Cleaning procedures

The Teflon-lined air and water hoses were rinsed thoroughly with methanol and acetone and put in an oven (60° Fahrenheit) for 1 h to ensure that all solvent had evaporated. The aluminum snow-maker was washed with Neutrad soap (Thermo Fisher Scientific, Waltham, MA, USA) and water, followed by sonication in methanol and an acetone rinse. The connecting stainless steel parts were also washed with Neutrad soap and water and then rinsed with acetone, hexane and acetone. All parts were set to dry in a clean hood. Teflon fittings were submersed in a 20% HCl bath for 2 h, rinsed with 1% HCl solution, followed by milliQ water and allowed to dry. Glass bottles were washed with Neutrad soap and water, rinsed with acetone and ashed at 475°C for 8 h. Teflon-lined lids were washed with soap and water, rinsed with hexanes and dried in a clean hood. Cleaned bottles were stored closed in sealed plastic bags.

Equation fits and derivations for labelled compound loss

Derivation of the first and second order rate laws to yield an equation equaling C(t). The final C(t) equation derived here was used to fit the experimental data to an equation which could be used to predict the concentration at any point in time based on the observed degradation of the isotopically labelled compounds in the snow. As discussed in the text all reaction orders, etc., are determined based on a threshold value after which no further degradation is observed, and complete degradation is assumed not to take place based on the available data. For first order reactions, the rate law is:

$$\frac{dC}{dt} = -k(C - C*) \qquad (S1)$$

when integrated yields:

$$\frac{\ln(C - C*)}{\ln(C_0 - C*)} = -kt \qquad (S2)$$

after some rearrangement to solve for C(t):

$$C(t) = e^{-kt}(C_0 - C*) + C* \qquad (S3)$$

For second order reactions, the rate law is:

$$\frac{dC}{dt} = -k(C - C*)^2 \qquad \text{(S4)}$$

when integrated yields:

$$\frac{1}{C - C*} = 2kt + \frac{1}{C_0 - C*} \qquad \text{(S5)}$$

after rearrangement to solve for C(t) is:

$$C(t) = \frac{1}{\left(\frac{2kt + 1}{C_0 - C*}\right)} + C* \qquad \text{(S6)}$$

where C_0 is the concentration at time t = 0 h, $C*$ is the threshold value, k is the reaction rate constant and t is time in h.

_n_-Alkane and acid flux value calculations

For non-labelled compounds a flux (depositional) component was added to describe the increase in concentration observed for _n_-alkanes and _n_-alkanoic acids, while still taking into account the degradation observed in the labelled compounds. Equation 1 in the main text was rearranged and solved for F, yielding:

$$F = \frac{\left[\frac{\Delta C}{\Delta t} + k(C - C*)\right]}{\left[\frac{SA}{m}\right]} \qquad \text{(S7)}$$

to calculate the flux values. As given in the text, the variables are defined as follows: C is concentration (ng kg^{-1}), t is exposure time (h), $C*$ is the threshold concentration (ng kg^{-1}), k is the reaction rate constant, here first order only (h^{-1}), SA is the surface area of the exposed snow (m^2), m is the mass (kg) of the snow being exposed (kg), and F is flux (ng h^{-1} m^{-2}). The reaction rate used to calculate the flux values is listed in Table 2. While it would not be expected that all compounds of the same compound class (e.g., PAHs) would have the same reaction rate constant, the flux values presented here are an estimate of possible flux values. The experimentally determined k-values from the labelled compounds were used to calculate the corresponding non-labelled compound flux values. For the non-labelled compounds which did not have an equivalent labelled compound, average rate constants for the corresponding compound class were used to calculate the flux values. For the _n_-alkanes the reaction rate constant for docosane-d_{46} was used, firstly, because the heptadecane-d_{36} did not degrade and, secondly, because the _n_-alkanes evaluated were closer in chain length (all greater) to

the docosane than to heptadecane. For the *n*-alkanoic acids the two reaction rate constants for the labelled compounds were relatively similar (0.070 and 0.067 h^{-1}), so the average (0.068 h^{-1}) was used to calculate the flux, except in the cases where the non-labelled and labelled compounds were the same, in which case the reaction rate constant of the corresponding compound was used.

In order to get the predicted concentration of the non-labelled first order compounds at any time t, taking the degradation and flux components into account, Eqn. 3 needed to be integrated and solved for C(t). Again, all equations are normalized to the threshold value of C*.

$$\frac{dC}{dt} = -k(C - C^*) + F\left(\frac{SA}{m}\right) \qquad \text{(S8)}$$

some rearrangement yields:

$$\frac{dC}{-kC + kC^* + F\left(\frac{SA}{m}\right)} = dt \qquad \text{(S9)}$$

where $kC^* + F\left(\frac{SA}{m}\right)$ can be viewed as a constant, and assigned to one variable, b, and –k as another constant, a, thus simplifying the equation to:

$$\frac{dC}{aC + b} = dt \qquad \text{(S10)}$$

After integration, Eqn. S10 yields:

$$\frac{1}{a[\ln(aC + b) - \ln(aC_0 + b)]} = t - t_0 \qquad \text{(S11)}$$

After simplifications,

$$\frac{1}{a\left[\ln\left(\frac{aC + b}{aC_0 + b}\right)\right]} = t \qquad \text{(S12)}$$

solving for C(t),

$$C(t) = \frac{e^{ta}(aC_0 + b) - b}{a} \qquad \text{(S13)}$$

and re-substituting a and b, to get the final equation:

$$C(t) = \frac{e^{-kt}\left[-kC_0 + kC* + F\left(\dfrac{SA}{m}\right)\right] - \left[kC* + F\left(\dfrac{SA}{m}\right)\right]}{-k} \quad \text{(S14)}$$

PAH flux calculations

The depositional flux calculations for the PAHs were simplified because unlike the alkanes and alkanoic acids, the concentration of the non-labelled PAHs did not increase over the exposure time. Their concentrations were higher than that predicted by the degradation of the labelled PAH compound, but remained relatively steady from 24 to 72 h of exposure. Therefore it was assumed that the average steady state concentration had been reached and all of the concentrations from 24, 48, and 72 h were averaged for each set. From that the following equation was used to estimate the depositional flux,

$$F \cdot SA = \left(\frac{dC}{dt}\right)m \quad \text{(S15)}$$

where dC/dt is equal to the initial rate law, $k(C-C*)^2$, m is the mass of the snow (kg), SA is the bulk surface area of the snow (m^2), and F is the deposition flux (ng h^{-1} m^{-2}).