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LETTERS TO THE EDITOR

Comments on "Relative contributions of tropospheric and stratospheric sources to nitrate in Antarctic snow" by M. R. Legrand and R. J. Delmas

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Recently, Legrand and Delmas (1986) have attempted to assess the relative contributions of tropospheric and stratospheric sources to nitrate in Antarctic snow. They concluded that the stratospheric contribution is unimportant and, in particular, that there is no evidence for a signal of solar activity in the ice sequence as we have claimed (e.g., Zeller and Parker, 1981; Laird et al., 1982). We would like to point out a number of shortcomings in their paper which affect the results and the conclusions that can be drawn.

When seeking to take issue with other scientists' work, it is customary to cite their most recently published data. In this case, Legrand and Delmas did not use the latest results available to them (e.g., Dreschhoff et al., 1983; Laird, 1983). A literature search in either the recent *Scientific Citation Indices* or in the indexes to *Current Antarctic Literature* would have uncovered these articles. The search would have revealed the considerable progress made in demonstrating the presence of a solar signal as well as in identifying a specific mechanism, namely solar flares or solar proton events (SPE's). In fact, several solar effects are apparently superimposed on the nitrate time series in Antarctica either directly, as in the case of ionization by aurorae and solar cosmic rays (SCR's), or indirectly, as in modulation of galactic cosmic rays (GCR's) by SCR's.

There are two major events related to solar activity that appear repeatedly in the Antarctic nitrate record on the time scales chosen by Legrand and Delmas. They are (1) a sharp peak in nitrate concentration and/or flux in 1972–1973 and (2) a prolonged period of lower nitrate concentrations between about 1600 and 1750. The importance of the 1972–1973 peak should not be underestimated. It corresponds to the most energetic solar flare of the satellite era which occurred in August 1972. This benchmark is prominent in Jackman et al.'s (1980) estimate of atmospheric NO_x production by various sources and shows up frequently in our data (e.g., Laird et al., 1982; Zeller et al., 1986). The nitrate low in the 17th–18th centuries that we have detected in three ice cores, two from the South Pole and one from Vostok, corresponds to the Maunder Minimum in solar activity near the end of the Neoglacial or Little Ice Age (Zeller and Parker, 1981; Dreschhoff et al., 1983).

While it is important to be current in the literature if another experimenter's data is to be questioned, it is imperative to analyze the data correctly. In this case, a serious error was made by Legrand and Delmas (1986) in their Fig. 7.2 graphical representation of the data from Fig. 1 in Laird et al. (1982), and this produced a major misinterpretation of the results. For a 6-year block of data (1965–1970), about 1.35 kg-N km^{-2} a^{-1} (6 kg-NO_3 km^{-2} a^{-1}) was added erroneously to each year's flux. This error, indicated here in Fig. 1, had the effect of (1) suppressing the highest single peak in 1972–1973 corresponding to the only major solar flare in recent history and (2) increasing significantly the apparent variability in our data. It is difficult to understand why such a critical error was never caught. Not only does the correct data in fact bear a resemblance to Jackman et al.'s (1980) curve of NO_x production

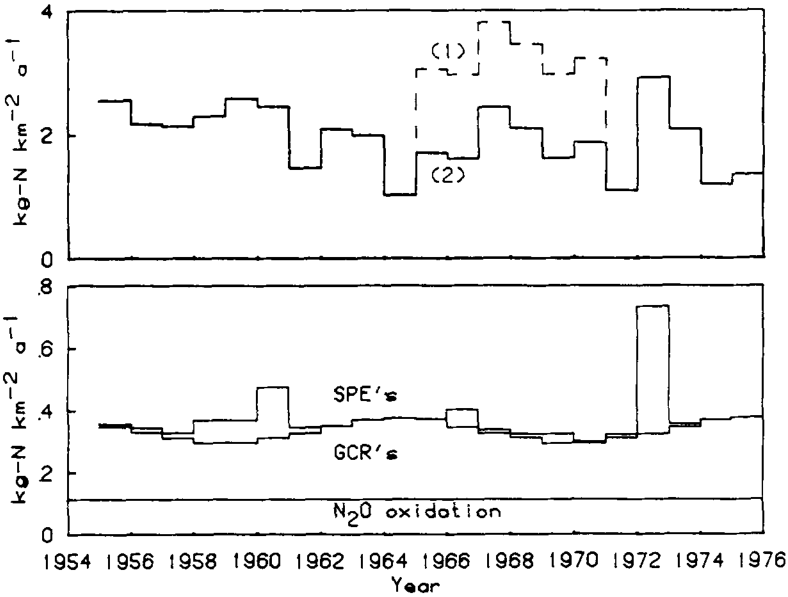

Fig. 1. Nitrate fluxes (kg-N km^{-2} a^{-1}) in South Pole snow from Laird et al. (1982) as depicted by Legrand and Delmas (1986), curve 1, and by Laird et al., curve 2. The bottom 3 curves estimate NO_x production as nitrogen in the polar stratosphere for 1955 to 1975 from N_2O oxidation and from galactic cosmic rays (GCR's) and solar proton events (SPE's) using 2.5 NO_x molecules per ion pair (modified from Jackman et al., 1980; and Legrand and Delmas).

by various sources, including SCR's, but it also has a significant linear relationship ($r > 0.6$, $p < 0.01$) with a 17-year time series of integral SCR fluxes from satellite measurements (Laird, 1983). It is most unfortunate that Legrand and Delmas's time series stops at 1969, thereby just missing the giant flare of August 1972, although the pit was excavated in 1977–1978 and the data should have been available.

It is particularly ironic that Legrand and Delmas chose to question our data on the basis of variability. In fact, we have recently completed an analysis of variance (ANOVA) of South Pole snow and nitrate deposition on 16 columns from 3 parallel trenches covering nearly 60 m^2 and up to 11 years (Laird et al., 1985; Laird, 1986), in which we have shown that the variance of this region is quite high due to drifting, low accumulation, frequent hiatuses in the ice sequence and pockets of anomalously high nitrate values. As glaciologists, Legrand and Delmas should be well aware of the problems at the South Pole (e.g., Gow, 1965), yet they chose not to mention them and insinuated that our data were faulty because the high variability (due in part to their serious digitizing error) was incompatible with their model. In fact, we are suspicious of the low variability in their flux data, which suggests that they picked a sampling site on the pit walls where the stratigraphy was particularly uniform and therefore less representative of true conditions. We think it absurd to presume that 11 years of data from a single column somehow refutes 51 years of data from three replicate columns on the basis of either variability or the presence of a solar signal.

Legrand and Delmas also cite the lack of detectable solar cycles in a Dome C ice core covering 210 years and dating back to about 1760. However, the Dome C nitrate profile does bear a resemblance to our South Pole and Vostok profiles and, based upon Fourier analysis

(Dreschhoff et al., 1983), South Pole nitrate exhibits a strong 22-year cycle and a weaker 11-year cycle. Legrand and Delmas apparently conducted no such analysis and once again their data stop just short of a benchmark period (the Maunder Minimum), so that meaningful conclusions about a solar signal cannot be drawn.

On the question of the source of the nitrate, we would like to make several points. If the nitrate is coming primarily from the troposphere, and in particular from midlatitude lightning, it should be efficiently scavenged long before it reaches the polar plateau (Jackman et al., 1980). In particular, it should have great difficulty penetrating the zone of maximum cyclogenesis just north of the south polar jet stream (Shaw, 1979). Hameed et al. (1981) recognized this fact and reflected it in their model of latitudinal variations of NO_x sources and sinks where the stratosphere:lightning source ratio of tropospheric NO_x ranged from 1:1 at 60°S to as much as 5:1 at 90°S, not 1:1 or less as claimed by Legrand and Delmas. This argument is supported by experimental data from Sanak et al. (1985) of ^7Be and ^{32}P at the French station Dumont d'Urville, showing that the stratospheric contribution is particularly high around Antarctica.

We strongly question the interpretation of Fig. 4. It is apparent from this figure that sea salt (Na^+) decreases inland as a generally linear function of elevation. Once the polar plateau is reached, there is little change in elevation or sea salt with distances inland. The basic geometry of a tropospheric source, whether it be from the oceans or from lightning, will be the same over Antarctica as air parcels moving inland will be uplifted and will precipitate. If the nitrate source were strictly tropospheric, a similar relationship should be found. Instead, in their Fig. 4 there is a generally linear increase in nitrate concentration poleward that cannot be attributed solely to less precipitation and the dilution/concentration effect. We find it inconceivable that the sodium and nitrate profiles would look so different if both had sources largely in the troposphere. Even tropospheric nitrate from high altitude lightning will have difficulty reaching the southern continent, because this species is removed rapidly in precipitation. However, stratospheric NO_x will have no such problems over Antarctica as its mean meridional motion is southward and downward due to the polar stratospheric vortex. In addition, NO_x is probably precipitated directly from the stratosphere as HNO_3 in ice crystals.

Finally, Legrand and Delmas regarded Jackman et al.'s estimate of NO_x production from SPE's as an upper limit because of spreading towards lower latitudes. We point out, however, that the polar stratospheric vortex mean meridional circulation opposes equatorward spreading at high altitudes and the circumpolar jet effectively reduces horizontal diffusion. In addition, their estimates using SCR's and GCR's were all based on production rates of 1.25 NO molecules per ion pair created. This number does not reflect N_2^+-produced NO, because below 80 km, N_2^+ reacts to form water clusters and is removed (Jackman et al., 1979). For our calculations, we are interested not in NO available for further reactions in the atmosphere, but in total NO_x (and hence NO_3) produced and available for deposition, including that in water clusters. Hence production rates of 2.5 NO per ion pair are probably a better estimate. Finally, SPE's are measured indirectly by ground-based riometers. Our estimates of NO_x production from SCR's and GCR's as measured by the IMP satellite series indicate that Jackman et al. are low by at least a factor of three (Laird, 1986). For these reasons, we do not believe that their numbers can be regarded as an upper limit but, rather, as probably low.

In conclusion, there is strong evidence in support of a dominant stratospheric source of nitrate in South Pole snow, and we believe the major component of this source can be attributed to solar activity in general and ionization from SCR's in particular. We were very surprised that the data presented by Legrand and Delmas in refutation of a solar contribution lack the time intervals of greatest relevance. On many of their points, there is no disagreement. In fact, we have used many of the same articles and lines of reasoning in the past to assess the various nitrate sources (e.g., Laird et al., 1982; Laird, 1986; Zeller et al., 1986). Where we differ most is in the interpretation of the data from these publications and in the conclusions.

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