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The infinitesimal generators of Lévy processes in Euclidean space are pseudo-differential operators with symbols given by the Lévy-Khintchine formula. This classical result relies heavily on Fourier theory which in the case when the state space is a Lie group involves abstract harmonic analysis requiring a formulation of the dual group of unitary irreducible representations. The notion of pseudo-differential operators can be extended to connected, simply connected nilpotent Lie groups by employing the Weyl functional calculus. In 2013 we demonstrated that the generators of Lévy processes in the 4 dimensional Engel group G_E are pseudo-differential operators which admit $C_c(G_E)$ as its core. Here we present an extension of these results to the 5 dimensional step 3 nilpotent Cartan group. (Received September 25, 2018)