Supplementary Method 1

Acute asthma models
Female B6.Cg-Fcεr1a<sup>tm1Knt</sup>Tg(FCER1A)1Bhk/J mice were sensitized via a 200-μL intraperitoneal injection of 20 μg ovalbumin (OVA; Sigma-Aldrich, St. Louis, MO, USA) with 2 mg aluminum hydroxide (Pierce, Thermo Scientific; OVA-alum), on days 1 and 14. The mice were subjected to intranasal challenge with 1% OVA in saline on days 28–30. NPB311 or omalizumab was administered intraperitoneally on day 27 and 1 h before each OVA challenge. Negative control mice received a phosphate-buffered saline (PBS) challenge instead of OVA.

Atopic dermatitis models
The back skin of B6.Cg-Fcεr1a<sup>tm1Knt</sup>Tg(FCER1A)1Bhk/J mice was shaved and tape stripped six times with 3M tape. A 100-μg portion of OVA in 100 μL of normal saline or 100 μL of saline alone was placed on a 1×1 cm patch of sterile gauze. Each mouse had a total of three 1-week exposures to the patch at the same site; exposures were separated by 2-week intervals. Mice were given the indicated concentration of NPB311 (1, 5, and 10 mg/kg, intraperitoneal injection) or dexamethasone (0.01%, epicutaneous administration) daily on day 47–49. Skin thickness was measured in micrometers.

Inflammatory cell counts in bronchoalveolar lavage fluid
After the last challenge, the lung was lavaged with 2×0.9 mL of PBS. The lavage fluids were centrifuged, bronchoalveolar lavage fluid (BALF) cells were isolated, and the supernatant was frozen for later analysis. The BALF cell pellets were resuspended in PBS, and total BALF leukocyte counts were determined for each mouse via light microscopy using trypan blue exclusion. The BALF cells were centrifuged onto slides using a Cytospin centrifuge (Shandon Cytospin 4 cytocentrifuge; Thermo Fisher Scientific, Waltham, MA, USA). Differential cell counts were determined by staining the air-dried and fixed cytospin slides with Diff-Quick staining reagent (Merck, Darmstadt, Germany).

Measurement of IgE
The serum total immunoglobulin E (IgE) level from blood collected via cardiac puncture was measured using a mouse IgE ELISA kit (BD Bioscience, San Diego, CA, USA) according to the manufacturer’s instructions. Microplates were coated with anti-IgE antibody in coating buffer. Serum samples and diluted standards were added, and detection antibody and SAv-HRP-conjugated streptavidin reagent were then applied onto the plate for 2 h. The reaction was developed with TMB, and the plates were read at 450 nm.