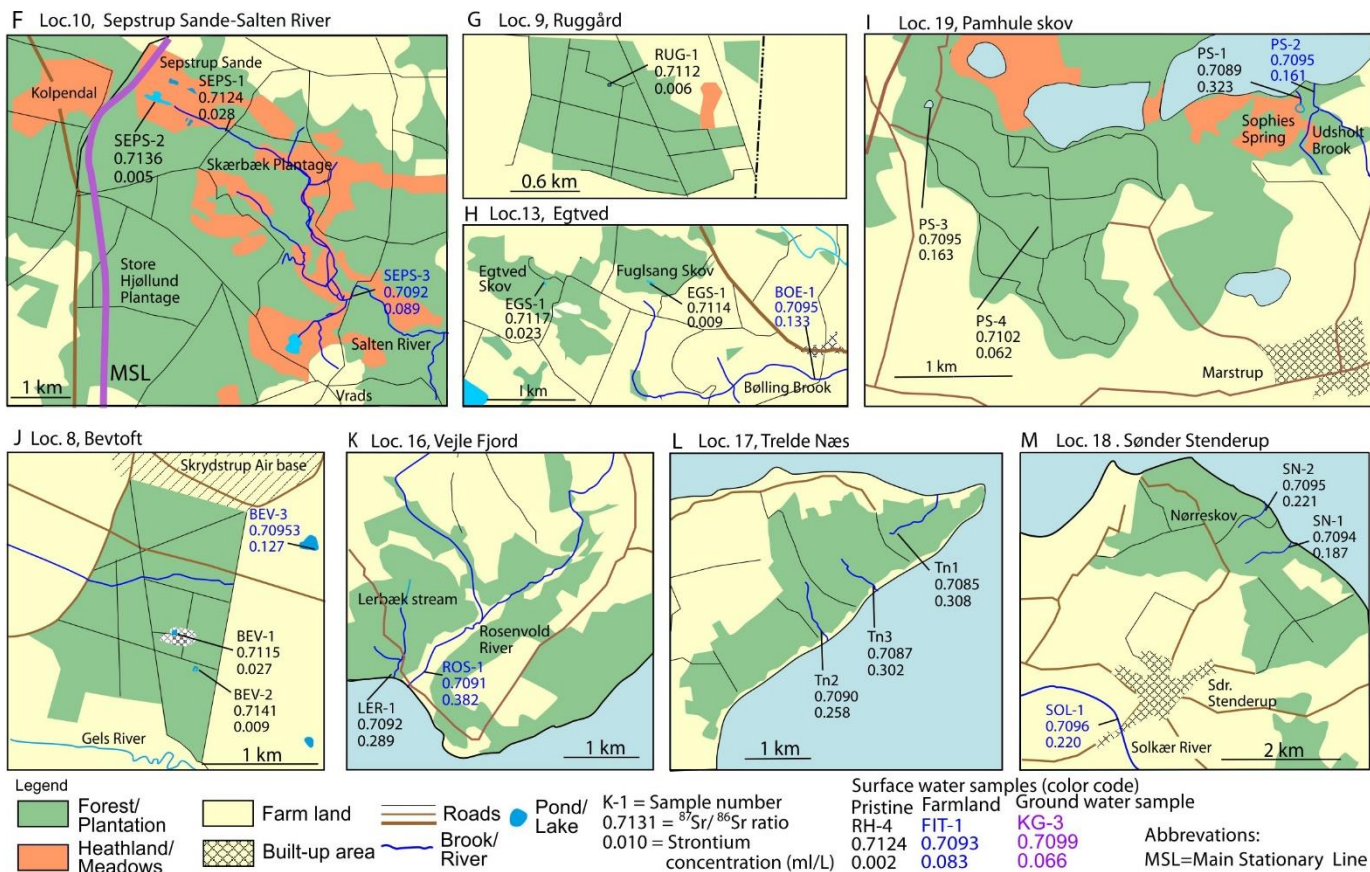
To compliment these rough estimates, we have examined the thickness of the non-calcareous zone in an east-west profile south of the Vejle Tunnel Valley from the tills of East Jutland to outwash plain of West Jutland (fig. S2). The cross-section passes through the most densely sampled part of the study area. The results clearly confirm previous estimates. In West Jutland, the thickness of the non-calcareous zone increases from about 15 m closest to the Main Stationary Line to more than 50 m about 15 km west of the line. In Central Jutland, the thickness of the non/calcareous zone varies between 2.5 and 16 m with an average thickness around 7 m. In East Jutland, the thickness varies between zero and 2-3 m.



**Fig. S2, part 1. Simplified maps of localities showing sample position and strontium data.**

The geographic location of localities is indicated in Fig.1B. The geographic coordinates, sampling date, and strontium data are given in table S1. For each sample, the  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio and the strontium concentration is indicated. Pristine samples are in black lettering; farmland samples are in blue lettering. Groundwater samples from pristine areas are in dark purple lettering (see fig. S1A). Maps are based on data from "Styrelsen for Dataforsyning og Effektivisering, skærkortet, WMS-tjeneste".





**Fig. S2, part 2. Simplified maps of localities showing sample position and strontium data.**

For explanation see text to fig. S2, part 1.

### Description of localities.

**Locality 1, Kompedal Plantage-Karup River** (fig. S1A). An overview of the locality is presented in main text Fig. 3. Here only the tributary Vallerbæk stream is shown. The locality is situated on the Karup Outwash Plain (Fig. 3). Vallerbæk stream flows in an approximately 150 m wide and up to 8 m deep erosional valley cut into the outwash plain during the deglaciation and Holocene. The uppermost 4 km of the stream runs within Kompedal Plantage and is classified as pristine. This part of the stream is tiny and often discontinuous at the surface. Sample K-7 was taken in a pond on farmland about 4 km NE of the forest and is hydrologically connected eastward to the Gudenå River system.

**Locality 2, Gludsted Plantage** (fig. S1C); **locality 10, Sepstrup Sande** (fig. S1F). Gludsted Plantage is located on the outwash plain in front of the tunnel valley at Sepstrup Sande (Loc. 10). The outwash plain and the tunnel valley are separated by the Main Stationary Line (MSL) (see main text). The streams on the outwash plain flow westward, while the streams at Sepstrup Sande flow eastward. The stream

running through Gludsted Plantage constitutes the beginning of Storå River. Within the forest, it is generally not continuous at the surface. Samples G-1 to G-5 were collected from pristine areas. Sample G-4 is from farmland 2 km east of the forest. Sample SADS-1 and 2 from the Sepstrup Sande tunnel valley are from pristine areas; SADS-3 is influenced by farming.

**Locality 3, Nørlund Plantage** (fig. S1D). Samples NP-1-NP-3 were collected on Nørlund “Hill Island” in the spring area of Hallund Brook, a tributary to the Skjern Å River. The upper part of the brook runs in a shallow meadow surrounded by coniferous forests. Samples NP-4 and NP-5 were collected from farmland on the outwash plain 1.7 km and 5 km, respectively, west of the forest. Sample HAR-1 is from Harrild Heath on the outwash plain south of Nørlund Hill Island. This pristine sample was collected from a side branch of Kvinde Brook.

**Locality 4, Frederikshåb Plantage; Locality 5, Randbøl Heath; Locality 6, Hærvejen** (fig. S1B). These localities are all located on the Grindsted outwash plain west of Vejle Tunnel Valley. The samples from Frederikshåb Plantage and Hestdalen were collected in a series of small lakes and ponds located in a shallow erosional valley cut into the outwash plain probably by meltwater from the tunnel valley. The lakes from Frederikshåb Plantage show highly variable water levels and they doubled in size during the fall of 2017. The samples from the very flat Randbøl Hede and Slauggard Plantage outwash plains were collected in ponds and small lakes of unknown origin. None of the samples from localities 4 and 5 are considered to have been affected by farming. The samples from Hærvejen have various origins including a natural spring and a raised bog (see table T1). Two samples (marked with red numbering in fig. S1B) are affected by farm land. The remaining samples are from pristine water.

**Locality 7, Gyttegård Plantage** (fig. S1E). The pristine sample was collected in an elongate pond completely surrounded by forest.

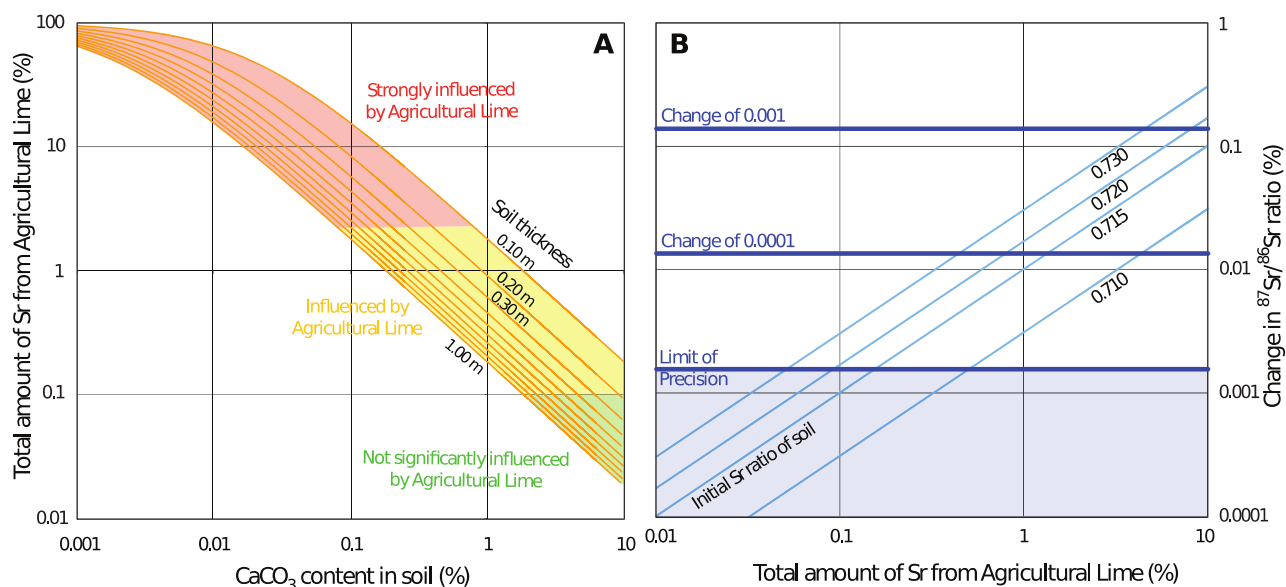
**Locality 8, Bevtøft Plantage** (fig. S1J). The locality is situated on the outwash plain separating Gram and the Toflund “Hill Islands” about 3 km east of the MSL and about 4 km southwest of the Bronze-age burial mound of the Skrydstrup Woman. Samples BEV-1 and BEV-2 were collected from two pristine ponds within the forest. However, the surroundings of BEV-1 are cleared for trees and are maintained with grass cutting machines and the pond may be contaminated. BEV-3 was sampled from an abandoned water-filled gravel pit located on farm land about 500 m east of the forest.

**Locality 9, Ruggård** (fig. S1G). The pristine sample was collected in a circular pond in the northern part of the forest.

**Locality 10**, see locality 2.

**Localities 12, Vejle Tunnel Valley** (Fig. S1B); **Locality 13, Egtved** (fig. S1H). Sample ROS-1 is from a natural spring situated at the end of the Vejle Tunnel Valley about 35 vertical meters below the outwash plain. It is classified as pristine. Sample VE-1 is from Vejle Å River and clearly affected by farming. The two samples from Fuglsang Forest and Egtved Forest are pristine, whereas the water sample from Bølling Brook is influenced by farming.

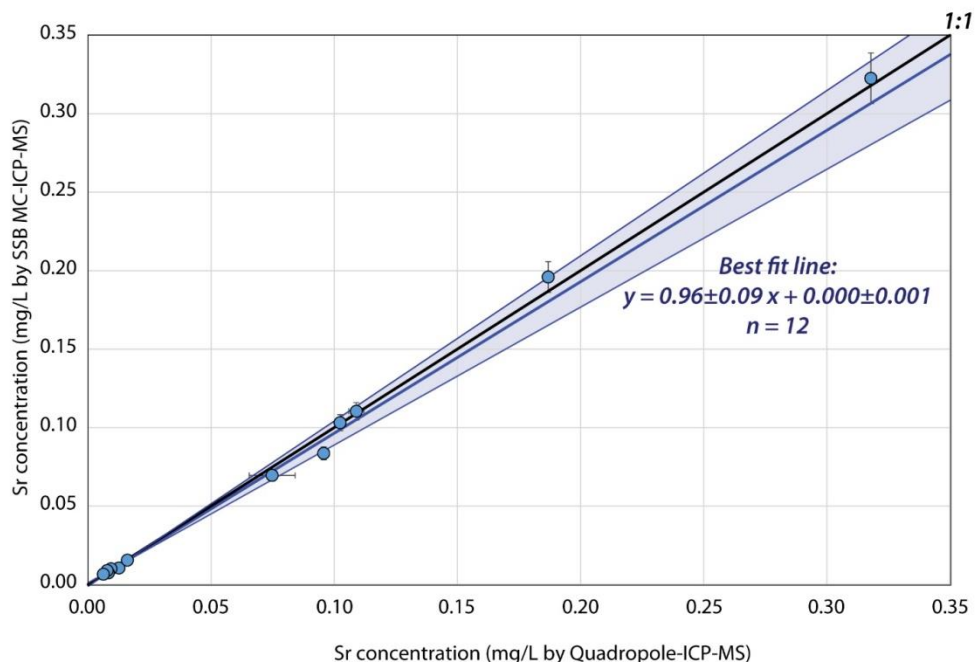
**Localities 16, Vejle Fjord** (fig. S1K); **Locality 17, Trelde Næs** (fig. S1L); **Locality 18. Sønder Stenderup** (fig. S1M) and **Locality 19, Pamhule Forest** (fig. S1I). **Locality 20, Hytterkobbøl**. These localities are all located in areas dominated by the calcareous, clayey East Jutland Till (Fig.1A,B). Samples were collected from brooks and streams except for the sample from Hytterkobbøl and the three pristine samples from Pamhule Forest (PS-1, PS-2 and PS-3), which were taken from small ponds and a spring.



**Fig. S3. Influence of soil thickness and soil composition on the level of disturbance of the  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio from agricultural Lime.** (A). The change in  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio as a function of how much of total strontium in a surface water sample is coming from agricultural lime (B). These calculations should be used as rough guidelines only as the Sr concentration in natural and agricultural lime and soil may vary, as may the Sr isotopic composition of agricultural lime used in different places.

**fig. S3. Supplementary text.**

How strongly the strontium isotopic composition of surface water is influenced by the use of agricultural lime is dependent on how much strontium there is in the soil, the isotopic composition of the soil and the thickness of the soil that the agricultural lime and rainwater effectively interact with. In fig. S3A, a simple estimate of the amount of the total Sr is coming from agricultural lime is given, based on how much calcium carbonate (or equivalent of other easily leachable source of Sr) is naturally in the soil, the thickness of the soil column the agricultural water is percolating through, and an average liming rate. For soils with less than 1%  $\text{CaCO}_3$ , the total amount of Sr coming from agricultural lime is around 1% or more. In fig. S3B a calculation of how much the Sr isotopic ratio shifts as function of the amount of agricultural lime and the natural Sr isotopic ratio is presented. The further the natural Sr isotopic ratio is from that of the agricultural lime (in this example a mid-Cretaceous limestone with  $^{87}\text{Sr}/^{86}\text{Sr} = 0.7078$  is used), the more it will be influenced by the agricultural lime. At a level of 1% Sr from agricultural lime the resulting shift in Sr isotopic composition is resolvable for all lithologies not dominated by carbonates and for areas with crustal signatures with high  $^{87}\text{Sr}/^{86}\text{Sr}$  ratios very small amounts of agricultural lime will severely alter the strontium isotopic composition of surface water.



**Fig. S4. Comparison of strontium concentrations measured by quadrupole ICP-MS and multi-collector ICP-MS.** Twelve samples were analyzed for their strontium concentration by both quadrupole ICP-MS (Agilent 7900) and standard-sample-bracketing multi-collector ICP-MS (Nu Plasma II). The results from the two methods are in good agreement, and suggests that the standard-sample-bracketing multi-collector give accurate results. An uncertainty of 5% is assigned to the Sr concentration data of the entire sample set measured by standard-sample-bracketing multi-collector ICP-MS, based on the reproducibility of standards.