Chronic and acute dietary exposure to pyrrolizidine alkaloids (PAs) was estimated in the European population via the consumption of plant-derived foods. This resulted in highest estimates of mean chronic dietary exposure of 34.5–48.4 ng/kg body weight (bw) per day in ‘Toddlers’ (LB–UB) and 154–214 ng/kg bw per day in the highly exposed population (LB–UB, also in ‘Toddlers’). Following a rather conservative scenario, the highest estimates of acute mean exposure and 95th percentile exposure were calculated for ‘Toddlers’, with mean exposure up to 311 ng/kg bw per day and 95th percentile exposure up to 821 ng/kg bw per day. Tea and herbal infusions were by far the main average contributors to the total exposure to PAs. Among consumers only, in the adult population, the mean chronic exposure via the consumption of honey ranged between 0.1 and 7.4 ng/kg bw per day (minimum LB–maximum UB), while for high consumers, it was between 0.4 and 18 ng/kg bw per day (minimum LB–maximum UB). In the young population, for the average consumers of honey, estimates were between 0.3 and 27 ng/kg bw per day (minimum LB–maximum UB), and between 0.7 and 31 ng/kg bw per day (minimum LB–maximum UB) among the high consumers. Ad hoc exposure scenarios for food supplements via consumption of pollen-based supplements showed chronic exposure to PAs that ranged between 0.7 and 12 ng/kg bw per day (minimum LB–maximum UB), while acute exposure was between 2.8 and 44 ng/kg bw per day (minimum LB–maximum UB), in both cases among consumers only. Likewise, the consumption of 150 mL infusion of 2 g of selected plant extracts led to exposures to PAs up to 67,000 ng/kg bw per day (e.g. infusion of Borage).