The spring 2010 Eyjafjallajökull (Iceland) eruption strikingly underlined the vulnerability of our globalized society to the atmospheric dispersal of volcanic clouds. Ash aggregation controls volcanic clouds dispersal by causing fine particles to settle prematurely from the cloud. Despite physical parameters of ash aggregates have been modeled and derived from ash fallout deposits of past eruptions, aggregates sedimentation still eluded direct measurement. Here we join field-based, high-speed-video analysis with laboratory experiments to provide the first in situ investigation and parameterization of the physical features and settling dynamics of ash aggregates from a volcanic cloud. On May 2010 we obtained high-speed video footages of both ash particles and aggregates settling from the Eyjafjallajökull volcano eruption cloud at a distance of 7 km from the vent, simultaneously sampling fallout products. Experimental laboratory determinations of the density, morphology, and settling velocity of individual ash particles enable their distinction from aggregates. The combination of field and experimental analyses allows a full characterization of the size, settling velocity, drag coefficient, and density distributions of ash aggregates as well as the size distribution of their component particles. We conclude that ash aggregation resulted in a tenfold increase in mass sedimentation rate from the cloud, aggravating the ash hazard locally and modifying cloud dispersal regionally. This study provides a valuable tool for monitoring explosive eruptions, capable of providing robust input parameters for models of cloud dispersal and consequent hazard forecast.