HMTSF-TCNQ is a quasi-one-dimensional (1D) conductor, with a metal-insulator transition due to the CDW formation at 30 K at ambient pressure. We have carried out $^{77}$Se and $^{13}$C-NMR measurements and found that the spin susceptibility on the donor HMTSF chain was already suppressed below 100K because of a large pseudo gap ~230 K opening and that the 30 K transition takes place mainly on the TCNQ chain. At high temperatures, the DC susceptibility, measured with a SQUID magnetometer, is well explained as the sum of individual spin susceptibility on both chains, while a large diamagnetism appears below 100K and becomes the largest below 30 K, where the electronic density of states disappears completely. This anomalous diamagnetism was reported earlier and explained as due to the Landau-Peierls mechanism on the 2D pocket formed by the reconstruction of quasi-1D Fermi surface. We propose another explanation of the diamagnetism on the basis of the inter-band effect on orbital diamagnetism, as discussed theoretically.