Supporting Information

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SI Materials and Methods

Photographing Specimens. Specimens were imaged with a mirrorless interchangeable lens camera (Sony a7R II) paired with a native 55 mm lens (Sonnar T* FE 55 mm F1.8 ZA), positioned at a fixed height of 72 cm over a self-contained light box (MK Digital Direct Photo-e-Box B10) outfitted with 28-W continuous full-spectrum fluorescent bulbs (6,500 K, 84 CRI) run through 120-V AC 60-Hz electronic ballasts. Specimens were illuminated by using top, side, and back bulbs in the light box, omitting the bottom (stage) bulbs and supplemental LED bulbs to ensure an even distribution of diffuse light from a single illuminant type source. At the beginning of each imaging session, the lighting elements were turned on and allowed to warm up for 20 min before shooting. Specimens were oriented so that the target area on the breast was positioned at the center of the camera’s field of view. The light box was fully enclosed during each exposure, except for a rectangular aperture on the top, sized to fit the camera’s field of view. Overhead lighting was turned off in each of the shooting locations, and windows were covered to further reduce ambient light leakage.

The images were captured in 14-bit uncompressed raw format and analyzed by using RawDigger software (Version 1.2.11), which provides access to raw data directly recorded by the digital camera’s CMOS sensor. Analyzing the raw sensor data directly enabled us to bypass the linearization step described by Stevens et al., 2007, and McKay, 2013 (32, 33), since the raw values have not been altered by nonlinear gamma encoding algorithms that are introduced when raw sensor data are converted into conventional image formats, such as JPEG or TIFF (34). Before shooting, we tested the linearity of the camera’s CMOS sensor following the procedure outlined in Stevens et al., 2007 (32) and we found that the sensor provided a linear response over the entire dynamic range (Fig. S9).

Exposure settings (shutter speed, aperture, and ISO) were optimized through a series of trials using reflectance standards. We conducted trials using four types of reflectance standards, including the XRite ColorChecker Passport (8-step), QPcard 101 (3-step), Labsphere Spectralon Diffuse Reflectance Standards (10 reference targets), and Munsell Neutral Value Scale matte finish (31-step). We found that each standard provided comparable results, but we selected the Munsell Neutral Value Scale as our primary standards because it was relatively affordable, provided the largest number of reference points, and included published reflectance percentages printed directly on the cards for easy reference. To determine exposure settings, we analyzed trial images in RawDigger with a goal of maximizing the dynamic range (defined as the distance between minimum and maximum light intensities) without introducing signal clipping on any of the color channels (R-G-B-G2), which occurs when certain clusters of pixels fall outside of the dynamic range due to overexposure (saturation). It is essential to refer to the raw data when assessing whether signal clipping has occurred, since the channel-specific histograms on many digital cameras’ displays incorporate gamma-encoding algorithms that make it difficult to tell whether signal clipping has actually occurred. Exposure settings maximizing dynamic range will often indicate overexposed areas on the camera’s built-in displays, when no signal clipping in the raw file has taken place.

The ISO was set to 100 to ensure a limited amount of digital noise. Based on the trials, an aperture of f/16 was chosen to minimize optical vignetting (light falloff), which is introduced at lower focal ratios, while providing a depth of field that would ensure that the target area appeared in focus for all specimens, which varied in height due to differences in natural size and preparation of the specimens. With these parameters in place, a shutter speed of 1/25 s was selected to maximize the dynamic range.

While the use of a light box ensured relatively even and continuous illumination compared with open studio lighting arrangements, perfectly consistent illumination is difficult to achieve in practice. Some unevenness was discovered in blank reference images, which was determined to have resulted from lens variables (optical vignetting and lens flare) and may have also been influenced by the arrangement of the bulbs in the light box. To account for these factors, the target area for each specimen was confined to a 3 × 3-inch square, which limited variance in illumination to <1%.

Under the constant lighting conditions that a light box provides, reflectance standards theoretically only need to be photographed once over the course of shooting to generate calibration regressions. In practice, however, some minor variations in overall illumination were discovered between the three locations, which may have been due to light leakage or slight variations in the voltage supply to the bulbs at each location. This variation, however, was easily accounted for by imaging the Munsell Neutral Value Scale reflectance standards at each location and calculating reflectance values for specimens with location-specific reflectance regressions. Since reflectance is expressed as a percentage, and these percentage values are relative to the standards, no additional adjustments were needed to normalize the color channels or calibrate the values across shooting locations. We photographed each card of the Munsell Neutral Value separately at The Field Museum and Carnegie Museum of Natural History, positioning each card at the center of the field of view in the same area where we measured reflectance from bird feathers. To determine reflectance regressions from these locations, we used all 31 reflectance standards (ranging from 3.1 to 90% reflectance). At the University of Michigan Museum of Zoology, we photographed the Munsell Neutral Value Scale fanned out in single photograph. For this sample, we only included 12 reflectance steps (ranging from 9 to 84.2% reflectance) that fell within the target area (Fig. S9).

Determining the Smoothing Function for the GAM. Smoothing parameters for GAMs can be determined in mgcv by using functions such as GCV that minimize residual deviance (goodness of fit) and degrees of freedom (21). With our final dataset, the GAM estimated a smoothing function of k = 10 (this model is plotted in Fig. S10), which recovered a smoother curve than k = 20 (Fig. 2). Oversmoothing, however, can obscure signals in the data (35, 36), which appears to be happening with k = 10 based on our knowledge of likely inflection points (such as the 1929 US stock market crash) that are present in the consumption data and the Greenland ice-core record. For reference, in Fig. S10, we include a variety of smoothing functions from k = 10 to k = 100. Based on the comparison of possible k values, k = 10 appears to apply an overly powerful smoothing operation in the GAM, forcing the first decline of black carbon to begin in the early 1920s rather than the end of the decade where we would expect it to appear based on consumption trends; k = 13 through k = 35 recovers trends that are effectively identical, which appears to recover important signals in the data that over smoothing misses; k = 36 and greater generate toohy trends that overrepresent random variations within the sample set. Based on the variation in the shape of different GAMs, we selected a smoothing
function of $k = 20$ to produce a relatively smooth trend line that still maintained a distinctive shape that allowed for comparison against consumption data.

**How Sampling Months Were Determined.** Beginning in late summer, each species used in the study initiates an annual molt to replace worn and soiled body feathers with fresh plumage. This molting period can last through the fall months (20). Natural variation in the timing of the molt produces a mix of birds with fresh and soiled plumage among specimens sampled from these months. This annual molt signal was apparent in our sample, with samples from fall months producing shifts in mean reflectance caused by the introduction of freshly molted birds, along with uncharacteristically broad ranges in reflectance values compared with other months (Figs. S4 and S5). Freshly molted individuals do not provide evidence for atmospheric conditions in a given year, warranting their removal from the final dataset. Since freshly molted birds begin to accumulate particulate matter immediately after the molting cycle is complete, rather than selectively evaluating which individuals had recently molted, all of the specimens sampled during these months were removed. We determined the months to exclude for each species based on abrupt shifts in mean reflectance between months, which are indicative of annual molting patterns. For example, in Horned Larks, reflectance values shift abruptly between July and August and then increase again between November and December, indicating that the sample of birds in the months of August–November includes a substantial number freshly molted individuals (Fig. S4). Following this method, the months of August–November were excluded for Horned Larks and Red-headed Woodpeckers, and the months of September–November were excluded for Field Sparrows, Grasshopper Sparrows, and Eastern Towhees (Fig. S4). We could be confident in these shifts given their seasonal timing, since overall black carbon emissions seasonally trend in the opposite direction for a given year in the Northern Hemisphere, as fuel consumption increases to meet heating needs when average temperatures drop (8, 15). We limited this inquiry to the years 1880–1950 because after midcentury, birds are substantially cleaner in all months, compromising our ability to detect monthly breakpoints.

**SI Evidence that Bird Specimens Accumulated Black Carbon from the Environment Before Collection**

To link reflectance data to black carbon levels for a single year, it had to be established that black carbon accumulation occurred before collection. Multiple lines of evidence indicated that the black carbon accumulated on bird specimens originated from the environment while the birds were alive and not from posthumous soiling or discoloration that occurred while being stored in a collection:

1. Since posthumous soiling would accrete continuously, if soiling had occurred over time in storage, it would not have been possible to observe seasonal differences, and any monthly trends that result from the annual molting cycle would have been erased or vastly diminished, particularly in older specimens. We found that consistent numbers of birds collected during the fall were much cleaner in a given year, indicating freshly molted individuals (Figs. S4 and S5). These patterns were observable even among birds that had been in the same collections as soiled birds, stored together since the time of collection.

2. We conducted a visual survey of bird specimens collected outside the US Manufacturing Belt from other parts of the United States or from less industrialized countries during our 135-y sampling period. If posthumous soiling had occurred within our sample, we would have expected specimens collected in these nonindustrialized regions to have exhibited comparable levels of soiling to those in our sample, which we did not find. A visual example of this evidence can be seen in Fig. S11, which shows five Horned Larks collected in Illinois and five Horned Larks collected along the western coast of North America. All 10 birds were collected during nonmolting months between 1903 and 1922, a period in which consistently high levels of black carbon deposition were found on bird specimens collected within the US Manufacturing Belt.

3. If specimens in our sample accumulated black carbon from sitting in museum collections, we would have expected specimens to have soiled ventral sides and cleaner dorsal sides because they generally rest in drawers with their breast and belly facing up. The dorsal side of the specimens would thus have been protected from soot precipitate. We found, however, that both sides of specimens exhibited soiling (Fig. S12).

4. If substantial posthumous soiling had occurred within our samples, we would have predicted that the oldest specimens would have been the sootiest based on gradual accumulation over time. However, we found a slight increasing trend in black carbon deposition between 1880 and 1910. Together, these lines of evidence suggest that any posthumous soiling from sitting in museum storage is negligible.
**Fig. S1.** Additional SEM micrographs, taken at different magnifications, from the Field Sparrows (*S. pusilla pusilla*) in Fig. 1. A–D are from the soiled 1906 specimen. E–H are from the clean 1996 specimen. The micrographs for each specimen are progressively higher in magnification. The white boxes in D and H outline the areas shown in Fig. 1.
Fig. S2. Map showing the collection localities for 1,345 of 1,347 specimens used in this study. The remaining two specimens lack county locality data. Counties are shaded based on the density of sampling within the county. The number of specimens from each county is printed within each county.
Fig. S3. Comparisons of old and young specimens for the four species pairs not shown in Fig. 2. (A) Grasshopper Sparrows (A. savannarum pratensis) from 1907 (Upper) and 1996 (Lower). (B) Horned Larks (E. alpestris pratensis) from 1904 (Upper) and 1966 (Lower). (C) Eastern Towhees (P. erythrophthalmus erythrophthalmus) from 1906 (Upper) and 2012 (Lower). (D) Red-headed Woodpeckers (M. erythrocephalus) from 1901 (Upper) and 1982 (Lower).
Fig. S4. Monthly trends in black carbon deposition for each species before 1950. Inverse reflectance is reported rather than reflectance to express drops in black carbon emissions, which register as increased reflectance values. The shaded areas are the months excluded from final analyses for each species, which are applied to all years. Sampling is sparse for Grasshopper Sparrows and Field Sparrows in the US Manufacturing Belt during fall and winter months because these species predominately migrate out of the region.
Fig. S5. Black carbon deposition for all 1,347 individuals sampled for this study, showing that specimens from molting months (red points) are substantially cleaner than specimens from winter–summer (black points). Black points are individuals included in the final dataset (n = 1,097), and red points are individuals from molting months that were excluded in final analyses (n = 250) (Fig. S4). Inverse reflectance is reported rather than reflectance to express drops in black carbon emissions, which register as increased reflectance values. Before 1950, individuals from molting months are noticeably cleaner than individuals from the rest of the year, warranting the exclusion of specimens from these months for all years.

Fig. S6. Black carbon deposition on specimens plotted against US coal consumption for the three time periods defined by the dashed lines in Fig. 2. (A) Between 1880 and 1910, black carbon deposition is not correlated with coal consumption. Black carbon deposition is high and remains relatively constant, trending upward only slightly as consumption increases sharply. (B) Between 1911 and 1960, black carbon deposition and coal consumption are positively correlated. (C) After 1960, black carbon deposition is decoupled from coal consumption. As consumption increases, black carbon deposition remains low. Before 1950, fuel consumption data are only available in 5-y intervals. We thus interpolated consumption values between points to estimate consumption for the year in which each specimen was collected before 1950. After 1950, yearly fuel consumption data are available.
Fig. S7. Black carbon deposition on specimens (five bird species) from the US Manufacturing Belt, collected between 1880 and 2015. Each point represents the z score for an individual specimen ($n = 1,097$), based on the inverse raw reflectance value taken from its breast and belly feathers. The black line in Upper is a GAM ($k = 20$) with 95% confidence limits (indicated by the shaded area), determined from the individual specimens (details on how $k$ was determined can be found in SI Materials and Methods and Fig. S10. Fig. S13 shows species-specific trends). The colored lines are consumption trends for biofuels and fossil fuels expressed in British thermal units (BTUs) (US Energy Information Administration). Before 1950, fuel consumption data are available in 5-y intervals. After 1950, fuel consumption data are yearly. Lower shows estimates of total US black carbon (BC) emissions from Bond et al., 2007 (11), which uses fuel consumption data and emission factor data to generate a historical emission inventory. The dashed line at 1910 denotes the progressive shift in cities within the US Manufacturing Belt from prosecuting to educating emissions violators. The dashed line at 1960 denotes the approximate moment after which black carbon emissions becomes decoupled from coal consumption.

Fig. S8. Black carbon deposition on specimens plotted against black carbon (BC) emissions estimates from Bond et al., 2007 (11) for the three time bins defined in Fig. 2. The second two time bins (1911 to 1960 and 1961 to 2014) are combined to illustrate the strong correlation across both intervals. (A) Before 1910, we recovered relatively constant, high levels of black carbon deposition on specimens, while Bond et al., 2007 (11) estimated a sharp rise in black carbon emissions. (B) After 1910, black carbon deposition is positively correlated with black carbon emissions estimates from Bond et al., 2007 (11). Our results independently recovered similar trends in atmospheric black carbon. Bond et al., 2007 (11) report BC emissions in 5-y intervals. We thus interpolated emissions values between points to estimate values for the year in which each specimen was collected.
Fig. S9. Raw R, G, and B channel-specific regressions based on the Munsell Neutral Value Scale reflectance standards for each shooting location. The regression equations for each channel were used to calculate channel-specific reflectance from raw CMOS sensor data recovered in RawDigger for each specimen.
Fig. S10. GAMs with various smoothing functions applied to the normalized 1,097-specimen dataset. $k = 10–12$ applies an overly powerful smoothing operation in the GAM; $k = 13–35$ recovers trends that are effectively identical, which appear to recover important signals in the data absent from the $k = 10–12$ models; and $k = 36$ (and greater) generates a toothy trend that overrepresents random variations within the sample set.
Fig. S11. Ten Horned Larks (*E. alpestris pratensis*) at The Field Museum, showing that specimens collected in nonindustrial regions do not exhibit comparable levels of soiling to birds collected within the US Manufacturing Belt. The five specimens in *Left* were collected in Illinois, inside the US Manufacturing Belt. The five specimens in *Right* were collected along the western coast of North America, outside of the US Manufacturing Belt. All 10 specimens were collected during nonmolting months (January–April) between 1903 and 1922.

Fig. S12. Images of the dorsal side of specimens from Fig. 1 and Fig. S3. These images, paired with Fig. 1 and Fig. S3, show that even soiling appears over the entire bird, indicating that the soiled birds in our sample acquired black carbon from the environment while alive. (A) Field Sparrows (*S. pusilla pusilla*) from 1906 (Upper) and 1996 (Lower). (B) Grasshopper Sparrows (*A. savannarum pratensis*) from 1907 (Upper) and 1996 (Lower). (C) Horned Larks (*E. alpestris pratensis*) from 1904 (Upper) and 1966 (Lower). (D) Eastern Towhees (*P. erythrophthalmus erythrophthalmus*) from 1906 (Upper) and 2012 (Lower). (E) Red-headed Woodpeckers (*M. erythrocephalus*) from 1901 (Upper) and 1982 (Lower).
Dataset S1. List of vouchered specimens used in the study along with reflectance data

In Column J, two-letter abbreviations are used for each state. Columns L–O report raw data recorded by the digital camera’s CMOS sensor. Column P reports the area sampled on each specimen. Columns Q–T report color channel-specific reflectance values, calculated from the raw sensor data and linear regressions from Fig. S10. CM, Carnegie Museum of Natural History, Pittsburgh; EATO, Eastern Towhee, FISP, Field Sparrow; FMNH, Field Museum of Natural History, Chicago; GHSP, Grasshopper Sparrow; HOLA, Horned Lark; RHWP, Red-headed Woodpecker; UMMZ, University of Michigan Museum of Zoology, Ann Arbor.