1	SUPPLEMENTARY DATA
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3	Tidal erosion and upstream sediment trapping moderate records of land-use change in a
4	formerly glaciated New England estuary
5	
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22

#### 23 S.1 Supplementary Methods

#### 24 S.1.1. Fluvial and estuarine bulk environmental parameters

Water samples were collected from the Merrimack River upstream of the estuarine turbidity maximum during the April 2014 spring freshet, the highest water [and presumably, therefore, sediment] discharge event of the year, at the river surface (0 m) and in the water column at 2 m, 3 m, 4 m, 5 m in depth and near the bottom (< 1m from the bottom) (Figure S1). Bottom sediment samples were collected from the Plum Island and Rowley estuaries and upstream of the estuarine turbidity maximum of the Parker and Ipswich rivers with a Van Veen grab sampler (Figure S1).

Total organic carbon (TOC) and total nitrogen (TN) contents, and stable isotope values for 32 TOC and TN ( $\delta^{13}C_{TOC}$  and  $\delta^{15}N_{TN}$ ) were determined in bulk for bottom grab samples (Ipswich, 33 River, Parker River, Plum Island estuary, and Rowley estuary) at the WHOI Organic Mass 34 Spectrometry Facility. Prior to analysis, samples were freeze-dried, homogenized, and powdered. 35 Samples for TOC, TN,  $\delta^{13}C_{TOC}$  and  $\delta^{15}N_{TN}$  were analyzed in triplicate on an elemental analyzer 36 coupled to a Finnegan Deltaplus isotope ratio mass spectrometer (EA/IRMS). TN and  $\delta^{15}N_{TN}$ 37 compositions were measured on raw, powdered sample aliquots. TOC and  $\delta^{13}C_{TOC}$  were 38 determined following fumigation acidification of powdered sample aliquots Whiteside et al. 39 (2011). These sample aliquots were sealed in a vacuum desiccator with a beaker of 50 mL 12N 40 HCl, fumigated for 60 to 72 hours at 60–65°C to remove carbonates, and dried in a separate 41 desiccator for an additional 24 hours prior to measurement. 42

Water samples from the Merrimack River were vacuum filtered through 25 mm glass fiber
filters (Whatman GF/F, 0.7 lm pore size). Particulate organic carbon (POC) and particulate

- nitrogen (PN) were measured on acidified filters using an elemental analyzer (Flash EA 1112
- 46 Series, Thermo Electron Corporation) following the methods of Hedges and Stern (1984). POC
- 47 stable carbon isotope ( $\delta^{13}C_{POC}$ ) values from all Merrimack River water samples except the 5 m
- depth were measured on acidified filters using isotope ratio-mass spectrometry.  $\delta^{13}C_{POC}$  values
- 49 were measured using a Costech ECS 4010 CHNSO Analyzer interfaced with a Delta V
- 50 Advantage Isotope Ratio Mass Spectrometer with the Conflo IV Interface at the Virginia
- 51 Institute of Marine Science and are presented relative to Vienna Pee Dee Belemnite. Analytical
- 52 precision of  $\delta^{13}C_{POC}$  measurements was within 0.2‰. Total suspended sediment (TSS) was
- 53 collected on pre-weighed 47 mm polycarbonate membrane filters (Whatman, 1.0 lm pore size)

and rinsed with milli-Q water to remove salts. TSS was measured gravimetrically after drying to

a constant weight ( $\pm 0.5 \text{ mg}$ ) at  $105\pm5^{\circ}$ C.

# 56 S.1.2. Radionuclide uncertainty and detector calibration

Analytical uncertainties for the radionuclide measurements were calculated using standard 57 counting statistics (Kaste et al. 2011):  $1\sigma = \sqrt{n}/n$ , where n = the number of detected counts. An 58 average of 1000 counts for <sup>210</sup>Pb yields an uncertainty of 3.3%. An average of 3000 counts for 59  $^{226}$ Ra yields an uncertainty of 1.8%. An average uncertainty of 3.8% was calculated for  $^{210}$ Pb<sub>ex</sub> on 60 the basis of propagation of uncertainty ranges ("error") for <sup>226</sup>Ra (1.8%) and <sup>210</sup>Pb (3.3%). The 61 range of uncertainty for <sup>137</sup>Cs radioactivity is equivalent to the detection limit of 0.2 Bq/kg. The 62 ranges of uncertainty of the CRS model age estimates were calculated using methods from 63 64 Appleby (2001).

Shielded ultra-low background Canberra 5030 Broad Energy Germanium gamma detectors
 were calibrated for <sup>210</sup>Pb and <sup>226</sup>Ra using certified uranium ore, and for <sup>137</sup>Cs using a calibrated
 radionuclide solution. <sup>210</sup>Pb was corrected for self-attenuation using the point-source method
 (Cutshall et al. 1983). Unsupported <sup>210</sup>Pb (<sup>210</sup>Pb<sub>ex</sub>) was determined by subtracting the estimated
 <sup>226</sup>Ra radioactivity, calculated from the average of the activities of <sup>214</sup>Bi (609.3 keV photopeak)

and  $^{214}$ Pb (352 keV photopeak), from total  $^{210}$ Pb activity.

## 71 S.1.3. Bulk and stable isotope organic carbon and nitrogen uncertainty

- Average precision (2- $\sigma$ ) for replicate measurements of TOC, TN,  $\delta^{13}C_{TOC}$  and  $\delta^{15}N_{TN}$
- measurements at Sites A (bank-proximal) and E (river-proximal) were 0.01%, 0.03%, 0.3‰, and

0.22 - 0.44% (1  $\sigma$ ), respectively. Average precision (2- $\sigma$ ) for replicate measurements of TOC,

75 TN,  $\delta^{13}C_{TOC}$ , and  $\delta^{15}N_{TN}$  at Site C (central) were 0.03%, 0.03%, 0.3‰, and 0.4‰ (1  $\sigma$ ),

76 respectively

## 77 S.1.4. Determination of Pb Abundance by X-Ray Fluorescence

78 We used high-resolution (1-cm) XRF scans to approximate elemental lead (Pb) abundance data as a proxy for the onset of national-scale U.S. Pb production. Relative elemental 79 abundance measurements were made using an Avaatech XRF Core Scanner at the University of 80 North Carolina at Chapel Hill with a 200 second count time, 30kV voltage, and 1000 µA tube 81 82 current. We used the software program WinAxilBatch to process the raw data through the 30kV lang model available from Avaatech. We rely on relative changes in elemental abundance 83 approximated by counts because matrix and dilution effects, as well as sediment properties (pore 84 water, grain size), make conversion of XRF data to concentrations difficult without additional 85 geochemical analysis (see Gregory et al. 2015 and references therein). Any use of trade, firm, or 86

product names is for descriptive purposes only and does not imply endorsement by the U.S.

- 88 Government.
- 89

## 90 S.2. Supplemental Results

## 91 S.2.1. Pollen assemblages

92 Two common pollen assemblages that indicate agricultural (land clearing) horizon markers, Ambrosia (ragweed) and Cerealia (cultivated grasses), were used for this study (Figure 7), 93 though down-core pollen count data for the entire assemblage were collected (see Tables S1, S2, 94 and S3). At Site A (bank-proximal), Ambrosia is first present (> 2% total abundance) at and 95 above 30.5 cm and Cerealia grains are first present at and above 35.5 cm. For Site C (central), 96 Ambrosia pollen is absent (<2% total abundance) below 22.5 cm and reaches a maximum at 17.5 97 cm, while *Cerealia* grasses are first present at and above 21.5 cm. At Site E (river-proximal), 98 Ambrosia abundance is variable throughout the upper 50 cm. 99

# 100 S.2.2. Bulk organic and stable isotope stratigraphy

101 TOC content is < 7% for all samples. At Site A (bank-proximal), grain-size-normalized TOC 102 (TOC) increases from an average value of 0.01 ( $\pm$  0.002) below 35 cm to an average value of 103 0.03 ( $\pm$  0.01) above 35 cm. TOC values from Site C (central) are generally higher (average 0.06  $\pm$ 104 0.03) between 140 – 60 cm, and lower (average 0.03  $\pm$  0.01) above 60 cm. For Site E (river-105 proximal), TOC values are also higher (average 0.07  $\pm$  0.04) from 140 – 50 cm and lower above 106 50 cm (average 0.04  $\pm$  0.02). Measured TN is also generally low (< 0.5%) in all samples, and are 107 thus interpreted with caution; however, trends of grain-size-normalized TN (TN) generally

108 mimic those of normalized TOC ( $R^2 = 0.96$ ).

 $\delta^{13}$ C values at bank-proximal Site A range between -20.3 and -24.0‰ with samples below 35 109 cm more depleted (-24.0 to -23.3‰) than between 35 and 10 cm (-21.9 to -20.3‰). Values 110 above 10 cm values range from -23.2 to -23.9‰). Both sites C (central) and E (river-proximal) 111 display trends with depth. At Site C (central) values for  $\delta^{13}$ C shift from -20.8 – -17.2‰ below 60 112 cm towards more negative values (-21.4 – -23.8‰) in the upper 60 cm. Values for  $\delta^{13}$ C at Site E 113 (river-proximal) shift from -15.2 - 21.7% below 60 cm to more negative values (-23.5 - -114 24.6‰) in the upper 60 cm. Values for  $\delta^{15}$ N are similar to trends for  $\delta^{13}$ C at Site A (bank-115 proximal), in which values are within a narrow range (3.4 to 5.6%).  $\delta^{15}$ N values differ at sites C 116 (central) and E (river-proximal), with  $\delta^{15}$ N peaks at Site C (central) at 80 cm (5.2 ± 0.1‰), 20 117 cm ( $4.8 \pm 0.1\%$ ), and 0 cm ( $4.5 \pm 0.4\%$ ), and a gradual up-core enrichment at Site E (river-118

proximal), ranging from 1.1‰ near the base to 4.1‰ at the top of the core.

# 120 S.2.3. Fluvial and estuarine bulk environmental parameters

Analysis of water and sediment samples from local rivers and estuaries provides baseline bulk environmental data from possible sediment sources to Joppa Flats (Table S4; Figure S1). TSS in the Merrimack River samples ranges from 9.6 to 15.0 mg/L and generally increases with depth in the water column. PN and POC from the Merrimack River suspended sediments range from 0.69 to 0.76% and from 6.43 to 7.41%, respectively. Bottom sediments from the Parker and Ipswich rivers and the Plum Island and Rowley estuaries generally contain low TOC (range: 0.04 to 0.97%) and TN (range: 0.01 to 0.07%) values.  $\delta^{13}C_{POC}$  measurements from the Merrimack River display no trend with depth in the water column. Merrimack River suspended sediment

River display no trend with depth in the water column. Merrimack River suspended sediment samples and bottom sediment samples from the Parker and Ipswich rivers are generally more

depleted (average  $\delta^{13}C_{TOC}$ : -28.5 ± 0.7‰) than the estuarine bottom sediment samples (average

130 depicted (average of Croc. -28.5  $\pm$  0.7 ‰) that the estuarme bottom sedment samples (average 131  $\delta^{13}C_{TOC}$ : -20.7  $\pm$  0.2‰).

#### 132 S.2.4. Pb Abundance

The initial increase in relative elemental Pb between 10 and 20 cm downcore is interpreted as the onset of national-scale U.S. lead production in ca. 1875 CE (Kemp et al. 2012), while the continued high levels are likely associated with the use of leaded gasoline in automobiles (Figure S4). The increase in elemental Pb is approximately coincident with the increase in <sup>210</sup>Pb in both cores, and therefore provides a secondary chronographic marker for the 20<sup>th</sup> century. The area of increased Pb abundance between 80 and 110 cm at Site C (central) is interpreted to be the result of a fine-grained organic-rich sediment layer.

140

#### 141 S.3. Supplementary tables

142 **Table S1.** Genus/family-level palynomorph abundance data from Site A (bank-proximal)

DEPTH (cm)	15.5	20.5	25.5	30.5	35.5	40.5
TREES/SHRUBS						
Abies			1	2	1	1
Acer	2		1	1	2	
Alnus	6	9	9	5	7	9
Betula	40	36	28	36	1	45
Carya	4	7	3	6	7	9
Celtis				1		
Cephalanthus	1					
Cornus			1			
Corylus	1					3
Cupressaceae				4		
Fagus	1	4		2	3	1
Fraxinus					1	
llex	2				2	1
Juglans	1		1	1	1	1
Liquidambar						
Myrica	1					
Nyssa	1	4	10	5	4	5
Ostrya/Carpinus	1		1	1		2
Picea glauca	19	12	15	19	16	12
Pinus indet.	92	77	104	81	92	77

Pinus strobus	16	7	22	46	45	54
Quercus	49	66	48	51	56	57
Salix					1	1
Tilia	2	1				
Tsuga	36	43	60	56	57	66
Ulmus	2	4	7	4	2	2
HERBS						
Amaranthaceae	2	2			1	1
Ambrosia	13	16	8	9	5	4
Artemisia	1	1				
Asteraceae indet.	2	1	3	2	3	
Cerealia				1	1	
Cyperaceae	2	1		1		
Fabaceae			1			
Heliotropum	1					
Liguliflorae					1	
Nymphaea	2					
Onagraceae	1					
Poaceae	5	9	8	10	3	5
Polygonum	1	1				
Rumex cf. acetosella			1	2	1	3
Typha	1					
LOWER VASCULAR PLA	NTS					
Osmunda		1	9	4	5	7
Lycopodium lucidulum			3	3		2
Sphagnum					1	
Monolete spores	5	11	7	11	8	
Trilete spores	1	3	3	3	5	5
ANGIOSPERM INDET.						
Tricolpate pollen	2	1		4	3	
Tricolporate pollen		2				
Triporate pollen	1	3	1	1		1

TOTAL COUNTED 317 322 355 372 335 374

143 Note: Ambrosia and Cerealia (bold) abundances were used in this study to identify the onset of agricultural land

144 clearing by Europeans in the Merrimack River watershed.

DEPTH (cm)	0.5	3.5	8.5	12.5	17	18	19	20	21	21.5	23	24	24.5	26	31	35.5	41
TREES/SHRUBS																	
Abies	5	3	4	2	1	2	1	1	3	4	5	4	11	1	7	13	19
Acer	3	3	4	4	1	1	2	2	3	2	1		2	2	1	2	3
Alnus	3	2		2	8	12	1	4	17	7	14	9	3	10	9	9	13
Betula	32	16	29	28	29	47	38	44	51	60	61	29	39	63	47	57	70
Carya	4	3	11	13	5	5	4	4	2	6	6	2	5	6	3	4	6
Castanea										1							
Celtis					1				1	1						1	
Cephalanthus	2		3	1							2	1					
Cornus					1									1			1
Corylus			2		1	5	1	2	3	3	1	1		2	1	3	2
Cupressaceae										1							
Fagus						3			1	1		2	1	9	6	4	6
Fraxinus	2	1	1				3	1	7	4	2	3	1	6	3	2	2
llex						1		1	1	1					2		3
Juglans	3	1						1			1	1					
Liquidambar	1		1	2			1						2	2		1	
Liriodendron			1														
Lythrum				1						1			1				
Myrica								1	1								1
Nyssa	7		1	6	7	8	14	12	1	8	10	6	14		5	8	3
Ostrya/Carpinus	1	1			2		1			1	1		1				1
Picea glauca	7	12	7	7	15	14	18	19	8	17	14	15	13	13	15	15	12
Picea mariana									1					1		1	2
Pinus indet.	39	60	42	50	80	54	40	44	50	44	44	37	48	60	75	76	75
Pinus strobus	90	100	86	76	74	81	86	64	48	59	63	66	75	52	47	21	28

**Table S2.** Genus/family-level palynomorph abundance data from Site C (central).

Quercus	89	49	71	86	69	96	75	66	76	95	107	77	88	94	69	61	80
Rosaceae											1						
Salix									2	4	1		3	2	2	1	1
Tilia		1			3	2	1	2			1			3		1	1
Tsuga	23	29	27	24	40	29	45	37	46	35	37	35	43	44	56	57	72
Ulmus	6	2	3	3	4	4	3	6	5	2	5	4	6	3	5	2	4
HERBS																	
Amaranthaceae	2	4		2		1	2	1	2	1	2				1		1
Ambrosia	7	4	9	3	3	17	7	10	12	11	11	2	7	3	8	5	5
Artemisia	1		2							1							1
Asteraceae indet.	4		3	1		3	4	3	1	3	2		1	4	6	2	2
Caryophyllaceae									1		1						
Cassia	1	1															
Cerealia		1			1	1				2							
Cyperaceae	1		2			1	1	5	5	2	1	2	2	1	5	2	1
Fabaceae				1	1					1	2		2	1			
Liguliflorae			1	1		1								1			1
Plantago						1			1	1	1						
Poaceae	9	5	5	11	10	21	12	20	15	16	15	8	9	7	11	7	15
Polygonum			1			1		1									1
Rumex cf. acetosella			1		2	4	1			1	2	1		4	3		1
Sagittaria									1								
Solanaceae				1													
Typha	2	1						1	1	3		2	2				
Vitis	1								2					1			

LOWER VASCULAR PLANTS																	
Osmunda	3	5	2	6	5	7	4	4	6	2	6	1	6	1	1	1	7
Lycopodium complanatum					2												
Lycopodium lucidulum								1	1				1		1	1	2
Pteris							1										1
Trilete spores		2	1	2	5	10	7	4	5	16	5	6	3	12	6		6
Monolete spores		8	6	12	12	7	8	4	4	13	9	8	10	8	5		5
Zonate spores														3			
ANGIOSPERM INDET.																	
Tricolpate pollen			3		2		4		3	4		1	2	4	2	3	7
Tricolporate pollen				1					2	2	1		1		2		
Triporate pollen				1							3			1	2	1	2
UN/Crumpled	1	1	4	5	3	3	5	1	2	2	3	1	2	5	3	1	4
TOTAL	348	314	329	347	384	439	385	365	389	436	438	323	402	425	406	361	463
Note: Ambrosia and Ce	erealia (	bold) ab	undance	s were us	ed in thi	s study to	o identify	the onse	et of agri	cultural l	and clear	ing by E	uropeans	in the N	Ierrimac	k River	

147 watershed.

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DEPTH (cm)	1.8	10.5	15.5	17.5	20.5	22.5	25.5	30.5	5
TREES/SHRUBS									
Abies	1	5	4	1	1		5	2	
Acer	6		2	3	1	2	3	2	
Alnus		4	2	5	1	8	4	6	
Betula	27	35	32	35	41	39	45	69	
Carya	11	3	3	5	5	9	5	7	
Cephalanthus	3			1	1	1			
Corylus			1				1	3	
Cupressaceae						1	1		
Fagus		2		1				1	
Fraxinus		1	2	8	3	8	5		
llex							1	2	
Juglans	1			1	1	2	2	1	
Liquidambar		1	1						
Nyssa		5	2	2	4	2	1	8	
Ostrya/Carpinus						2		1	
Picea glauca	6	7	5	7	8	5	3	10	
Pinus indet.	54	28	33	72	36	54	32	48	
Pinus strobus	65	34	69	37	82	50	35	61	
Quercus	59	58	78	73	90	95	68	62	
Salix	1	1	1		7	1	1		
Tilia	2				8	1	1		
Tsuga	37	10	18	21	21	14	25	49	
Ulmus	2	4	2	3		2	4	5	
HERBS									
Amaranthaceae	2	3	3	2		3	3		
Ambrosia	5	17	15	3	3	5	12	6	
Artemisia		5	4	2	1	1	2		
Asteraceae indet.	3	7	3	4		3	8	3	
Campanula							1		
Caryophyllaceae		1							
Cerealia	1				1			2	
Cyperaceae		2	1		1	1	1	3	
Ericaceae		1	1					2	
Fabaceae		1		3			1	1	
Heliotropum						1			
Lamiaceae		1							

**Table S3.** Genus/family-level palynomorph abundance data from Site E (river-proximal).

Liguliflorae							2		
Parthenocissus				1					
Plantago		2	1				2		
Poaceae	8	17	13	18	14	18	14	4	4
Rumex cf. acetosella			1	1			1		
Sagittaria				2		1			
Solanaceae					1				
Typha	1		1	2		4			
Vitis					1				
LOWER VASCULAR PLANT	ſS								
Osmunda	2		2	2	1	2	1	9	1
Lycopodium complanatum							1	1	
Lycopodium lucidulum				1					
Trilete fern spores	1	1	6	3	2	5	9	3	2
Monolete fern spores	3	3	6	7	2	4	7	6	5
ANGIOSPERM INDET									
		З			1	2			
Tricolporate pollen	1	0	2	З	2	2	З	1	
Triporate pollen			1	0	2		0		1
Total	302	262	315	329	340	346	310	378	356

Total302262315329340346310378356Note: Ambrosia and Cerealia (bold) abundances were used in this study to identify the onset of agricultural land 149

clearing by Europeans in the Merrimack River watershed. 150

152	Table S4.	Bulk envir	onmental	properties	from wa	ter samples	of the M	Ierrimack Rive	er (Sample II	O MR ## Apr14) and	d Van V	√een grab

samples of the Parker and Ipswich rivers and Rowley and Plum Island estuaries.

Sample ID	Average TSS (mg/L)	TSS SD (mg/L)	TOC (%)	TN (%)	C:N	δ <sup>13</sup> C (‰ VPDB)	δ <sup>15</sup> N (‰)
MR Surf Apr14	9.7	1.2	6.43	0.69	9.38	-28.68 ± 0.07	18.26 ± 6.25
MR 2m Apr14	9.6	1.1	6.48	0.79	8.20	-28.85 ± 0.11	24.64 + 12.62
MR 3m Apr14	12.6	0.3	6.72	0.71	9.45	-28.85 ± 0.22	14.88 ± 0.65
MR 4m Apr14	11.9	1.3	7.21	0.72	9.97	-28.66 ± 0.08	15.26 ± 8.28
MR 5m Apr14	11.6	0.8	7.56	0.84	8.99	N/A	N/A
MR Bot Apr14	15.0	0.3	7.41	0.76	9.80	-28.58 ± 0.83	23.343 ± 7.55
PR (Parker River)	N/A	N/A	0.97	0.07	16.7	-26.80 ± 0.31	5.94 ± 0.26
IR (Ipswich River)	N/A	N/A	0.34	0.02	17.1	-28.75 ± 0.11	$6.24 \pm 0.48$
RRE (Rowley River Estuary)	N/A	N/A	0.87	0.07	14.2	-20.84 ± 0.25	3.67 ± 0.34
PIE (Plum Island Estuary)	N/A	N/A	0.04	0.01	9.4	-20.48 ± 0.25	4.47 ± 0.19

# 154 S.4. Supplementary Figures





157 sample codes (Image: Landsat).



**Figure S2.** Comparison of relationship between grain size (% mud) and total organic carbon (left) and total nitrogen (right) contents prior to (top) and following (bottom) normalization to grain size. Note the reduced  $R^2$  (linear regression) values following normalization.

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Figure S3. Topobathymetric LiDAR from the U.S. Army Corps of Engineers (2018) reveals
evidence of increased deposition north of Site E (river-proximal) and increased scour south of
Site E, providing further evidence for the interpretation of increased mixing in the upper 50 cm
of sediment at Site E.



167

168 **Figure S4**. Relative downcore abundance (counts) of elemental Pb at Sites A (bank-proximal)

- and in C (central) derived from X-ray fluorescence (XRF) scans.
- 170

## 171 S.5. Supplementary References

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