The Atmosphere During the Younger Dryas

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One of the most dramatic climate change events observed in marine and ice core records is the Younger Dryas, a return to near-glacial conditions that punctuated the last deglaciation. High-resolution, continuous glaciochemical records, newly retrieved from central Greenland, record the chemical composition of the arctic atmosphere at this time. This record shows that both the onset and the termination of the Younger Dryas occurred within 10 to 20 years and that massive, frequent, and short-term (decadal or less) changes in atmospheric composition occurred throughout this event. Changes in atmospheric composition are attributable to changes in the size of the polar atmospheric cell and resultant changes in source regions and to the growth and decay of continental biogenic source regions.

During the last ~70,000 to 10,000 years, the North Atlantic experienced rapid changes in sea-surface temperature coincident with rapid changes in the bulk chemistry, dust concentration, and temperature of precipitation, which are unparalleled in the Holocene record (*1–4*). Some of these events have been related to processes that controlled the rate of thermohaline overturn and consequently the heat budget of the North Atlantic (*5*). Advances in our understanding of the coupled ocean-atmosphere responses that characterize these events (*2, 6*) depend on the retrieval and interpretation of high-resolution paleoclimate records. The most highly resolved view of the paleoatmosphere of the North Atlantic region is contained in ice core records recently recovered from two concurrent deep drilling activities located at Summit, Greenland: the U.S.–Greenland Ice Sheet Project (GISP2) and the European Greenland Ice Core Program.

In this report, we describe glaciochemical records (chloride, sulfate, nitrate, sodium, magnesium, potassium, calcium, and ammonium) from the GISP2 ice core (72.6°N, 38.5°W; elevation, 3200 m) for the period of the most significant rapid climate change event of the last deglaciation in the North Atlantic region, the Younger Dryas (YD). All chemical sampling and analysis undertaken on the GISP2 core utilized specially adapted glaciochemical techniques (*7*). High-resolution (mean of 3.48 years per sample during the YD), continuous glaciochemical series were developed based on the use of multivariate annual layer dating of the core (*8, 9*). This annual layer counting has also allowed redating of the YD as an event of 1,300 ± 70 years that terminated at 11,640 ± 250 years ago (*8*).

The GISP2 glaciochemical series yields a unique record of the major soluble constituents transported in the atmosphere and deposited over central Greenland. The calcium series covers the period ~10,000 to 20,000 years ago (Fig. 1). Crustal materials are the major source for calcium in modern Greenland snow (*10*); hence, calcium concentrations provide a sensitive monitor of changes in crustal dust loading over Greenland. The YD and Oldest Dryas (OD) stand out prominently in the record as periods of substantially elevated dust concentrations. Furthermore, a series of peaks are evident, spaced approximately every 400 to 500 years: in the early Pre-Boreal (PB) at 11,400 ± 250 years ago (*11*); in the YD at 11,810, 12,220, and 12,640 ± 250 years ago; and in the Bolling/Allerod (BA) at 13,180, 13,650, and 14.020 ± 520 years ago.

The atmospheric chemical signals recorded as concentration in ice sheets may be modulated by accumulation rate (*12*). Because the YD is characterized by a factor of 2 decrease in accumulation rate (*8*), we examine chemical inputs in terms of flux [species concentration times accumulation rate (*13*)]. Each flux series is similar to its respective concentration series except for ammonium and nitrate (Fig. 2). In order to investigate all of the soluble species measured in this study, the remainder of our discussion is in terms of flux.

The various chemical fluxes can be differentiated into five groups (crustal, sea salt, ammonium, nitrate, and sulfate) based on chemical association or signal characteristics, or both, described below. Each flux signal is presented in two parts (Fig. 3) to facilitate the examination of major patterns [splines (*14*)] and high-frequency variability (residuals).

Calcium and magnesium are primarily contributed to the core site from crustal sources (*10*). Increased fluxes imply more intense circulation over continental regions, increased aridity (*12*), or changes in source area. Crustal source fluxes rise sharply at the onset of the YD, and during the YD there were at least three lesser increases superimposed on an overall decline (Figs. 2 and 3). Fluxes declined abruptly at the end of the YD but increased again during the early PB. Fluxes were quite variable (decadal and less fluctuations seen in residuals) throughout the YD, in particular during the early YD.

The fluxes of those species derived primarily from sea salt, namely, sodium, chloride, and potassium (*10*), show a pattern similar to that of the crustal group, although the sea salt group does not display as dramatic a decline in overall flux over the YD as the crustal group.

Ammonium deposited in Greenland ice

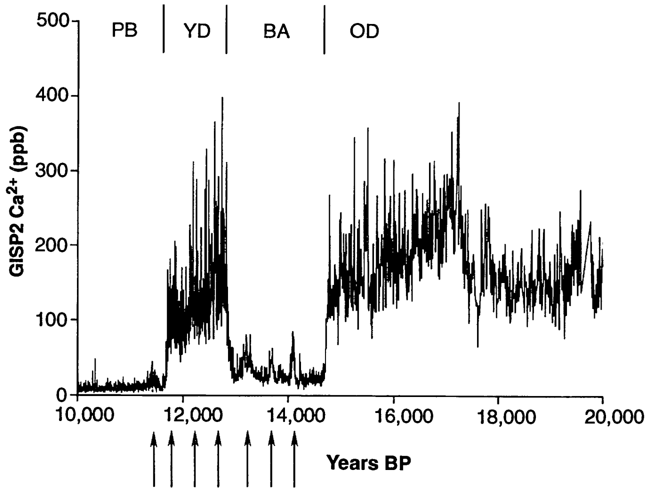

Fig. 1. Calcium concentration (ppb) covering the period ~10,000 to 20,000 years ago. Sample resolution is ~2 years through the Holocene, a mean of 3.48 years within the YD and BA, and ~3 to 15 years during the OD. Arrows mark periods of increased calcium concentration referred to in the text. Years BP, years before present.

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is emitted originally as NH_3 from continental biogenic sources (15). Prominent peaks in ammonium (residuals) from central Greenland cores have been attributed to biomass burning (16). Ammonium flux increases briefly during the earliest YD (Figs. 2 and 3) and then remains relatively low through the YD. It increases and shows high variability after the YD.

Nitrate is contributed by a variety of sources: lightning, NO_x produced from N_2O oxidation in the lower stratosphere, galactic cosmic rays, or surface sources (17). Nitrate fluxes rise only immediately after the YD.

Sources of sulfate to the core site include a combination of volcanic activity, marine biogenic sources, and evaporite sources. Prominent volcanic events may be represented by some of the residuals in the series, but the overall importance of volcanic source sulfate is less than other sources in the recent record (18) and is as yet undifferentiable during periods of high dust. The sulfate flux shows several oscillations during the YD, with the largest increase during the BA.

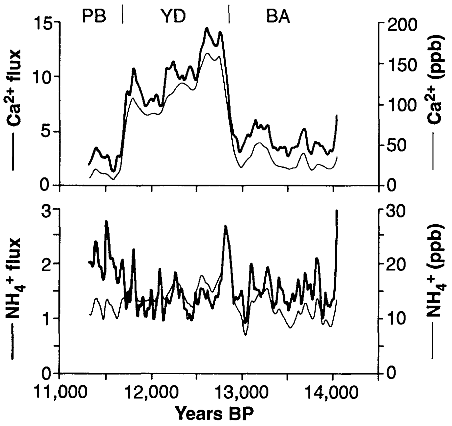

Fig. 2. Comparison between concentration (ppb) and flux (in kilograms per kilometer squared per year) for calcium and ammonium [robust spline smooth (70%) versions] over the time period 11,322 ± 250 to 14,035 ± 520 years ago.

Table 1. The first three eigenmodes for the zero-meaned YD flux series reported as flux (in kilograms per kilometer squared per year). Positive and negative values within eigenmodes reflect only the relative association between variables. Bartlett's test (23) for equality among the eigenmodes showed that the joint probability of equality was less than 0.001.

Species	Eigenmode 1	Eigenmode 2	Eigenmode 3
Calcium	1.98	1.05	1.16
Magnesium	0.20	0.12	0.09
Sodium	0.52	−0.02	−0.38
Chloride	0.85	−0.35	−0.89
Potassium	0.06	0.00	0.00
Ammonium	0.12	−0.66	0.58
Nitrate	0.72	−1.12	−0.71
Sulfate	2.40	3.00	3.2

Because our chemical record is multivariate and individual species may have multiple or related sources, associations between species provide an additional tool for investigating source type and transport path. We applied empirical orthogonal function (EOF) analysis (19) to the YD portion of the flux series to investigate associations. Our data are particularly well suited to this approach because the combined series measure the bulk of the soluble chemistry in the atmosphere. We consider that the principal components (eigenmodes) deduced from this analysis represent the chemical composition of "air mass types" affecting central Greenland during the YD. Results of the first three eigenmodes explain ~82% of the variance in the YD flux series (Table 1). EOF analyses that explain a large proportion of the variance in a series are believed to have a strong likelihood of being physically meaningful (19).

Much of the total variance is explained in eigenmode 1 (58.5%). Because all species are positively loaded (synchronous input), we assume that this eigenmode provides a description of the chemical composition of the "background" atmosphere (dominated by sulfate, calcium, chloride, and nitrate).

Eigenmode 2 (13.8% of the total variance) is most heavily loaded on sulfate, nitrate, calcium, and ammonium. It reveals a negative correlation between crustal and sea salt species, suggesting different transport routes or input timing from these two source regions. Sulfate associated with this eigenmode may be largely crustal rather than biogenic in origin because it is correlated with crustal species. Nitrate is correlated with sea salt species, suggesting transport in a form such as $NaNO_3$. Ammonium is associated with nitrate and sea salt species rather than crustal species, suggesting an air mass composed of NH_4NO_3. The low sodium/chloride ratio in eigenmode 2 indicates that the chloride is from a more distant marine source (allowing fractionation to result in an increase in excess chloride).

Eigenmode 3 (10% of the variance) is loaded primarily on sulfate, calcium, sea salt, nitrate, and ammonium. As for eigenmode 2, sea salt is negatively related to crustal species and sulfate is associated with crustal species. However, sodium/chloride ratios for this eigenmode suggest less distant transport. Increased nitrate is once again associated with sea salt species, but ammonium is associated with sulfate [perhaps as $(NH_4)_2SO_4$] and crustal species.

Our EOF analyses demonstrate that increased wind strength alone cannot account for the chemical flux series distribution during the YD. At least 25% of the total variance (eigenmodes 2 and 3) and perhaps a portion of eigenmode 1 (although not demonstrable with this analysis) require

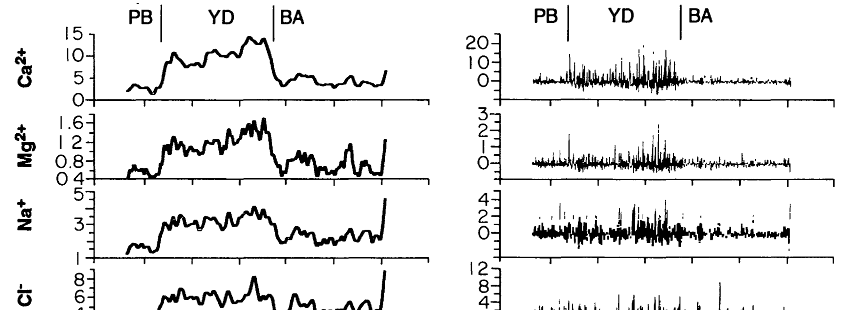

Fig. 3. Robust spline smooth (left) (70%) and residual from spline (right) (both as flux in kilograms per kilometer squared per year) for all species covering the time period 11,322 ± 250 to 14,035 ± 520 years ago.

either source changes or different transport paths to explain flux series associations. Furthermore, this analysis allows the characterization of a "background" atmosphere and two independent air mass types. The air mass type characterized by eigenmode 2 is composed of continental species (calcium, magnesium, and evaporite source sulfate) and contains long-traveled sea salt plus NH_4NO_3 and $NaNO_3$ introduced to the air mass during periods when continental source loading was low. That characterized by eigenmode 3 is primarily composed of continental source species (same as eigenmode 2 plus ammonium) and relatively short-traveled sea salt species, plus $NaNO_3$ introduced during periods of low continental source loading.

The data lead to the following reconstruction. During the BA, fluxes of crustal and sea salt species over the Greenland ice sheet were generally higher than during the Holocene but significantly lower than during the YD. Detailing of periods of increased atmospheric loading of crustal and sea salt species during the BA may be useful in reconstructing climate at this time. Between 13,713 and 13,531 ± 520 years ago, there is evidence of a massive increase in sulfate flux. The size of this increase and its timing relative to the size and timing for other species could suggest a localized source such as a polynya or a prolonged period of volcanism. Ammonium and nitrate fluxes appear to be relatively unperturbed during most of the BA. However, an increase in ammonium at 12,859 to 12,786 ± 250 years ago may signal the destruction of BA biomass in response to the onset of cooler and drier YD climate conditions.

Sea salt, crustal, and sulfate species increased to YD levels dramatically over a period of ~10 years. At least three periods of polar cell expansion punctuated the YD. Frequent, massive, short-term (decadal or less) variations in crustal and sea salt fluxes characterize the early YD, reflecting the significant atmospheric reorganization produced by the transition into the YD. If much of the North American ice cover (as opposed to volume) was still intact, as indicated by glacial geologic evidence (*20*), the largest source for increased fluxes of crustal species directly upwind from Greenland at this time would probably have been outwash surfaces south of the ice sheet margin (*21*). Incorporation of this material into Greenland-bound air masses is substantiated by the increased aridity that characterized glacial-type conditions south of the North American ice sheet and by reconstructions of atmosphere and ocean circulation during such periods (*22*). The larger increase in crustal (calcium) versus sea salt species flux at this time further indicates the involvement of a new source area (outwash) for Greenland-bound air masses. We propose that the consequent transport of this new source to central Greenland required expansion of the polar atmospheric cell and that increased fluxes of crustal species provide a monitor of the relative size and strength of this cell. Evidence for different air mass compositions may eventually be unraveled to decipher the complexities of polar cell changes.

Overall high sulfate fluxes plus several maxima characterized the YD. The strong association between sulfate and crustal source species rather than with sea salt species suggests that marine biogenic source sulfate may not have been a significant component of the YD atmosphere.

Crustal and sea salt loading of the atmosphere decreased abruptly (<20 years) at the termination of the YD, consistent with earlier observations of abrupt temperature and dust changes at this time (*2*). A brief period (~100 to 200 years) of polar cell expansion characterized the early PB, evidenced by increased dust levels at this time (Fig. 1). The onset of the PB is marked by massive fluctuations in ammonium, which may reflect either dramatic variations in continental biogenic source strengths or massive fluctuations in the size of the polar cell reminiscent of the climate reorganization at the onset of the YD. Increased nitrate flux at the onset of the PB may be related to the availability of ammonium to produce NH_4NO_3 or increased access to nitrate sources due to changes in circulation.

Several smaller events, with chemical species behavior similar to that of the YD, characterize the BA period, and at least one such event follows the YD. This sequence of events in combination with maxima during the YD may provide evidence of climate forcing on the order of ~400 to 500 years. We suggest that the dramatic chemical flux changes in the YD and the lesser maxima of the BA and PB are produced by changes in the size of the polar cell (changes in atmospheric catchment area) plus changes in chemical species source strength. The magnitude and complexity (massive decadal-scale fluctuations) of the soluble species record documented in the YD portion of the GISP2 core provide evidence of an extremely dynamic atmosphere thus far unparalleled in the Holocene. Investigation of the forcing of such an extreme environmental state offers a new view of climate change.

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