SUPPLEMENTARY INFORMATION

Antarctic-wide array of high-resolution ice core records reveals pervasive lead pollution began in 1889 and persists today

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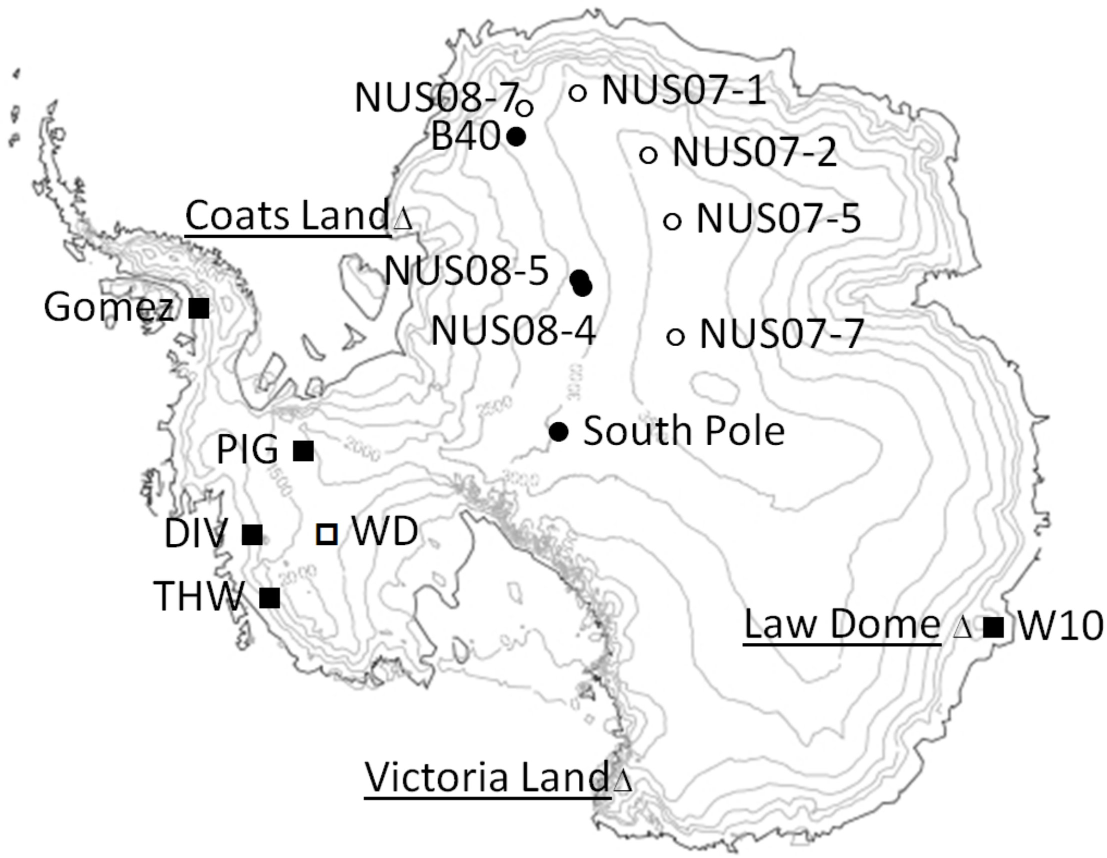
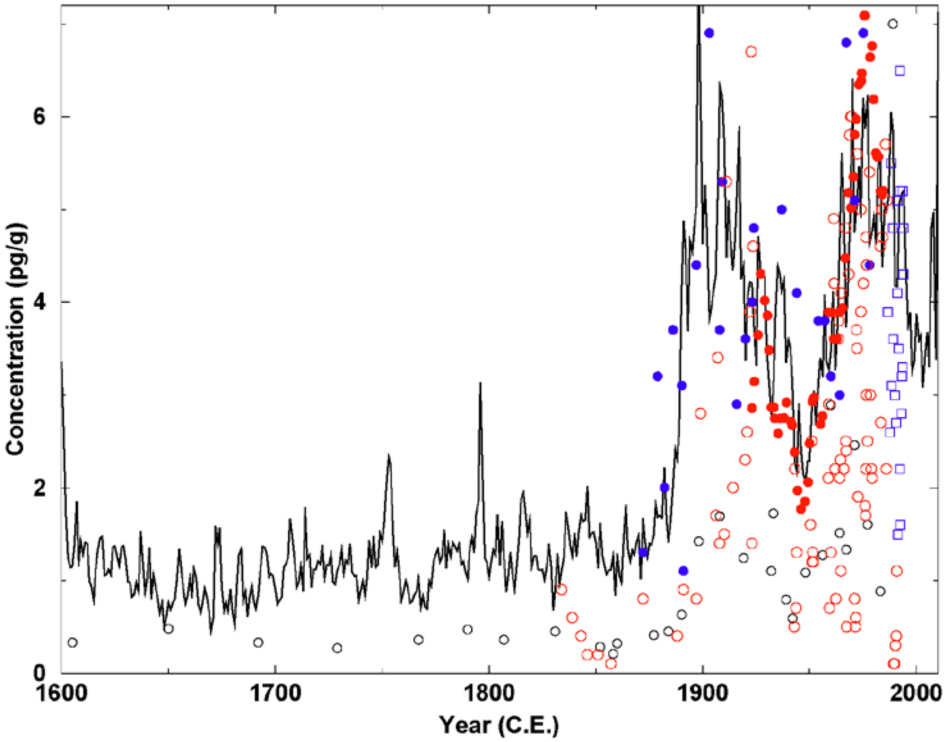
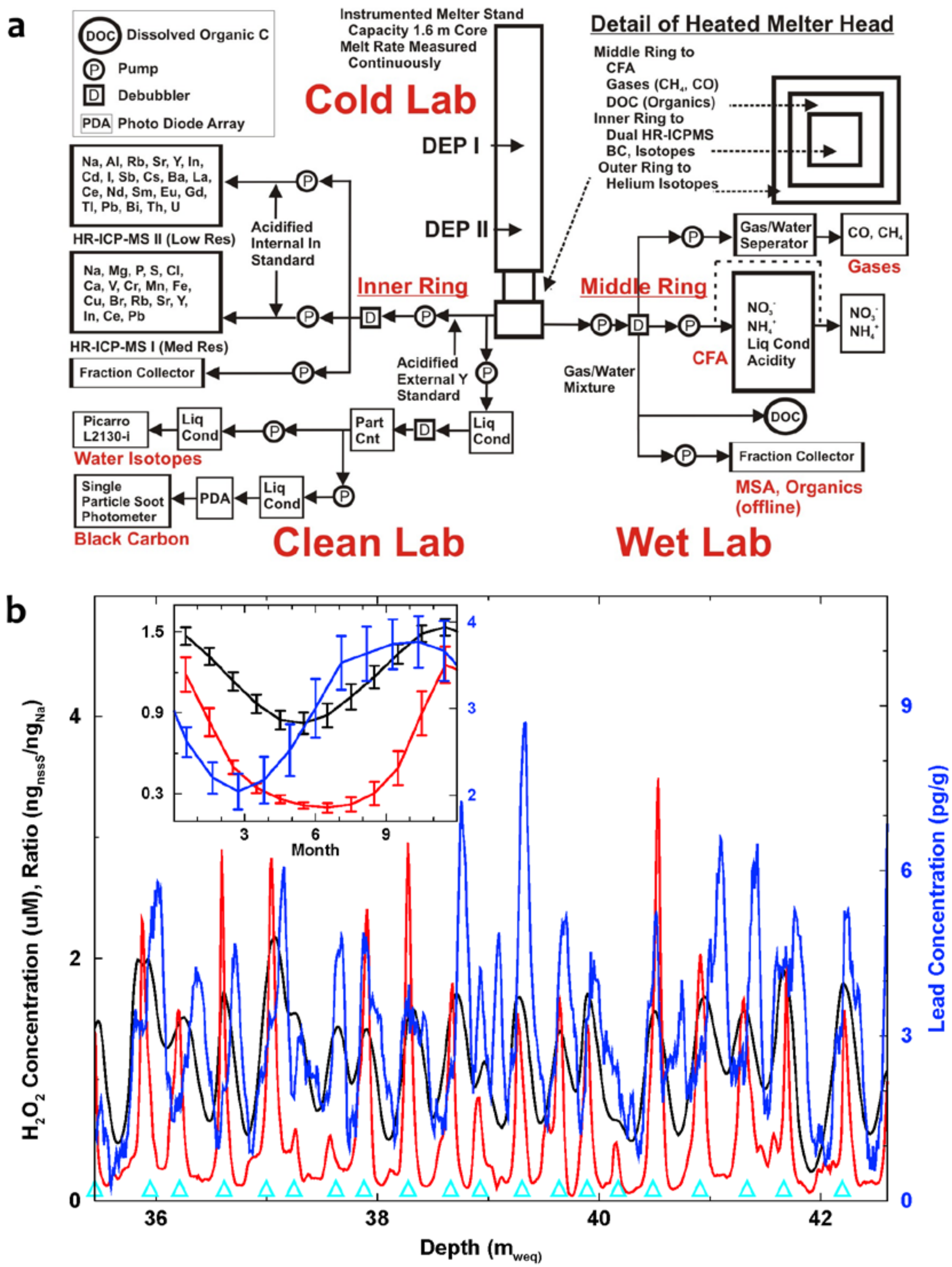


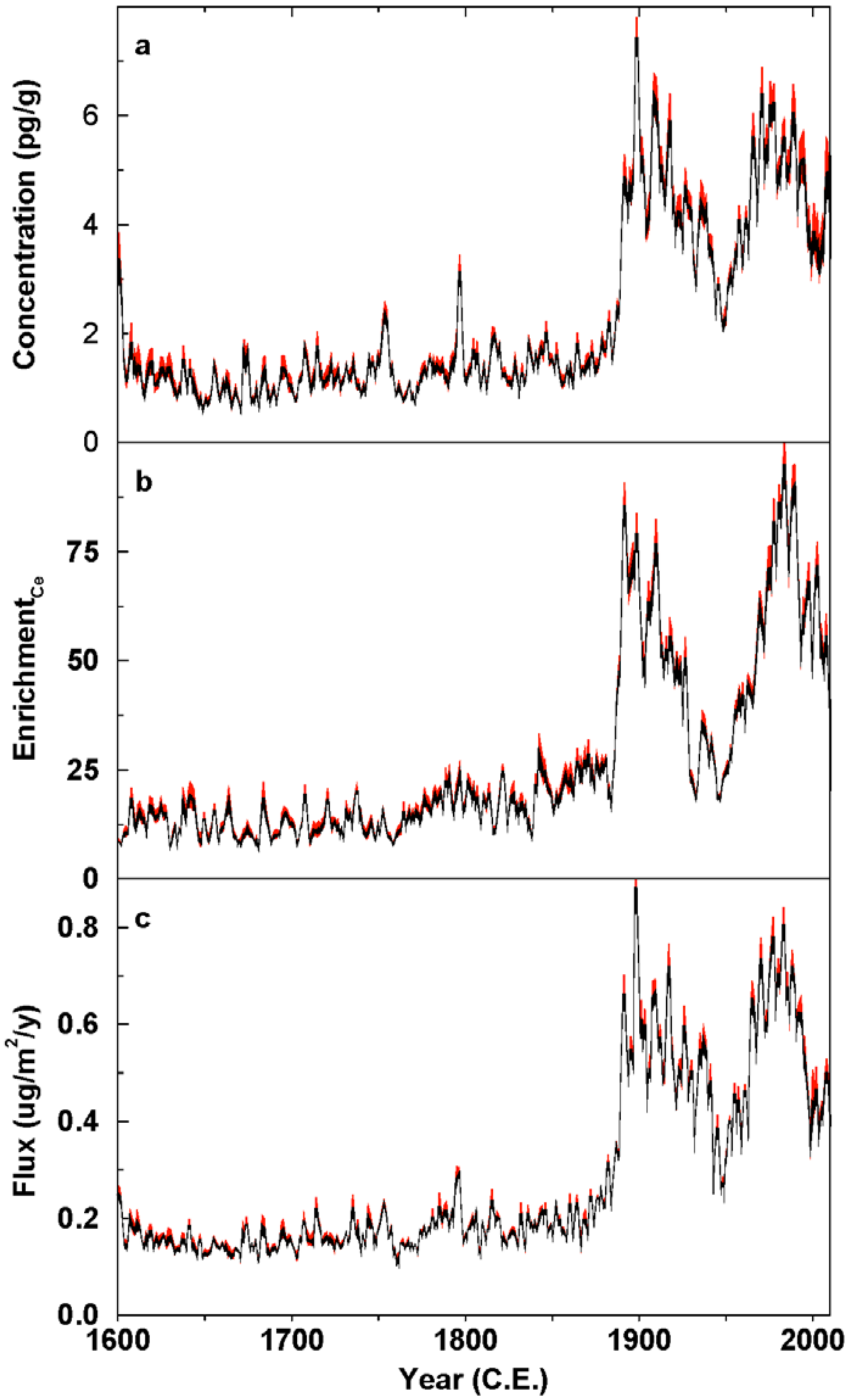
Supplementary Figure S1: Locations of ice cores used in this and previous studies. Ice cores used in this study (■, ● ,□,○); locations used in previous studies (∆); (■,□) lead record at that site measured at sub-annual resolution. Sites (■, ●) also were used in the lead isotope composite. Map generated using Panoply Data viewer; Schmunk, Robert B. (2014). Panoply Data Viewer (version 4.0.1) [Software]. Available from http://www.giss.nasa.gov/tools/panoply/.



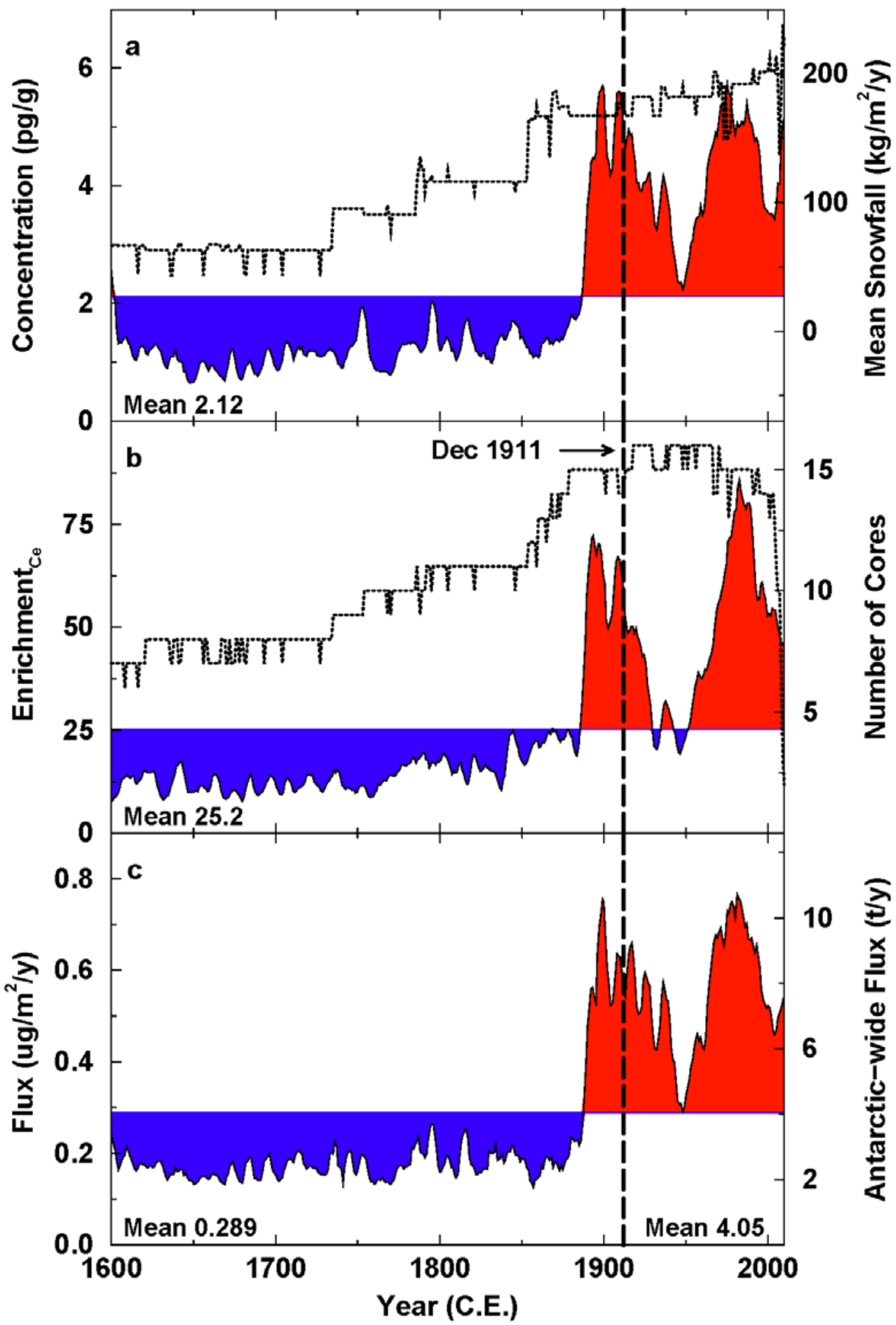
Supplementary Figure S2: Comparison between previous Antarctic lead concentration records (Coats Land[3] [●],[○][9]; Law Dome[6] [O]; Victoria Land ice core[7] [●] and snowpit[7] [□]); and the Antarctic composite record from this study (—). The composite record demonstrates the dramatic increase in lead pollution in Antarctica by comparison with the preindustrial period back to 1600 C.E, that levels in the early 20th century were as high as any time since, and this pollution still persists today.



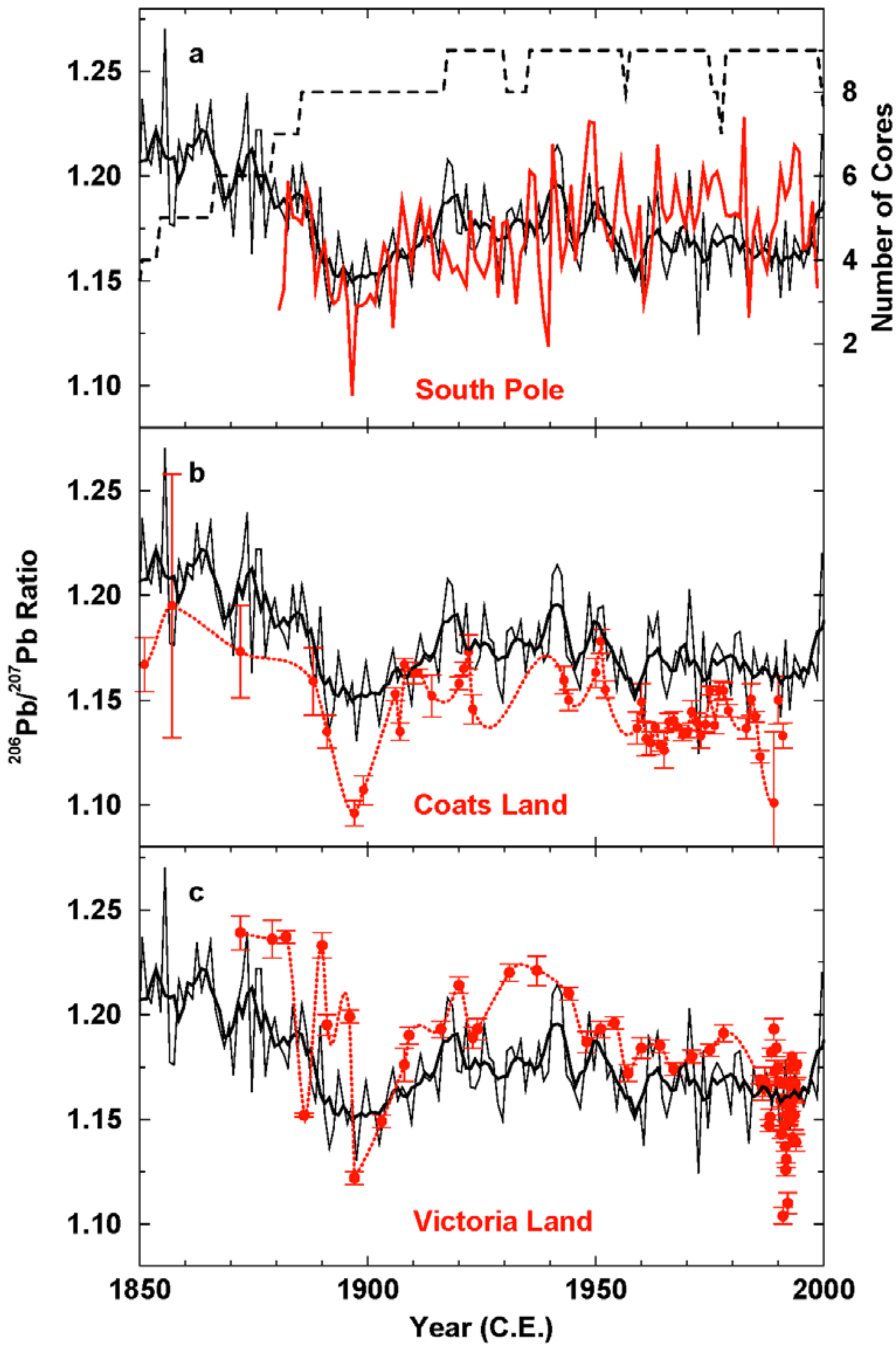
Supplementary Figure S3: (a) Schematic of analytical system. (b) Demonstration of the high-resolution aerosol records obtainable with this technique. This record is from the high-accumulation site (DIV2010) in western Antarctica and spans the period 1900-1920 C.E. Shown are sub-annual records of lead (—) and the non-sea salt sulfur- sodium ratio (—), the peaks of which were used to pick the depth of each summer layer. The lower resolution, hydrogen peroxide signal (H_2O_2, —) was used to support the picking decision. The positions of the annual picks are shown by (∆). Inset: The average annual cycle in each species. Lead shows a consistent spring peak in concentration. Error bars are the standard error of the mean value.



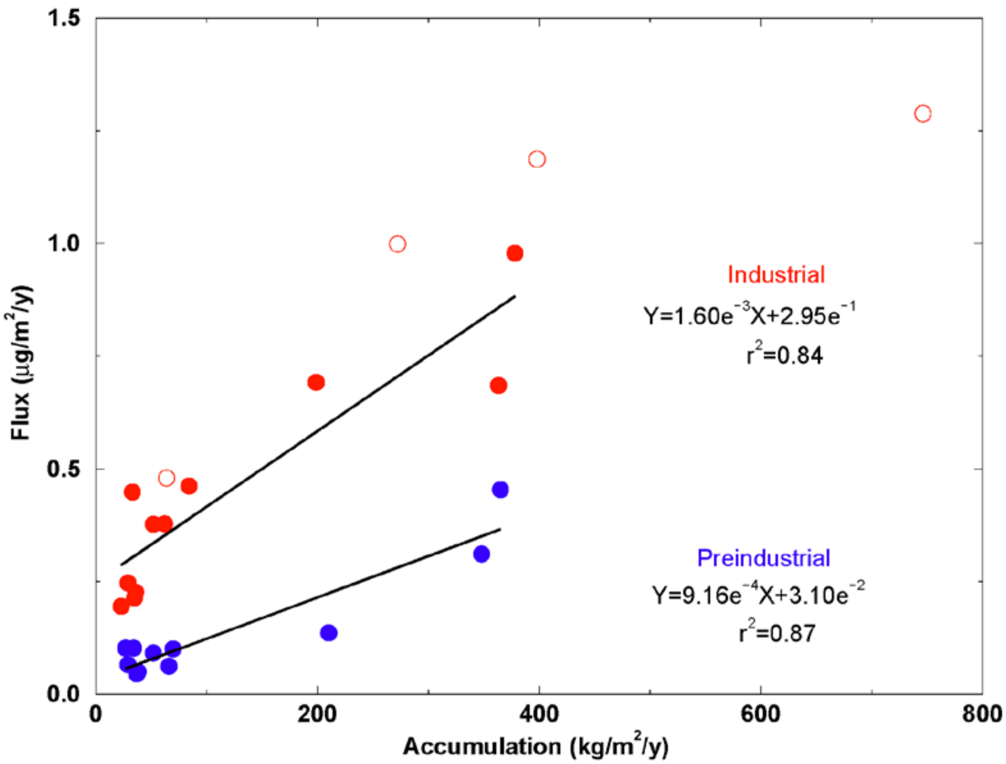
Supplementary Figure S4: Composite ice core records of lead in Antarctica with uncertainties in the annual mean values. The annual mean (geometric mean) of lead flux (concentration and enrichments) is the black line. Red shading indicates the standard error about the mean (geometric mean) for annual flux (concentration and enrichment) measurements. The small deviation of the individual ice cores records from the composite record supports the notion that lead fluxes are well mixed across Antarctica, justifying development of the composite lead records using records from all over Antarctica.



Supplementary Figure S5: Composite ice core record of lead in Antarctica. Records are in five-year average resolution; vertical dashed line indicates the year Amundsen and Scott reached the South Pole. (a) Lead concentration, pg g^{-1} and mean snowfall for the composite record (····). (b) Enrichment of lead relative to the dust proxy, cerium. The number of cores used in the composite is shown by (····). (c) Flux of lead µg m^{2} y^{-1}. Quoted mean values are for the time period 1600-2010. Areas shaded in red indicate time periods where the lead value was above the stated mean; blue shading indicates values below the mean.



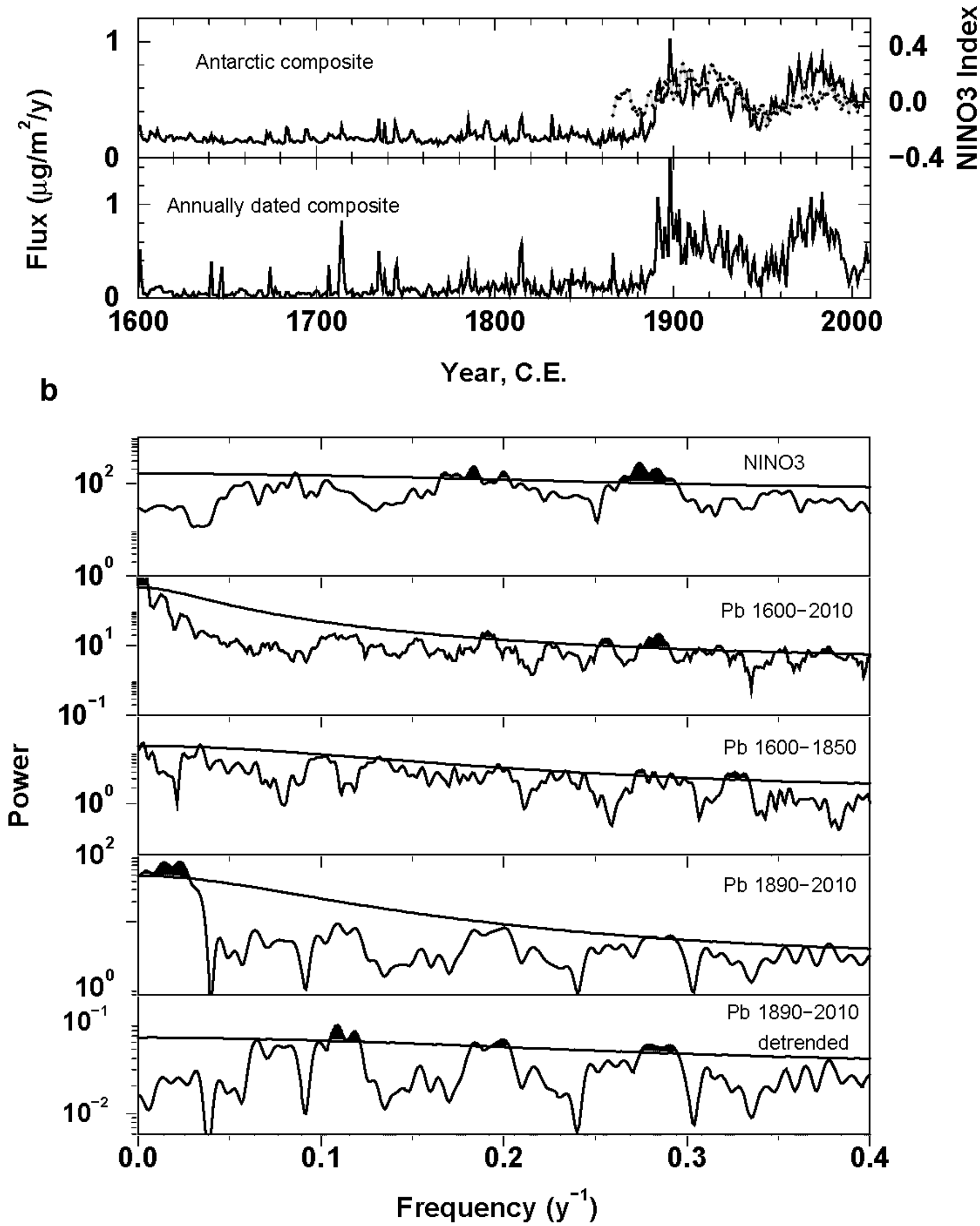
Supplementary Figure S6: Antarctic composite lead isotopic ratios compared with results of other studies. Antarctic composite (—) is displayed as a five-year running average of the annual data (**—**).The number of ice cores used in each part of the composite is shown by (**- - -**). A spline has been fit through the discrete isotope measurements as a visual aid (····). (a) South Pole; measured as part of this study included in the composite record, (b) Coats Land[8], (c) Victoria Land[7].



Supplementary Figure S7: Correlation plot between the long-term averaged lead deposition flux and snowfall accumulation rates at each site. This relationship was used to scale the lead flux at each site and correct for differences in accumulation rates. Preindustrial and industrial relationships cover the years 1600-1850 (●) and 1890-2000 (●), respectively. Only ice cores that extended into both time periods were used in the correlations (filled circles). The ice cores not used in the correlation still follow the same trend (○). The y-intercept of the fit is an indication of the dry deposition flux. For background (polluted) conditions, the dry deposition rate was 0.0310±0.004 (0.247±0.009) µg m^{-2} y^{-1}, and the wet deposition rate was 0.152±0.004 (0.279±0.008) µg m^{-2} y^{-1} when evaluated at the average net snowfall rate for Antarctica of 166 kg m^{-2} y^{-1} [32], so dry deposition accounted for ~17% of the total during the background period 1600 to 1850 C.E. but ~47% during the polluted period 1890 to 2000. We combined the preindustrial and industrial sections of the records by weighting the contribution of each record using a linearly increasing scalar during the period 1870–1888 C.E.

a

b



Supplementary Figure S8: Spectral analysis of the flux composite record built from the eight ice core records that were annually dated. (a) Comparison between the total Antarctic flux composite (—) (with NINO3 time series overlaid [•-]) and the geometric average of the annually dated composite that was used in the spectral analysis. (b) Spectral analysis of time series. Frequencies that have spectral power greater than 90% confidence interval are shaded in black. Spectral analysis was performed using the Kspectra tool-kit[24] using the multi-taper technique with a resolution parameter of two and three tapers and significance levels of 90% computed from a red-noise model fit to the input series. The industrial era record was de-trended by a baseline made of a 15-year running average of the annual lead record. The NINO3 record displays significant frequencies of ~three and five years. These same frequencies also are present through the entire lead flux record demonstrating the effect of atmospheric circulation on transport efficiency and thus Antarctic lead deposition. The spectral power over the industrial period is dominated by the ~60-90 year frequency which is describing the influx of lead pollution in Antarctica. Spectral analysis of this time series de-trended with a 15-year running mean shows the increased spectral power of the ENSO frequencies as well as the strengthening of an ~nine-year oscillation.

Supplementary Table S1: Site details of ice cores used in the Antarctic array.

East Antarctic Sites	Latitude	Longitude	Elevation (m)	Lead Isotopes Measured	Dating Method*	Year Drilled	Total Depth (m)	Depth at 1600 C.E. (mweq)[∝]	Bottom Year, (C.E.)	Top Year, C.E.	Accumulation, kg m-2 y-1	Year Analyzed
NUS07-1	73 43S	007 59E	3174	No	Volcanic	2007/08	30.3	-	1755	2005	52	2009
NUS07-2	76 04S	022 28E	3582	No	Volcanic	2007/08	90.37	13.26	337	1993	34	2008
NUS07-5	78 39S	35 38E	3619	No	Volcanic	2007/08	89.49	10.30	-111	1989	27	2009
NUS07-7	82 04S	54 53E	3725	No	Volcanic	2007/08	90.66	12.03	49	2005	29	2009
NUS08-4	82 49S	18 54E	2552	Yes	Volcanic	2008/09	30.1	-	1622	2008	37	2009
NUS08-5	82 38S	17 52E	2554	Yes	Volcanic	2008/09	92.02	14.41	346	2008	37	2010
NUS08-7	74 53S	01 36E	2700	No	Volcanic	2008/09	80.35	31.84	1255	2008	70	2010
South Pole	90 0 0S	0 0 0E	2800	Yes	Volcanic	1999/2000	17.06	-	1880	1999	64	2010
B40	70 0 0S	0 4 5E	2892	Yes	Volcanic	2012/2013	200	27.06	-538	2012	62	2013
W10	66 44 0S	112 50 0E	1390	Yes	Annual	2008/09	125.93	-	1735	2008	351	2010
West Antarctic Sites	**Latitude**	**Longitude**	**Elevation (m)**									
WDC05A	79 28 5S	112 4 1W	1759	No	Annual	2005/06	69.77	-	1775	2005	200	2007
WDC05Q	79 28 5S	112 4 1W	1759	No	Annual	2005/06	129.44	81.2	1520	2005	203	2007
PIG2010	77 57 25S	95 57 42W	1593	Yes	Annual	2010/11	59.43	-	1918	2010	403	2011
DIV2010	76 46 13S	101 44 15W	1329	Yes	Annual	2010/11	111.71	-	1786	2010	372	2011
THW2010	76 57 9S	121 13 13W	2020	Yes	Annual	2010/11	61.74	-	1867	2010	274	2011
Gomez	73 35 0S	71 22 0W	1400	Yes	Annual	2006/07	134.01	-	1854	2006	694	2010

* Volcanic: Ice cores dated using volcanic synchronization to WDC05Q, Annual: Ice cores dated with annual layer counting of multiple chemical parameters.
∝ mweq: Depth in meters water equivalent.