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journal homepage: [www.elsevier.com/locate/epsl](http://www.elsevier.com/locate/epsl)Rapid emplacement of the Central Atlantic Magmatic Province as a net sink for CO<sub>2</sub>Morgan F. Schaller<sup>a,\*</sup>, James D. Wright<sup>a</sup>, Dennis V. Kent<sup>a,b</sup>, Paul E. Olsen<sup>b</sup><sup>a</sup> Earth and Planetary Sciences, Rutgers University, 610 Taylor Rd. Piscataway, NJ 08854, USA<sup>b</sup> Lamont–Doherty Earth Observatory of Columbia University, Rt. 9W, Palisades, NY 10964, USA

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## ABSTRACT

Recent evidence from the ~201.5 Ma Central Atlantic Magmatic Province (CAMP) in the Newark rift basin demonstrates that this Large Igneous Province produced a transient doubling of atmospheric pCO<sub>2</sub>, followed by a falloff to pre-eruptive concentrations over ~300 kyr. This paper confirms the short-term findings from the Newark basin, and tests the million-year effects of the CAMP volcanism on Early Jurassic pCO<sub>2</sub> from strata in the corollary Hartford basin of Eastern North America (ENA) also using the pedogenic carbonate paleobarometer. We find pCO<sub>2</sub> levels for pre-CAMP background of 2000 ± 700 ppm (at  $S(z) = 3000 \pm 1000$  ppm), increasing to ~5000 ± 1700 ppm immediately above the first lava flow unit, consistent with observations from the Newark. The longer post-extrusive Portland Formation of the Hartford basin records a fourth pulse of pCO<sub>2</sub> to ~4500 ± 1200 ppm, about 240 kyr after the last lava recorded in the ENA section. We interpret this fourth increase as due to a major episode of volcanism, and revise the main CAMP duration to 840 ± 60 kyr. The Portland also records a post-eruptive decrease in pCO<sub>2</sub> reaching pre-eruptive background concentrations of ~2000 ppm in only ~300 kyr, and continuing to levels below pre-CAMP background over the subsequent 1.5 Myr following the final episode of eruptions. Geochemical modeling (using modified COPSE code) demonstrates that the rapidity of the pCO<sub>2</sub> decreases, and fall to concentrations below background can be accounted for by a 1.5-fold amplification of the continental silicate weathering response due to the presence of the CAMP basalts themselves. These results demonstrate that a continental flood basalt capable of producing a short-term perturbation of the carbon system may actually have an overall net-cooling effect on global climates due to a long-term net-decrease in pCO<sub>2</sub> to below pre-eruptive levels, as previous models have suggested followed the emplacement of the Deccan Traps.

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## 1. Introduction

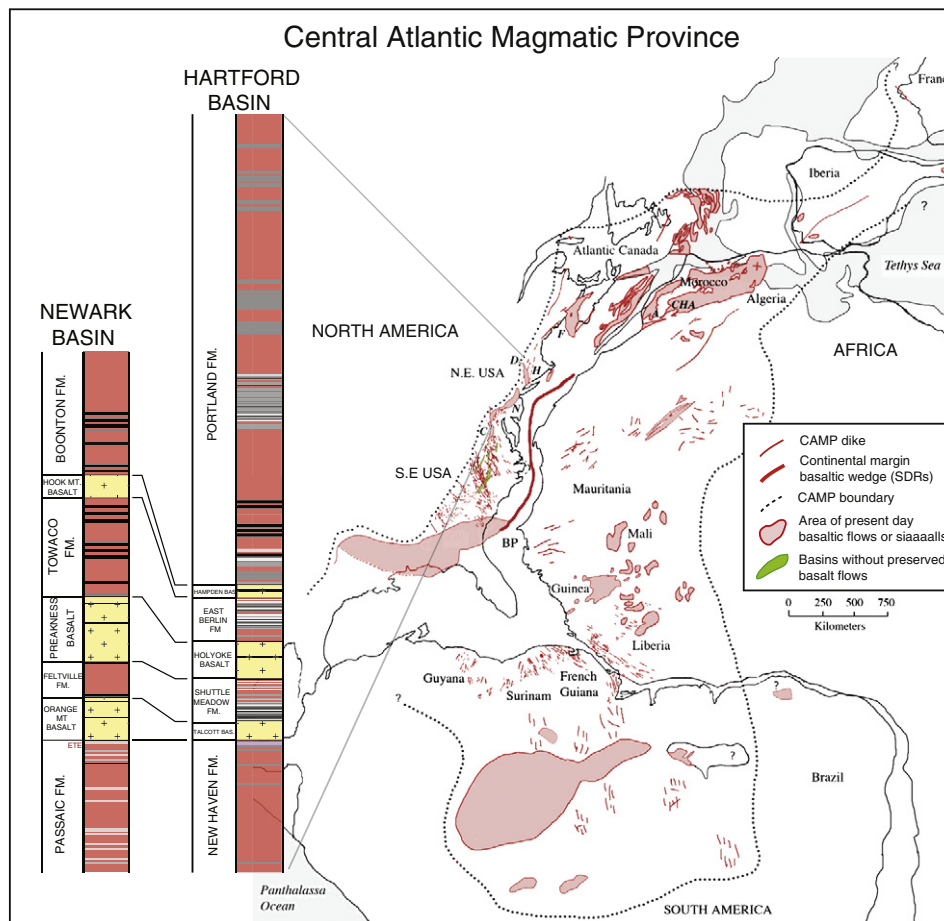
Covering millions of square kilometers of continental area, continental flood basalts are Large Igneous Provinces (LIPs) that may produce in excess of several million cubic kilometers of lava (Coffin and Eldholm, 1993). The great volume and aerial extent of these eruptions, and their intriguing association with mass extinction events throughout earth history (Courtilot and Renne, 2003; Wignall, 2001), has spurred interest in the potential of LIPs to alter the composition of Earth's atmosphere through the emission of gases – primarily SO<sub>2</sub> and CO<sub>2</sub> (Self et al., 2008). Until recently, geochemical modeling and effusive flux estimates seemed to suggest that CO<sub>2</sub> degassed during continental flood volcanism probably has a negligible long-term effect on the atmosphere (e.g., Caldeira and Rampino, 1990; Self et al., 2006). Most recently, Schaller et al. (2011a) tested the direct effects of a LIP using the pedogenic carbonate paleobarometer (Cerling, 1999) on sediments in superposition with the extrusives of the Triassic–Jurassic Central Atlantic Magmatic Province (CAMP) in the Newark basin of eastern North America (Fig. 1).

Schaller et al. (2011a) demonstrated that the short term (10<sup>4</sup>–10<sup>5</sup>-year) effects of a continental flood basalt on atmospheric pCO<sub>2</sub> may be substantial – a doubling of pCO<sub>2</sub> in apparent response to each magmatic episode – and yet quite transient with a steady decrease in atmospheric pCO<sub>2</sub> over the ~300 kyr following each volcanic episode.

Several independent geochemical models have predicted such a transient response following the eruption of the Deccan Traps (Caldeira and Rampino, 1990; Dessert et al., 2001). The model of Dessert et al. (2001) specifically predicts atmospheric CO<sub>2</sub> concentrations falling some 20% below pre-eruptive background by about 1 million years after the eruption of the Deccan Traps, an effect attributed to an acceleration of global silicate weathering further amplified by the presence of fresh weatherable basalt. However, due to the absence of continuous sedimentary sections where the appropriate lithology is interbedded with volcanics, such a response to any LIP other than the CAMP has not yet been explicitly identified. Encouragingly, immediately post-eruptive pCO<sub>2</sub> decreases have been observed in the Newark basin following the CAMP eruptions (Schaller et al., 2011a), and the corollary Hartford basin of eastern North America contains a much longer post-extrusive section (Kent and Olsen, 2008) where the long-term response of the system may be tested. In this paper we first use the pedogenic carbonate paleobarometer (Cerling, 1999) to demonstrate increases in pCO<sub>2</sub> coincident with

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**Fig. 1.** The distribution of the remaining CAMP volcanics and stratigraphy of the Newark and Hartford basins of Eastern North America. Modified from Whiteside et al. (2007), which is based on McHone (2000).

pulses of CAMP volcanism in the equivalent strata of the Hartford basin. We further use the thick post-CAMP Portland Formation to evaluate the long-term decrease in  $p\text{CO}_2$  as a function of increased global silicate weathering, testing the hypothesis that the presence of  $2\text{--}4 \times 10^6 \text{ km}^3$  of fresh CAMP basalt (McHone et al., 2003) is capable of reducing  $p\text{CO}_2$  to below pre-eruptive background concentrations.

## 2. The CAMP record in the Newark and Hartford basins

The Central Atlantic Magmatic Province (CAMP; Marzoli et al., 1999) is perhaps the largest continental flood basalt of the Phanerozoic, with latest Triassic and earliest Jurassic tholeiitic basalts and mafic intrusions preserved on four continents, and an aerial extent of greater than  $1.12 \times 10^7 \text{ km}^2$  (McHone et al., 2003) (Fig. 1). CAMP basalts span the Mesozoic rift zone of the Pangean supercontinent, and despite heavy erosion, are well exposed in the rift basins of eastern North America and Morocco, where they have been extensively studied. More poorly understood constituents of the CAMP are preserved in West Africa, Europe, and extensive regions of South America. However,  $^{40}\text{Ar}/^{39}\text{Ar}$  radiometric ages for this vast collection of lava flows and intrusives are essentially indistinguishable within the dating error (e.g., Hames et al., 2000; Jourdan et al., 2009; Knight et al., 2004; Marzoli et al., 2004; Marzoli et al., 2011; Nomade et al., 2007; Verati et al., 2007), and center the eruptive events around  $\sim 201 \text{ Ma}$ . More precise U–Pb dates place some of the first flow units at  $201.38 \pm 0.31 \text{ Ma}$  (Schoene et al., 2010) in Eastern North America.

The half-graben rift basins of the Newark Supergroup in Eastern North America contain Late Triassic to Early Jurassic age strata, the

majority of which are shallow to deep lacustrine, fluvial, alluvial and playa facies (Olsen, 1997; Smoot, 1991). The lacustrine sedimentary units in each basin display a rhythmic succession of facies that reflect periodic fluctuations in lake depth (Van Houten, 1962), which have been related directly to Milankovitch orbital forcing of tropical precipitation (Olsen, 1986; Olsen and Kent, 1996). The stratigraphy of the Newark basin is precisely known from a continuous cored section through the center of the basin (Kent et al., 1995; Olsen et al., 1996a), and this record of orbitally forced sedimentation provides the astronomical chronometer used to construct the geomagnetic polarity time scale for more than 33 Myr of the Late Triassic and Early Jurassic (Kent and Olsen, 1999; Kent et al., 1995; Olsen and Kent, 1996, 1999).

The CAMP extrusive interval sits near the top of the Newark basin section and is composed of three lava flow units (stratigraphically upward, the Orange Mountain, Preakness, and Hook Mountain Basalts), with significant sedimentary sequences above each of them (Feltville, Towaco, and Boonton Formations, respectively) (Figs. 1 and 4). Milankovitch cycle-stratigraphy on these sediments has constrained the duration of volcanism in the Newark basin to  $\sim 600 \pm 20 \text{ kyr}$  (Olsen et al., 1996b; Olsen et al., 2003; Whiteside et al., 2007), where the Feltville Formation represents  $\sim 260 \text{ kyr}$ , the Towaco Formation  $\sim 290 \text{ kyr}$ ; the first flow unit (Orange Mountain Basalt) was apparently extruded within a single precession cycle. The Hook Mountain Basalt is also within a precession cycle whereas the duration of the Preakness Basalt can presently only be constrained to  $\sim 60 \pm 20 \text{ kyr}$  (Whiteside et al., 2007). In these primarily lacustrine units, paleosols with varying degrees of pedogenic carbonate development formed during the dry phases associated with low lake

depth. Schaller et al. (2011a) used these soils to test both the magnitude and duration of the effects of the CAMP volcanism on atmospheric pCO<sub>2</sub>.

In the Hartford basin, the oldest strata are the fluvial and marginal lacustrine facies of the Triassic New Haven Formation, where pedogenic carbonate-bearing paleosols are abundant (Hubert, 1978). The first volcanic unit is the Talcott Basalt, followed by the Holyoke and Hampden basalts, interbedded with the cyclic lacustrine sequences of the Shuttle Meadow and East Berlin Formations and the Smith's Ferry and Park River members of the lower Portland Formation (Figs. 1 and 3). Interspersed throughout these lake sequences are more weakly developed paleosols, where periodic subaerial exposure of the primary sediments allowed soil-forming processes to operate on the lake margins (e.g., Tanner, 2003).

The predictive framework of Milankovitch cycle stratigraphy has been used to demonstrate that the flow units and interbedded sedimentary sequences of the Hartford basin are directly equivalent to those present in the Newark basin (Olsen, 1988; Whiteside et al., 2007) (Figs. 1 and 4). Therefore, the Shuttle Meadow Formation represents ~260 kyr, and the East Berlin Formation ~290 kyr. Recently, the magnetostratigraphy of the Hartford basin has extended the Newark timescale into the Early Jurassic (Kent and Olsen, 2008), through sampling of the largely lacustrine lower 2000 m of the thick post-extrusive Portland Formation, which is the primary focus of the current study.

### 3. Estimating pCO<sub>2</sub> from pedogenic carbonates

Pedogenic carbonate nodules were collected from paleosols located stratigraphically above and below each of the CAMP flows in the Hartford basin from both cores and outcrop. Samples from New Haven Formation, distributed 500 and 1200 m below the Talcott Basalt, provided a pre-CAMP baseline. Since the mostly fluvial New Haven lacks cycle or magnetic stratigraphy, these samples are treated as a single relative background value for comparison to the Newark basin record. The samples situated ~1200 m below the Talcott Basalt are from exposures dated using U–Pb at 211 ± 2.1 Ma by Wang et al. (1998), and we estimate the sample at 500 m below the basalt to be ~205 Ma. Samples from the lower Shuttle Meadow Formation are from a cored section near Berlin, CT (Silver Ridge Core B-1 (see Whiteside et al., 2011)) as well as from outcrop. The upper 75% of the East Berlin Formation is well exposed but suitable paleosols are rare. The lower 600 m of the post-extrusive Portland Formation is covered in high resolution with significant overlap by a series of short geotechnical cores taken by the Army Corps of Engineers (ACE) during construction of the Park River flood diversion tunnel project (Kent and Olsen, 2008; Pienkowski and Steinen, 1995). Samples from exposures in the middle to upper Portland Formation were assembled using the stratigraphy of Kent and Olsen (2008), and our uppermost sample is from near the top of the mostly lacustrine part of the Portland Formation (see Table 1 for all sample and core location data).

Organic and inorganic carbon isotope measurements from these paleosols were input into the soil diffusion model of Cerling (1999):

$$C_a = S(z) \frac{\delta_s - 1.0044\delta_\phi - 4.4}{\delta_a - \delta_s}$$

where  $C_a$  is the concentration of atmospheric CO<sub>2</sub>,  $S(z)$  is the concentration of CO<sub>2</sub> at soil depths greater than 50 cm contributed by the respiration of soil organic matter,  $\delta_s$  is the  $\delta^{13}\text{C}$  of soil CO<sub>2</sub>,  $\delta_\phi$  is the  $\delta^{13}\text{C}$  of soil-respired CO<sub>2</sub>, and  $\delta_a$  is the  $\delta^{13}\text{C}$  of atmospheric CO<sub>2</sub>. All  $\delta$  values are relative to Vienna PeeDee Belemnite (VPDB).

The carbon isotopic ratio of soil carbonate ( $\delta_{cc}$ ) is used as a proxy for  $\delta_s$ , which involves a temperature-dependent equilibrium fractionation between CaCO<sub>3</sub> and CO<sub>2</sub>, described by:

$$10^3 \ln \alpha = 11.709 - 0.116(T) + 2.16 \times 10^{-4}(T)^2$$

(Cerling, 1999) where  $\alpha$  is the fractionation factor, and temperature ( $T$ , in °C) is fixed at 25 °C, as appropriate for the tropical Newark basin. Because there is no carbon isotope fractionation due to respiration,  $\delta_\phi$  is related directly to the carbon isotopic ratio of soil organic matter ( $\delta^{13}\text{C}_{\text{org}}$ ). The carbon isotopic ratio of the atmosphere is calculated from the measured  $\delta^{13}\text{C}_{\text{org}}$  by the following relationship (Arens et al., 2000):

$$\delta_a = (\delta^{13}\text{C}_{\text{org}} + 18.67)/1.10$$

which assumes consistent fractionation by photosynthesis. This builds carbon cycle perturbations directly into the model. The only adjustable parameter of the model is the concentration of CO<sub>2</sub> in the soil derived from the respiration of organic matter ( $S(z)$ ), which is a function of soil productivity. The paleosols sampled in this study were relatively productive argillic and vertic calcisols (Mack et al., 1993), with mean depth to the Bk horizon ( $D_s$ ) at 40 ± 15 cm soil depth, which we use to estimate  $S(z)$  by the following empirical relationship:  $S(z) = 66.7D_s + 588$  (Retallack, 2009). This results in a mean  $S(z)$  of ~3256 with an SE of ± 893 ppm. Given the difficulty inherent in accurately estimating depth to the Bk horizon in individual paleosols from drill cores, and in accordance with the work of Breecker et al. (2009, 2010), we use an  $S(z)$  value of 3000 ± 1000 ppm for all pCO<sub>2</sub> estimates, which gives a final pCO<sub>2</sub> range that is indistinguishable from the error that would result from systematic use of the empirical depth to carbonate relationship. Individual soil profiles were not decompacted (Retallack, 2009), and our  $S(z)$  approximations are therefore conservatively low.

A combination of thin sections and polished slabs are used to identify primary micritic calcite from diagenetic material (Driese and Mora, 2002), with a sampling preference for small, isolated carbonate nodules that are generally in better isotopic equilibrium with the surrounding soil than larger nodules or calcretes (Schaller et al., 2011a). Great care was taken to avoid phreatic calcretes, and to sample only those displaying clear evidence of having formed in the vadose zone (Rasbury et al., 2006). Samples for organic carbon isotope analysis were taken from as close to the soil-surface as possible to avoid measuring recalcitrant organics unrelated to the active organic carbon pool during soil formation (Nadelhoffer and Fry, 1988), and several samples from each profile were homogenized to provide a more representative organic carbon value.

The  $\delta^{13}\text{C}$  value of pedogenic carbonate is known to decrease predictably with soil depth (Quade et al., 1989), and characterizing this trend makes it possible to differentiate between the relative influences of the atmospheric vs. soil-respired CO<sub>2</sub> reservoirs. Therefore, multiple (at least 5) down profile isotopic measurements were made on each paleosol to identify an equilibrium  $\delta_{cc}$  value with depth in the soil (Schaller et al., 2011a) (Fig. 2). Using the mean of these depth-controlled measurements ensures that the mixing between the atmospheric and soil-respired reservoirs is at equilibrium with respect to the diffusion model, removing significant uncertainty in the isotopic maturity of a given paleosol profile.

Chronostratigraphic placement follows the cycle and magnetic stratigraphy of Kent and Olsen (2008) and Olsen et al. (2005). In the Park River ACE core suite, this cyclostratigraphy provides direct stratigraphic age control to the level of orbital precession, and correlates unambiguously to the post-extrusive Boonton Formation of the Newark basin (Kent and Olsen, 2008; Olsen et al., 2005).

### 4. Results

The depth-equilibrated mean stable carbon isotopic composition of pedogenic carbonate ( $\delta^{13}\text{C}_{cc}$ ) from exposures 500 to 1200 m below the Talcott basalt in the New Haven Formation ranges from −7.7 to −7.2‰ (relative to VPDB), which are

**Table 1**  
Samples from both outcrop and core from the Hartford basin. Formations in bold are CAMP basalt units.

Sample	Core or outcrop		Formation	Depth – below/+ above Talcott Base (m)	Age (Ma) <sup>a</sup>	Down-profile mean $\delta^{13}\text{C}_{\text{carb}}$ (‰ PDB) <sup>b</sup>	$\delta^{13}\text{C}_{\text{OM}}$ (‰ PDB) <sup>c</sup>	Atmos. pCO <sub>2</sub> (ppm) (at Sz = 3000 ± 1000 ppm)	
	Lat	Lon							
HNH3-50	41.55649	–72.91159	New Haven	–1200 <sup>d</sup>	211.9 <sup>d</sup>	–7.4	–26.2	1775	±592
HNH2-47	41.55723	–72.91175	New Haven	–1200 <sup>d</sup>	211.9 <sup>d</sup>	–7.7	–26.4	1703	±568
HNH691B <sup>e</sup>	41.55956	–72.90775	New Haven	–1175 <sup>d</sup>	211.9 <sup>d</sup>	–7.2	–26.8	2218	±739
HNH691C <sup>e</sup>	41.55981	–72.90845	New Haven	–1170 <sup>d</sup>	211.9 <sup>d</sup>	–7.6	–26.9	2064	±688
HNHAAD <sup>e</sup>	41.39441	–72.8872	New Haven	–500 <sup>d</sup>	~205	–7.6	–27.0	2065	±688
			<b>Talcott Basalt</b>	0	201.38				
HSMSRB1-165	41.5850	–72.7565	Shuttle Meadow	120	201.26	–3.4	–26.2	5186	±1773
HSMAAA <sup>e</sup>	40.67295	–72.83597	Shuttle Meadow	200	201.18	–6.6	–26.0	3074	±1025
			<b>Holyoke Basalt</b>	241					
HEBFD26T-82	41.758639	–72.69509	East Berlin	540	200.84	–6.3	–26.2	2249	±746
			<b>Hampden Basalt</b>	550					
HPFD24T-112	41.758101	–72.693243	Portland	597	200.78	–4.6	–27.7	5473	±1824
HPFD24T-107	41.758101	–72.693243	Portland	599	200.78	–5.3	–26.1	2980	±993
HPFD24T-92	41.758101	–72.693243	Portland	603	200.77	–4.9	–26.2	3530	±1177
HPFD14T-82	41.758201	–72.690903	Portland	605	200.77	–3.0	–25.2	4391	±1464
HPFD20T-165	41.757929	–72.68815	Portland	631	200.75	–3.8	–25.2	4518	±1506
HPFD20T-99	41.757929	–72.68815	Portland	651	200.73	–3.6	–24.9	3574	±1191
HPFD16T-206	41.757913	–72.684209	Portland	671	200.71	–7.5	–27.8	2727	±909
HPFD16T-154	41.757913	–72.684209	Portland	686	200.69	–6.3	–27.2	3019	±1006
HPFD16T-126	41.757913	–72.684209	Portland	695	200.68	–7.1	–27.7	2811	±937
HPFD16T-48	41.757913	–72.684209	Portland	718	200.66	–4.8	–26.0	3455	±1152
HPFD13T-165	41.757635	–72.677278	Portland	726	200.65	–6.0	–26.7	2988	±996
HPFD22T-210	41.757761	–72.680457	Portland	744	200.63	–6.2	–27.1	3072	±1024
HPFD12T-210	41.757841	–72.675222	Portland	745	200.63	–5.8	–25.5	2317	±772
HPFD18T-138	41.758023	–72.682255	Portland	746	200.63	–4.9	–25.9	3280	±1093
HPFD12T-32	41.757841	–72.675222	Portland	799	200.58	–6.9	–26.6	2204	±735
HPFD19T-188	41.757686	–72.672524	Portland	801	200.58	–6.8	–27.0	2545	±848
HPFD23T-80	41.757715	–72.671683	Portland	847	200.53	–3.3	–25.5	4496	±1499
HPFD19T-32	41.757686	–72.672524	Portland	848	200.53	–4.6	–26.6	4125	±1375
HPFD30T-175	41.757341	–72.668001	Portland	913	200.46	–3.1	–25.7	5092	±1697
HPFD30T-86	41.757341	–72.668001	Portland	940	200.44	–5.9	–26.6	2882	±961
HPFD29T-185	41.757496	–72.664766	Portland	983	200.39	–5.6	–26.4	3003	±1001
HPFD29T-97	41.757496	–72.664766	Portland	1010	200.37	–7.3	–25.5	1496	±499
HPFD7T-101	41.758055	–72.664892	Portland	1025	200.35	–6.9	–26.4	2117	±706
HPFD27T-154	41.757519	–72.663522	Portland	1043	200.33	–5.7	–27.3	2649	±883
HPFD27T-94	41.757519	–72.663522	Portland	1062	200.31	–6.2	–26.6	2716	±905
HPEL <sup>e</sup>	41.5712	–72.6198	Portland	1143	200.23	–7.7	–26.0	1528	±509
HPEJ <sup>e</sup>	41.5723	–72.6163	Portland	1224	200.15	–9.3	–26.0	915	±305
HPJFA <sup>e</sup>	41.9747	–72.6598	Portland	1582	199.79	–9.5	–25.8	785	±262
HPJPEG <sup>e</sup>	41.9903	–72.5937	Portland	2379	198.99	–7.7	–26.6	1768	±589

<sup>a</sup> Age based on cycle and magnetic stratigraphy tied to 201.38 absolute ages of first extrusive unit (Schoene et al., 2010).

<sup>b</sup> Analyzed on a Micromass Optima – dual inlet IRMS.

<sup>c</sup> Analyzed on a Eurovector EA connected to VG Isoprime IRMS.

<sup>d</sup> Depth estimate ± 200 m error, based on projection of the bedding dip. Age estimates from U/Pb date of 211.9 ± 2.1 Ma (Wang et al., 1998) on same/adjacent exposure.

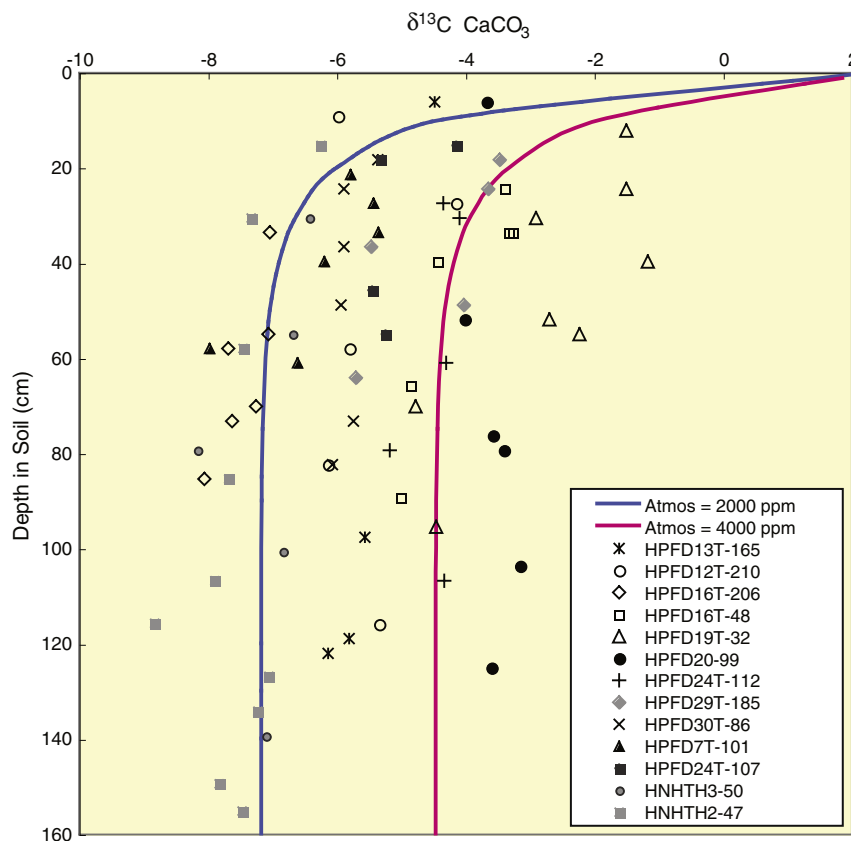
<sup>e</sup> Samples from outcrop.

comparable to values from the Passaic Formation of the Newark basin (see Table 1). These values are very close to those of Suchecki et al. (1988) from the New Haven Formation, which help to establish a consistent average pre-eruptive background. Stratigraphically above the Talcott Basalt, in the lower Shuttle Meadow Formation, pedogenic carbonates have  $\delta^{13}\text{C}$  values of –3.2‰, which decrease to –6.6‰ toward the top of the formation (Fig. 3). Comparable values of –6.3‰ are found at the top of the East Berlin Formation, which sits stratigraphically above the Holyoke Basalt (the second flow unit). The  $\delta^{13}\text{C}$  of soil organic matter was between –26.3‰ and –26.0‰ in the Shuttle Meadow and upper East Berlin formations. These values are consistent with results from the Feltville and Towaco Formations of the Newark basin (Schaller et al., 2011a), and the high  $\delta^{13}\text{C}$  of pedogenic carbonate above the Talcott Basalt (equivalent of the Newark Orange Mountain Basalt) probably represents the increased influence of the atmospheric CO<sub>2</sub> reservoir on pedogenic carbonate formed at depth.

Calculated pCO<sub>2</sub> levels through the New Haven Formation are treated as a single value, averaging 1900 ± 650 ppm. Those samples from exposures roughly 1200 m below the Talcott basalt have a U–Pb age of 211.9 ± 2.1 Ma (Table 1) (Wang et al., 1998), and calculated pCO<sub>2</sub> values of ~1800 to 2200 ± 700 ppm

(Table 1; all pCO<sub>2</sub> estimates at  $S(z) = 3000 \pm 1000$  ppm), which are comparable to the ~2000 ± 700 ppm of Schaller et al. (2011a) from the pre-CAMP Passaic Formation (204 to 201.5 Ma) in the Newark basin. These samples from the lower New Haven indicate that a pre-eruptive Triassic pCO<sub>2</sub> baseline may have persisted for at least ~10 Myr before the initial CAMP eruptions, a significant extension of the available Newark record.

Just above the Talcott Basalt in the Shuttle Meadow Formation, pCO<sub>2</sub> increases to ~5000 ± 1500 ppm followed by a decrease to ~3000 ± 1000 ppm just beneath the Holyoke Basalt. These pCO<sub>2</sub> levels are comparable to those observed in the first post-eruptive strata of the Feltville Formation in the Newark basin, at ~4400 ± 1400 ppm directly on top of the Orange Mountain Basalt (Fig. 4). Atmospheric pCO<sub>2</sub> at the top of the East Berlin Formation, just below the Hampden Basalt, is roughly 2200 ± 750 ppm, which is consistent with the ~2000 ppm background values found at the top of the Towaco Formation of the Newark basin (Fig. 4). Although the resolution of these few inter-CAMP points in the Hartford basin is sporadic, their consistency with the record from the Newark strata (Fig. 4) reinforces the independence of individual values and the global applicability of this approach.



**Fig. 2.** Stable carbon isotopic composition of pedogenic carbonate ( $\delta_{cc}$ ) with estimated soil depth from representative paleosols in the Portland Formation (symbols encompass analytical error). Note that these shallowest nodules represent depths to the uppermost carbonate nodules, which should not be confused with depth to the Bk horizon (Retallack, 2009). Observed  $\delta_{cc}$  is compared to the  $\delta_{cc}$  values predicted by the diffusion model at atmospheric  $\text{CO}_2$  concentrations of 2000 ppm (blue line) and 4000 ppm (pink line) (after Quade et al., 1989). For this demonstration only, atmospheric  $\delta^{13}\text{C}_{\text{CO}_2}$  was set to  $-6.5\text{‰}$ , soil  $\delta^{13}\text{C}_{\text{org}}$  was set to  $-26.5$ , with an exponential production function and characteristic depth of production at 15 cm (other parameters described in text). For all  $p\text{CO}_2$  estimates made in this study, the  $\delta^{13}\text{C}_{\text{org}}$  was measured directly and used as a model input. Soil carbonate above 20 cm in the profile was rare; note that stabilization of measured  $\delta_{cc}$  is commonly well below 50 cm soil depth. Only the mean of the equilibrium  $\delta_{cc}$  values were used to calculate  $p\text{CO}_2$  in this study. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

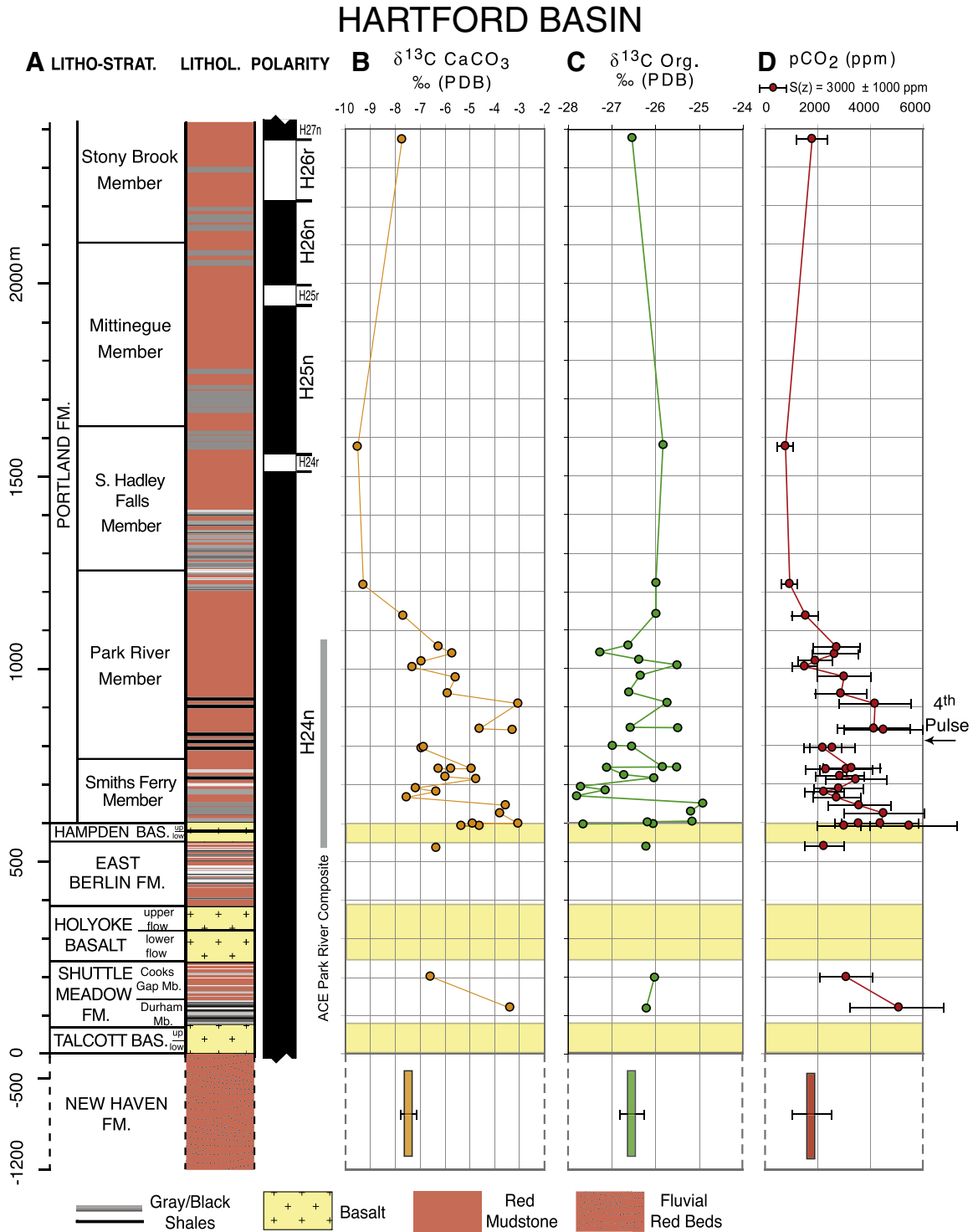
In the post-extrusive Portland Formation, pedogenic carbonates have  $\delta^{13}\text{C}$  values of  $-3$  to  $-5\text{‰}$  just above the Hampden Basalt, decreasing to  $-7\text{‰}$  about 200 m up section (Fig. 3). Around 240 m above the Hampden Basalt, the  $\delta^{13}\text{C}_{cc}$  again increases to  $-3.5\text{‰}$ , followed by a falloff through the rest of the sampled Portland Formation to a minimum of  $-9.5\text{‰}$  ~2400 m (1.8 Myr) above the Hampden Basalt. The  $\delta^{13}\text{C}$  of soil organic matter ( $\delta^{13}\text{C}_{\text{OM}}$ ) also shows variability through the Portland Formation, ranging from  $-24.9\text{‰}$  to  $-27.8\text{‰}$  (Fig. 3). Similar to the Newark record,  $\delta^{13}\text{C}_{\text{OM}}$  decreases slightly just above the Hampden Basalt, followed by an immediate swing toward higher values. Overall, the lower Portland Formation exhibits  $\sim 3\text{‰}$  variability in  $\delta^{13}\text{C}_{\text{OM}}$  that stabilizes up section.

Calculated atmospheric  $p\text{CO}_2$  in the Portland Formation shows a few distinct trends. In soils formed directly on top of the Hampden Basalt,  $p\text{CO}_2$  levels are  $\sim 5400 \pm 1500$  ppm, with some fluctuation in the calculated  $p\text{CO}_2$  level before settling to  $\sim 4300 \pm 1200$  ppm (Fig. 3). This apparent fluctuation, observed in a single core above the Hampden Basalt, is driven by the  $\delta^{13}\text{C}$  of soil organic matter, because the  $\delta^{13}\text{C}$  of pedogenic carbonate is relatively stable in the first 10 m above the basalt. Because these soils are relatively weakly developed and organic matter was rare, the few apparently lower  $p\text{CO}_2$  values are likely a product of  $^{13}\text{C}$  enriched recalcitrant organic matter that is unrelated to the active decomposition pool during pedogenesis (Nadelhoffer and Fry, 1988). Aside from these few points, the majority of the calculated  $p\text{CO}_2$  changes in the Hartford basin appear to be driven by the  $\delta^{13}\text{C}$  of pedogenic carbonate.

Above the initial increase, atmospheric  $p\text{CO}_2$  gradually decreases to near background levels in the first 200 m of the post-extrusive

Portland Formation. At about 240 m above the Hampden Basalt,  $p\text{CO}_2$  again peaks to  $\sim 4500 \pm 1200$  ppm, which is captured laterally across 2 individual cores, followed by a steady decrease to below background levels through the rest of the 1700 m of sampled section (Fig. 3).

Because the Park River Member is equivalent to a part of the Boonton Formation of the Newark basin, the  $p\text{CO}_2$  values found in the Portland are expected to be consistent with the Boonton record, where an increase to  $\sim 5000$  ppm is noted just above the Hook Mountain Basalt (Fig. 4), and where the next few samples some 350 m up section yield  $p\text{CO}_2$  levels of  $\sim 3000$ – $2500$  ppm. These values from the upper Boonton Formation were previously thought to be anomalously high, but are in fact directly consistent with similar high concentrations following the  $p\text{CO}_2$  peak at 240 m in the Portland Formation. It appears that the full expression of the final observed  $p\text{CO}_2$  pulse of the Portland (the fourth of the Newark–Hartford section) was simply not captured by the low resolution record from the Boonton Formation due to lack of appropriate lithology, and yet the few points present in the upper Newark match their chronostratigraphic equivalents in the Hartford. The  $p\text{CO}_2$  estimates from the Portland Formation show excellent reproducibility across section between overlapping individual cores. Overall, the Hartford basin  $p\text{CO}_2$  record is tightly consistent with that of the Newark basin ( $\sim 250$  km to the south), despite independent age control used for correlation (error within a few precession cycles between basins) and our use of parameterized  $S(z)$  values of  $3000 \pm 1000$  ppm, and temperatures fixed at  $25^\circ\text{C}$  in the records generated from both basins (Figs. 4 and 6).

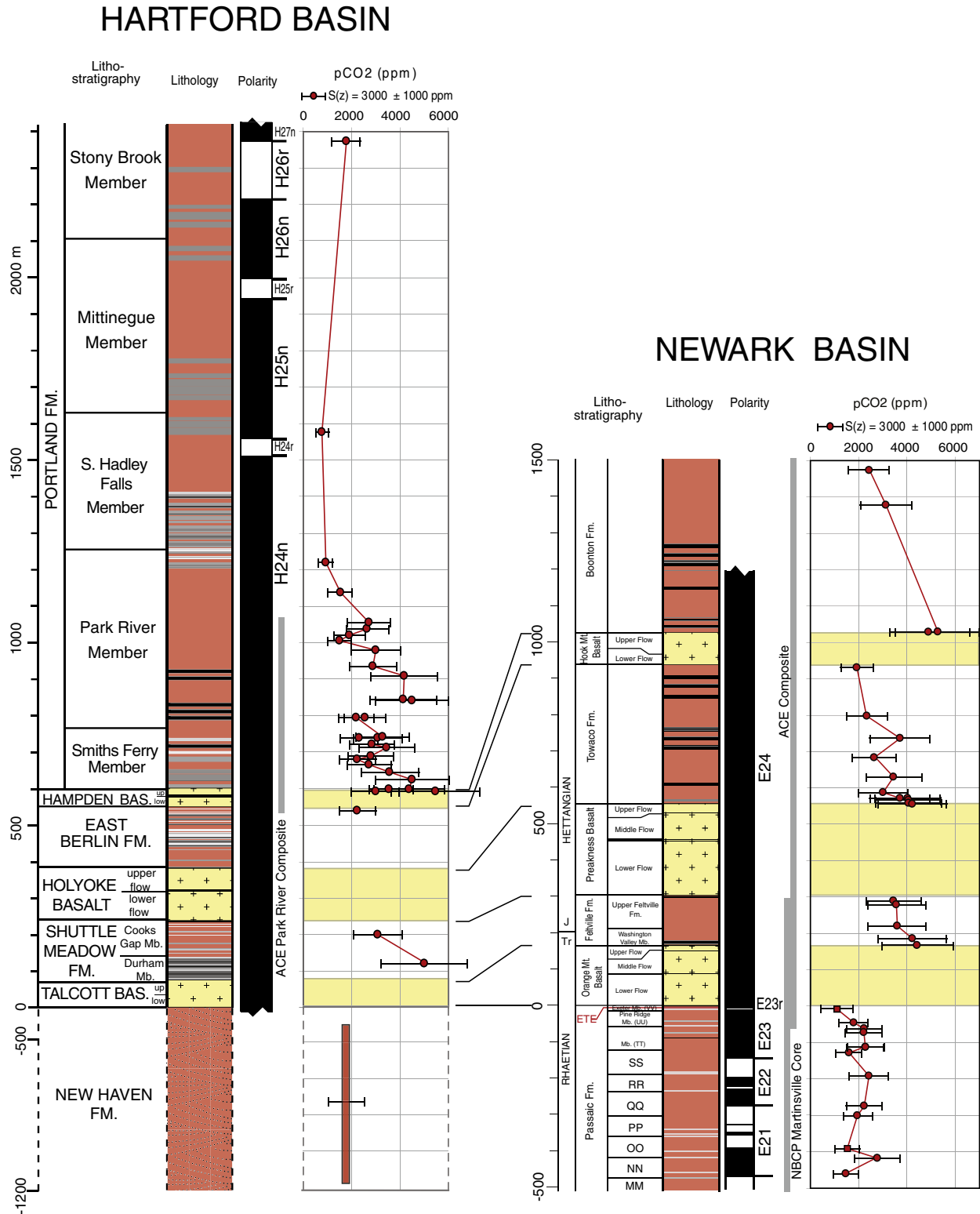


**Fig. 3.** A. Lithologic and polarity stratigraphy of the Hartford basin from Kent and Olsen (2008). B. Profile equilibrated mean  $\delta^{13}\text{C}$  values of pedogenic carbonate. C.  $\delta^{13}\text{C}$  of organic matter from as close to the paleosol surface as possible. D. Calculated  $p\text{CO}_2$  based on input variables in B and C at 25 °C. The concentration of respired  $\text{CO}_2$  in the soil  $[S(z)]$  was estimated at  $3000 \pm 1000$  ppm, and the resulting range of  $p\text{CO}_2$  is indicated by the  $\pm$  error bars on each estimate. For B, C, and D, only the mean  $\pm$  standard deviation of the 5 sample locations from the New Haven Formation is shown because of stratigraphic uncertainty; see Table 1 for depths below the Talcott Basalt.

#### 5. A fourth major pulse of volcanism?

With one exception, each pulse of  $\text{CO}_2$  observed in the Newark and Hartford records is found only directly above a unit of CAMP lavas, and each observed  $p\text{CO}_2$  increase has accordingly been attributed to the volcanic activity responsible for the emplacement of the underlying extrusive unit (Schaller et al., 2011a). The observed increase in  $p\text{CO}_2$  ~240 m from the base of the Portland Formation, or ~240 kyr after the

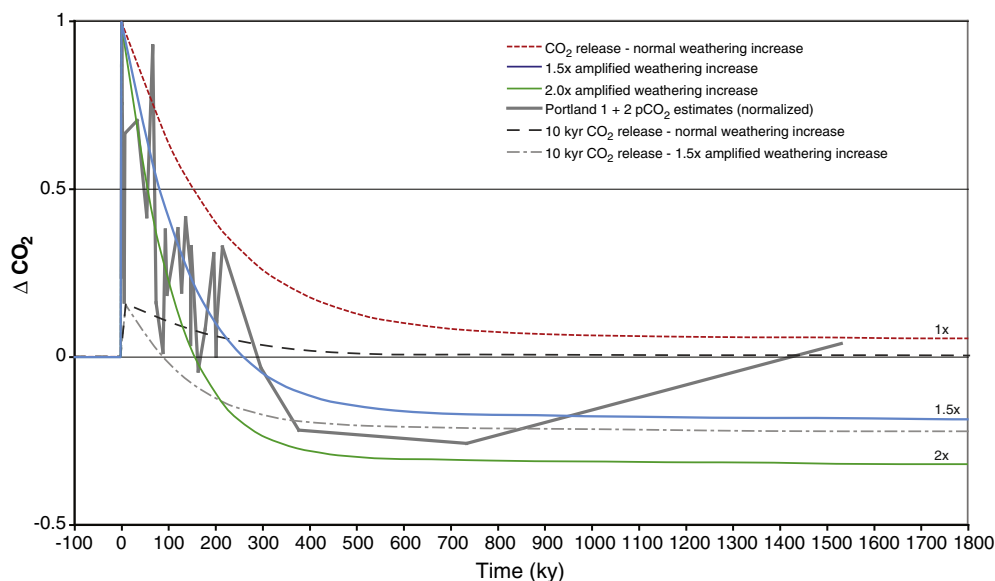
extrusion of the Hampden Basalt, is of roughly the same size and duration as those in the Newark and Hartford record that are directly preceded by laterally extensive extrusive units (Fig. 5). Also, the  $\delta^{13}\text{C}$  of organic matter shows little indication of another (perhaps lighter) source of carbon feeding this  $\text{CO}_2$  pulse (e.g., Svensen et al., 2004). Therefore we attribute this fourth rise to a fourth episode of substantial and rapid volcanism, similar in magnitude to those episodes corresponding to the observed lava flow units in the Newark and Hartford basins.



**Fig. 4.** Calculated  $pCO_2$  from the Hartford basin compared to the Newark basin  $pCO_2$  record of Schaller et al. (2011a). Error bars represent calculated  $pCO_2$  at soil respired  $CO_2$  concentrations  $[S(z)]$  of  $3000 \pm 1000$  ppm. The Army Corps of Engineers (ACE) suite of Park River cores is denoted by gray bar in the Hartford basin, covering the lower Portland Formation in high resolution. For comparison to the pre-CAMP background of Schaller et al. (2011a), samples from the New Haven Formation are shown as a mean of the  $pCO_2$  estimates between  $\sim 500$  m and  $\sim 1200$  m below the Talcott Basalt (see Table 1), with an error corresponding to  $S(z)$  values of  $3000 \pm 1000$  ppm.

However, there is no solid evidence in Eastern North America for a fourth major pulse of CAMP activity (see Olsen et al., 2011 for review; Weems and Olsen, 1997). A thin unnamed basalt caps the Culpeper basin sequence about  $\sim 1300$  m above the Sanders Basalt. However, there is structural ambiguity (Lee and

Froelich, 1989), and the unnamed basalt may in fact be a faulted portion of the underlying Sanders Basalt (Weems and Olsen, 1997), making it an unlikely candidate for the volcanic episode that produced the fourth  $pCO_2$  increase observed in the Hartford basin.



**Fig. 5.** The  $p\text{CO}_2$  record of the post-extrusive Portland Formation (at  $S(z) = 3000$  ppm) (gray line), normalized to their respective increases above background, combined with one another, and scaled to an initial eruptive pulse at  $t = 0$ , for comparison to the results of geochemical modeling using a modified version of the COPSE model (Bergman et al., 2004). The model equilibrates the atmosphere and ocean on the 1000-year timescale. In the reference run (red dotted-line), the model achieves steady state for 100 kyr, at which point atmospheric  $\text{CO}_2$  was doubled over 1000 years (by adding  $3.5 \times 10^{17}$  mol of  $\text{CO}_2$ ) to simulate the eruption of a single pulse of CAMP volcanics and their  $\text{CO}_2$  release. The ensuing decrease in  $p\text{CO}_2$  is due to consumption by increased continental weathering rate resulting from the high  $p\text{CO}_2$  and resultant increased temperatures. In the test simulations, the ocean-atmosphere  $\text{CO}_2$  reservoir was again doubled in size, while the increased rate of silicate weathering was concordantly further amplified by 1.5 (blue line) and 2 times (green line), in an attempt to simulate the effects of freshly erupted basalts on the continental weathering budget. A second set of test simulations shows the effect of the same  $\text{CO}_2$  pulse released over 10 kyr without an amplification of the weathering increase (black dashed line), and with a  $1.5 \times$  amplification (gray dot-dash line). The model was run for a total of 1.9 Myr. Linear correlation of each 1000-year release simulation to the observed normalized Portland  $p\text{CO}_2$  estimates at  $S(z) = 3000$  ppm (using least-squares) gives  $R^2 = -0.03$  for the reference simulation,  $R^2 = 0.62$  for the 1.5 time amplification of the weathering increase, and  $R^2 = 0.46$  for the 2 time amplification. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Similarly, four distinct flow units have been recognized in the corollary High Atlas region of Morocco, deemed the Lower, Intermediate and Upper Units, and the Recurrent Basalt, on the basis of major element geochemistry (Bertrand, 1991). Although there is debate regarding the detailed stratigraphic placement of the Lower Basalt (Deenen et al., 2010; Marzoli et al., 2004; Whiteside et al., 2007), the Recurrent is geochemically similar to Hook Mountain and Hampden Basalts (Deenen et al., 2010; Marzoli et al., 2011), leaving the volcanism responsible for the Recurrent a possible, but still unlikely tracer for the fourth  $p\text{CO}_2$  increase observed in the Hartford basin. The South American CAMP extrusive section remains largely unstudied stratigraphically, and it is unclear how these volcanics are related to the ENA section. Despite CAMP dikes, sills and rarer lava flows preserved over several million square kilometers of northern South America (see Marzoli et al., 1999 for review), it is at present impossible to determine the precise temporal or stratigraphic relationship of these volcanic units at the level necessary to isolate a single widespread eruptive event as the cause for the fourth  $p\text{CO}_2$  pulse observed in the Hartford basin. In addition, the age(s) of the Clubhouse Crossroads Basalt and their possible relationship to the voluminous seaward-dipping reflectors offshore is unclear (Olsen et al., 2003), and hence some of this igneous activity cannot be readily dismissed as a potential source for the fourth  $p\text{CO}_2$  pulse.

Most importantly, as evidenced by the fourth  $p\text{CO}_2$  peak in the Hartford basin, the CAMP-induced  $p\text{CO}_2$  increases should be identifiable in any section with the appropriate lithology and high sedimentation rate, regardless of the presence of CAMP lavas. Identifying these four  $p\text{CO}_2$  pulses in other basins is an essential step toward understanding the stratigraphy of the extrusive zone, and potentially adds a higher degree of precision to global correlation. In that light, the identification of a fourth apparent volcanogenic pulse in the Hartford basin allows us to revise the total duration of CAMP induced high  $p\text{CO}_2$  as observed in the ENA basins from  $600 \pm 20$  kyr (Olsen et al., 2003; Whiteside et al., 2007), to  $840 \pm 60$  kyr, based on the amount

of time present between the base of the Talcott Basalt and the last apparent pulse of  $\text{CO}_2$  in the Portland Formation using the chronostratigraphy of Kent and Olsen (2008).

## 6. Weathering of the CAMP basalts

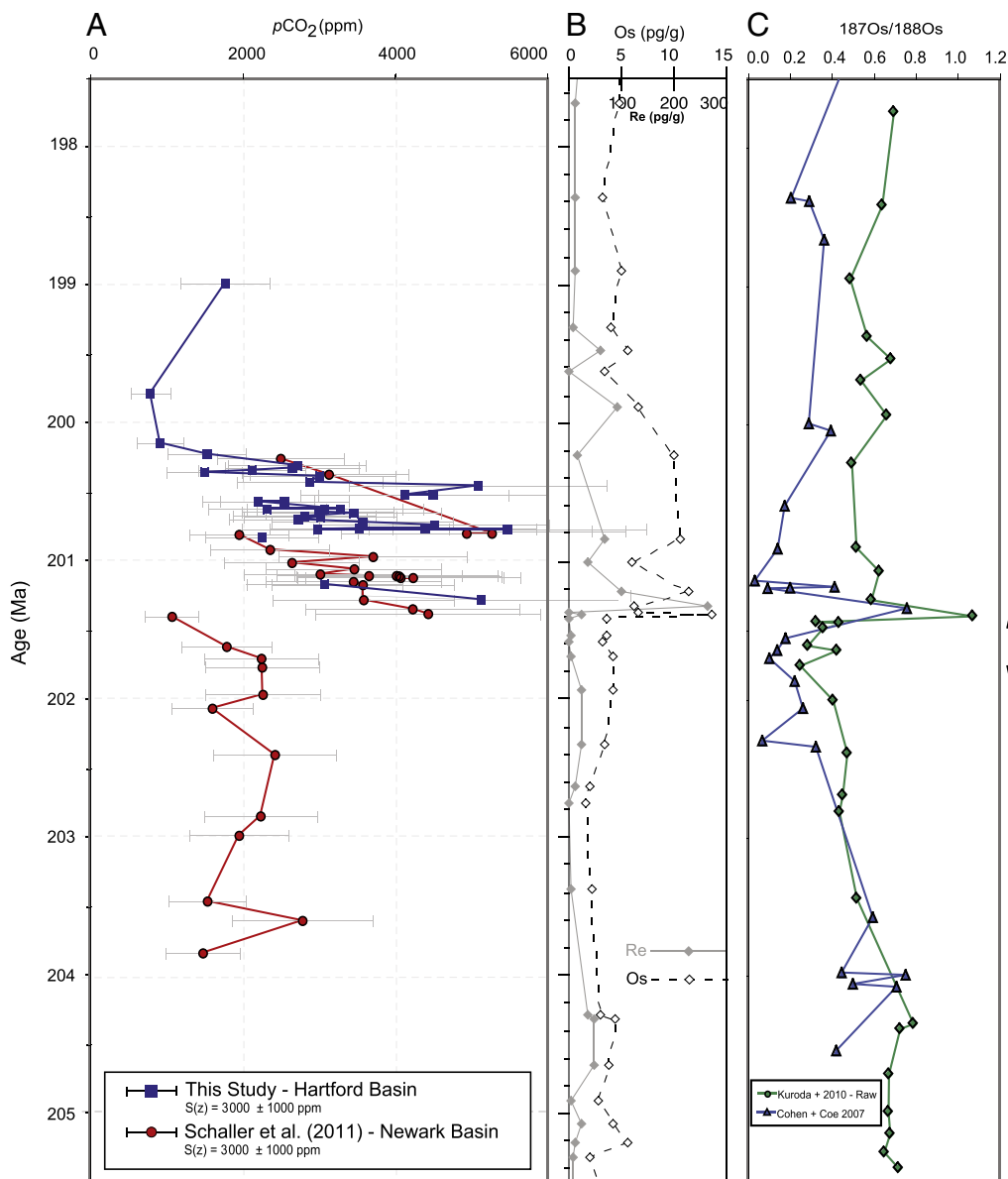
A notable characteristic of the post-eruptive CAMP record is the striking decrease in atmospheric  $p\text{CO}_2$  within a few hundred thousand years of basalt emplacement (Fig. 6). In the Newark record  $p\text{CO}_2$  falls from an excess of  $\sim 4000$  ppm to nearly background levels of  $\sim 2000$  ppm within  $\sim 300$  kyr of the extrusives. Similarly, in the post-extrusive Portland Formation of the Hartford basin, we observe a decrease over a comparable timescale (following both the Hampden Basalt and the fourth  $p\text{CO}_2$  increases discussed above). Unlike the others,  $p\text{CO}_2$  continues to decline following the final pulse in the Portland Formation, and ultimately levels off at concentrations below pre-eruptive background. We hypothesize that both the rapidity of the  $p\text{CO}_2$  decreases, and the fall in  $p\text{CO}_2$  to below background are due to the rapid consumption of  $\text{CO}_2$  by an overall increase in continental weathering, which is accelerated by the relatively rapid hydrolysis of the freshly erupted CAMP silicates themselves.

### 6.1. Modeling the post-extrusive $p\text{CO}_2$ decrease

Here, we use a geochemical model to demonstrate that the rapid decrease in atmospheric  $p\text{CO}_2$  following the CAMP eruptions may indeed be due to basalt-induced amplification of the predicted increase in continental weathering. We use a modified version of the COPSE biogeochemical model (Bergman et al., 2004) in perturbation mode, which couples the carbon, oxygen, phosphorus and sulfur cycles, with explicit feedbacks between atmospheric  $p\text{CO}_2$  and temperature to the rate of continental silicate weathering.

In the case of the Orange Mountain Basalt in the Newark basin (the oldest flow unit), the duration of eruption is well constrained





**Fig. 6.** A. Composite Newark and Hartford pCO<sub>2</sub> record of this study. Newark data are from Schaller et al. (2011a). B. The [Os] and [Re] records from the Kurusu section in Japan, plotted using the accumulation rates of Kuroda (2010), with the record correlated using the peak in <sup>187</sup>Os/<sup>188</sup>Os to the corresponding <sup>187</sup>Os/<sup>188</sup>Os peak in the isotope record of Cohen and Coe (2007). (note difference in Re and Os scales). C. <sup>187</sup>Os/<sup>188</sup>Os ratios from the Kurusu section in Japan (Kuroda et al., 2010) and the St. Audrey's Bay, U.K. (Cohen and Coe, 2007). Here we have used the peak in <sup>187</sup>Os/<sup>188</sup>Os in St. Audrey's Bay (Cohen and Coe, 2007) as a marker for a more appropriate correlation to the Kurusu section (contra Kuroda et al., 2010), which is afforded by the error in biostratigraphy and the residence time of Osmium in the oceans (Oxburgh, 2001). Placement offset noted by arrow.

to within a single precession cycle (Olsen et al., 2003; Whiteside et al., 2007), and a doubling of pCO<sub>2</sub> over pre-eruptive background is observed directly on top of the uppermost lava flow of the Orange Mountain. Each volcanic episode shows roughly the same pCO<sub>2</sub> response, with a similar durational constraint (with the exception of the Preakness and equivalent Holyoke basalts), and we are not aware of any data, geochemical or otherwise, that precludes an eruption time as short as 1000 years (1 kyr) for a single extrusive unit (e.g., see Schaller et al., 2011b). The absence of significant sedimentary strata or deep weathering profiles between individual lava flows of a single extrusive unit (e.g., within the Hampden Basalt) favors a short amount of time between lava flows, especially considering the high long-term sediment accumulation rates of ~1 meter per thousand years (Kent and Olsen, 2008). Therefore, we use a total eruptive duration of 1000 years for a single extrusive unit in our geochemical modeling, and for reference, show the results of a 10,000-year (10 kyr) degassing scenario with an identical release.

The perturbation mode of the modified COPSE model starts from a 100 kyr steady state where mid-ocean ridge degassing is balanced by silicate weathering as a function of pCO<sub>2</sub> and temperature at initial conditions. The model couples the surface ocean-atmospheric C-reservoir on a 1000-yr time-scale, and runs at 100-yr timesteps. In our simulations, we roughly double the size of the atmospheric fraction of the coupled ocean-atmosphere reservoir by adding 3.5 × 10<sup>17</sup> mol of CO<sub>2</sub> over 1000 years to reproduce the observed doubling of atmospheric CO<sub>2</sub> immediately following the CAMP eruptions, which is roughly one third of the total CO<sub>2</sub> potentially degassed from a total estimated CAMP basalt volume of the ~3 × 10<sup>6</sup> km<sup>3</sup> (Schaller et al., 2011a; Schaller et al., 2011b; Self et al., 2006). We run the model for a total of 1900 kyr, and in the reference run simply watch the weathering response of the system to this doubling over the ensuing 1800 kyr of model time. In an attempt to simulate the effect of an increase in the weathering potential of the

continental surface, we further amplify the increased rate of continental silicate weathering at the time of CO<sub>2</sub> perturbation by 1.5 and 2 times the normal effect.

## 6.2. Model results

The reference simulation shows the prescribed doubling of pCO<sub>2</sub> after the 100 kyr steady state, followed by an asymptotic drawdown to relatively stable pCO<sub>2</sub> levels that are slightly higher than the pre-perturbation steady-state by 600 kyr after the release (Fig. 5, dashed red line). This suggests that the silicate weathering system is capable of removing the majority of a doubling perturbation within ~600 kyr, but the increase in weathering due to the rapid transient increase in pCO<sub>2</sub> (and greenhouse temperature) is not enough to lower pCO<sub>2</sub> levels below pre-eruptive background, as observed in the post-CAMP record from the Hartford basin.

A 1.5 times amplification of the increased weathering rate at the time of CO<sub>2</sub> perturbation returns the system to pre-eruptive background pCO<sub>2</sub> levels by ~270 kyr, and eventually falls to about 20% below steady-state background (Fig. 5, blue line). A 2 times amplification returns pCO<sub>2</sub> to background levels by 180 kyr post-perturbation, eventually falling to well below pre-eruptive background (~30%) for the remaining 1500 kyr of simulation (Fig. 5, green line). The 10 kyr reference simulations (Fig. 5, black and gray dashed lines) both show roughly 12% of the initial atmospheric pCO<sub>2</sub> response as the 1 kyr runs using the same  $3.5 \times 10^{17}$  mol release.

The pCO<sub>2</sub> decreases to below pre-eruptive background in the Portland Formation cannot be a function of a normal increase in weathering due to a CO<sub>2</sub> perturbation (as in the reference run), indicating that some amplification of the weathering increase is required. However, a 2 times amplification (above the background increase due to increased temperature and CO<sub>2</sub> alone) reduces pCO<sub>2</sub> to levels further below background than observed in the Portland Formation. Therefore, we suggest that a ca. 1.5 times amplification of the silicate weathering increase adequately describes the two post-eruptive decreases observed in the Portland. This demonstrates that the highly weatherable basalts erupted into the equatorial humid belt must be partially responsible for the rapidity of the pCO<sub>2</sub> drawdown, and the decrease to below pre-eruptive background, by amplifying the increased in global weathering rates by perhaps 1.5 times. It should be noted that the timing and duration of the pCO<sub>2</sub> fluctuations are well-constrained parameters of the Newark–Hartford pCO<sub>2</sub> record, due to the precision of the astrochronology on these strata (Kent and Olsen, 1999, 2008; Olsen and Kent, 1996; Olsen et al., 1996a; Olsen et al., 2003). Therefore, the rate of drawdown is entirely dependent on calculated pCO<sub>2</sub> values and not an artifact of poor age control.

Since the difference between the test simulations and the reference run is essentially a function of basalt area available for hydrolysis, we can estimate the proportion of basalt necessary to further amplify the rate of continental weathering by a factor of 1.5. Assuming that a continental flood basalt may weather at a rate 10 times that of granitic continental crust (Dessert et al., 2003; Gaillardet et al., 1999), increasing the weathering potential of the total continental surface by a factor of 1.5 may be achieved by erupting enough basalt to cover ~1/18th the continental area, or  $\sim 8.3 \times 10^6$  km<sup>2</sup> (assuming total land area of roughly  $1.5 \times 10^8$  km<sup>2</sup>). This figure is comparable to estimates of the CAMP eruptive area at  $1.12 \times 10^7$  km<sup>2</sup> (McHone et al., 2003). Additionally, it appears that the rate of post-extrusive drawdown is faster in the later part of the composite Newark–Hartford record than earlier in the eruptive history (see Fig. 6). This may be due to the continuously increasing amount of basalt available to participate in weathering reactions with increasing time from the initial eruptions.

## 6.3. Comparison to the Deccan

Modeling experiments similar to those performed here have been used to evaluate the effects of the Deccan LIP (Caldeira and Rampino, 1990; Dessert et al., 2001), which lacks direct estimates of pCO<sub>2</sub>. In their most aggressive eruptive scenario, Caldeira and Rampino (1990) release  $2.0 \times 10^{17}$  mol CO<sub>2</sub> over an eruptive duration of 100 kyr, resulting in a meager ~65 ppm increase in pCO<sub>2</sub>. This CO<sub>2</sub> release is close to that used in the present study, but Caldeira and Rampino (1990) assume a much longer eruptive duration, which results in a highly attenuated effect (e.g., see Schaller et al., 2011b). They find that equilibrium is reestablished ~1.5 Myr after the eruption of the Deccan Traps; however, their model incorporates only a modest increase in continental weathering at the time of perturbation, and does not include an amplification to account for the Deccan basalts, so pCO<sub>2</sub> does not fall below pre-eruptive background.

In contrast, Dessert et al. (2001) use a much larger single CO<sub>2</sub> release ( $1.6 \times 10^{18}$  mol CO<sub>2</sub> over 100 kyr), which amounts to a ~1800 ppm increase, or a little over a doubling of atmospheric pCO<sub>2</sub>, which is similar to the increase observed in response to the CAMP. More importantly, the presence of the Deccan basalts themselves resulted in the return of pCO<sub>2</sub> to pre-eruptive levels by 1.2 Myr after perturbation, followed by a continued decline to equilibrium at levels 20% lower than pre-eruptive concentrations. They conclude that the increased continental weathering potential afforded by the presence of the Deccan lava pile was essential in rapidly reducing atmospheric pCO<sub>2</sub> below steady state background.

The results of Dessert et al. (2001) for the Deccan are consistent with the findings of this study of the CAMP, but have a significantly longer relaxation time: pCO<sub>2</sub> does not return to pre-eruptive background until ~1.2 Myr after perturbation in the Dessert et al. (2001) simulation, compared to the ~300 kyr return observed in the CAMP record. Several factors probably account for this difference. First, the Dessert et al. (2001) simulation releases a large quantity of CO<sub>2</sub> over a relatively long eruptive period (100 kyr) to induce a ~doubling of atmospheric pCO<sub>2</sub>, which may take a significantly longer amount of time to remove completely. A much smaller total amount of CO<sub>2</sub> can be released over a much shorter duration (e.g.,  $3.5 \times 10^{17}$  released over 1000 years, as in our simulations) and result in roughly the same doubling of atmospheric pCO<sub>2</sub>. Thus, on timescales greater than 1000 years, or the nominal overturning time of the oceans (Broecker and Peng, 1982), nearly 10 times more CO<sub>2</sub> is needed to produce the same atmospheric effect, which may require additional CO<sub>2</sub> sources (e.g., see Schaller et al., 2011b; Rampino and Caldeira, 2011). This is apparent in our simulations (Fig. 5) where  $3.5 \times 10^{17}$  mol of CO<sub>2</sub> are released on both 10 kyr and 1 kyr timescales; the 10 kyr release resulted in ~12% of the initial atmospheric pCO<sub>2</sub> increase as that of the 1 kyr scenario. Secondly, a more minor contributing factor is that the weatherable area of the Deccan Traps was estimated to be  $\sim 2 \times 10^6$  km<sup>2</sup>, which is considerably smaller than the total estimated area of the CAMP eruptions ( $\sim 1.12 \times 10^7$  km<sup>2</sup>), and hence has a more limited potential to absorb CO<sub>2</sub>. Thirdly, CAMP straddled the paleoequator where weathering may have been more intense than that of the Deccan, which was emplaced in the tropical arid belt (~26°S latitude) although it was subject to more intense weathering much later when India drifted into the equatorial humid belt (Kent and Muttoni, 2008).

Parenthetically, the rate of CO<sub>2</sub> release will also affect the degree of ocean acidification. Hautmann (2004) hypothesized that drastic increases in pCO<sub>2</sub> around the Triassic–Jurassic transition may have caused rapid ocean acidification, and hence undersaturation of seawater with respect to CaCO<sub>3</sub>, based on a global carbonate gap in the stratigraphic record. Berner and Beerling (2007) use a geochemical model to test this hypothesis, and conclude that a minimum release of  $8.3 \times 10^{17}$  mol CO<sub>2</sub> (10,000 GtC) in less than 100 kyr is necessary to induce wide-spread CaCO<sub>3</sub> dissolution. However, a degassing scenario where  $\sim 3 \times 10^{17}$  mol CO<sub>2</sub> is released in ~1 kyr, as we suggest

is recorded in the Newark Group where four such pulses of roughly  $\sim 3.5 \times 10^{17}$  mol CO<sub>2</sub> each are observed, is probably sufficient to produce a CaCO<sub>3</sub> undersaturated ocean.

#### 6.4. Other indicators of increased pCO<sub>2</sub> and global weathering rates

Our pCO<sub>2</sub> estimates are corroborated by other independent reconstructions at lower resolutions. In particular, stomatal densities have shown a doubling to tripling of pCO<sub>2</sub> across the ETE, followed by a 600 kyr plateau at elevated concentrations (McElwain et al., 1999). Despite showing the same relative change as the Newark Group paleosol estimates, the absolute values of the stomata-based approximations are appreciably lower and show little variability within the CAMP interval. This apparent lack of resolution within the CAMP interval is probably because both the stomatal index and density proxies have an asymptotic response to increasing pCO<sub>2</sub> (Beerling and Royer, 2002b), and therefore become insensitive at extremely high concentrations, showing little further change as saturation is approached. The offset between the paleosol estimates and the leaf stomata record may also be due to calibration issues, where the stomatal density proxy may underestimate pCO<sub>2</sub> (Beerling and Royer, 2002a).

Pedogenic carbonate pCO<sub>2</sub> reconstructions from the western U.S. (Cleveland et al., 2008) are comparable to the pre-eruptive background levels found in the New Haven and Passaic formations of this study. The Ghost Ranch and Montoya sections of Cleveland et al. (2008) lack CAMP volcanics and instead rely on a chronology derived from fluvial sequence stratigraphy (Cleveland et al., 2007). Without evidence that these sections extend into the Early Jurassic, long distance-correlation to the Newark record at the stage level overlaps the Ghost Ranch and Montoya pCO<sub>2</sub> estimates with our pre-CAMP baseline. Although their estimates are highly variable, they are not inconsistent with pre-CAMP data from the Passaic (Schaller et al., 2011a) or New Haven Formations presented here ( $\sim 2000 \pm 700$  ppm). The variability in the Cleveland et al. (2008) data can be attributed to their lack of multiple down-profile isotope measurements, such that equilibration with soil CO<sub>2</sub> is unknown, lack of individual organic measurements from each soil, and also to their use of temperatures estimated from the  $\delta^{18}\text{O}$  of pedogenic calcite, which has not been rigorously calibrated for use in the paleo-record (Dworkin et al., 2005).

Several marine sections spanning the Late Triassic to Early Jurassic show evidence for a marked increase in basalt weathering product. Most notably, substantial fluctuations in the  $^{187}\text{Os}/^{188}\text{Os}$  of seawater between  $\sim 203$  and 197 Ma are found in marine sections from St. Audrey's Bay, UK (Cohen and Coe, 2007), and the Kurusu section from Japan (Kuroda et al., 2010), an effect which is attributed to increased flux of unradiogenic  $^{188}\text{Os}$  from the weathering of the CAMP (Cohen and Coe, 2007) (Fig. 6). To explain this relative increase in  $^{188}\text{Os}$ , Cohen and Coe (2007) conclude that perhaps as much as 50% of the estimated  $3 \times 10^6$  km<sup>3</sup> of basalt could have been removed in a 3 Myr window following the eruptions, a rate which is close to the current weathering rate of ocean island basalts (Gaillardet et al., 1999). This interpretation is also supported by 2- to 3-fold increases in Os and Re concentrations in the Kurusu section in Japan (Kuroda et al., 2010) that we correlate to the St. Audrey's Bay record via the  $^{187}\text{Os}/^{188}\text{Os}$  peak in both, within the error allowed by the biostratigraphy (Ward et al., 2004). The relatively low  $^{187}\text{Os}/^{188}\text{Os}$  ratio that occurs just after the initial CAMP eruptions indicates that unradiogenic  $^{188}\text{Os}$  was a significant proportion of the weathering flux, and the subsequent slow  $^{187}\text{Os}/^{188}\text{Os}$  increase suggests that the proportion of unradiogenic component progressively decreased as the highly weatherable CAMP silicate was consumed. This scenario is consistent with our uppermost pCO<sub>2</sub> estimate from the Portland formation,  $\sim 1.5$  Myr after the last pCO<sub>2</sub> increase, which suggests the return of atmospheric pCO<sub>2</sub> to equilibrium levels following

consumption of the CAMP basalts (Fig. 5). However, our scenario leaves the decrease in  $^{187}\text{Os}/^{188}\text{Os}$  leading up to the initial CAMP eruptions unexplained.

## 7. Summary and conclusion

This paper presents evidence for the short and long-term effects of the CAMP volcanism on Early Jurassic pCO<sub>2</sub> from strata in the Hartford basin of Eastern North America. We use the pedogenic carbonate paleobarometer (Cerling, 1999) to confirm the findings of Schaller et al. (2011a) in the corollary Newark basin, and to test the million-year scale effect of the CAMP eruptions.

We find that the Hartford basin pCO<sub>2</sub> record is consistent with observations from the Newark basin, where a pCO<sub>2</sub> maximum is identified just after each volcanic episode (to levels around  $\sim 4500 \pm 1200$  ppm). The significantly longer post-extrusive Portland Formation of the Hartford basin shows evidence of a fourth pulse of CO<sub>2</sub> (to  $4500 \pm 1200$  ppm) about 240 kyr after the last lava recorded in the Eastern North American section. We contend that this increase in CO<sub>2</sub> is volcanic in origin and may be attributed to a fourth pulse of widespread magmatism not represented by lavas in the ENA section. Using the fourth Newark Group pulse of CO<sub>2</sub>, found 240 kyr after the Hampden Basalt, we revise the estimated CAMP duration as observed in the Eastern North American section to  $\sim 840 \pm 60$  kyr.

The Portland Formation record also shows a rapid post-eruptive decrease in pCO<sub>2</sub> that reaches pre-eruptive background concentrations of  $\sim 2000 \pm 700$  ppm by  $\sim 250$  to 300 kyr, consistent with observations from the Newark basin. Furthermore, the longer Portland Formation exhibits a long-term decrease in pCO<sub>2</sub> over the subsequent 1.5 Myr following the final eruptive episodes, to levels below the pre-CAMP background. We use a geochemical model based on the COPSE code to demonstrate that the rapidity of the decreases, and the fall to concentrations below background can be accounted for by a 1.5 times amplification of the continental silicate weathering response due to the presence of the CAMP basalts themselves. If basalt has 10 times the reactivity of continental crust, such an amplification would require  $\sim 8.3 \times 10^6$  km<sup>2</sup> of basalt to be erupted, comparable to estimates for the areal extent of the CAMP at  $1.12 \times 10^7$  km<sup>2</sup> (McHone et al., 2003). This finding is generally corroborated by other modeling efforts showing a similar response of the carbon system to the emplacement of the Deccan Traps with  $1.6 \times 10^{18}$  mol CO<sub>2</sub> released over 100 kyr (Dessert et al., 2001). However, our result departs in that we achieve a more rapid response of the carbon system by releasing far less CO<sub>2</sub> ( $3.5 \times 10^{17}$  mol) over an eruption time of 1000 years for any one of the CAMP extrusive units. A longer eruption time therefore would require significantly more CO<sub>2</sub> to be released to achieve the same effect.

Together, these results indicate that continental flood basalts may result in an extreme short-term perturbation of the carbon system, followed by a long-term net-drawdown in pCO<sub>2</sub> to below pre-eruptive levels, implying that LIPs may have an overall net-cooling effect on climate through the connection between pCO<sub>2</sub> and temperature.

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