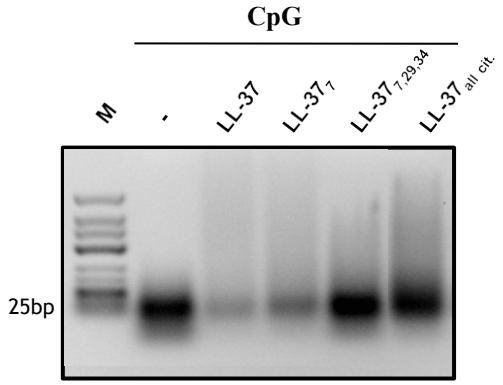
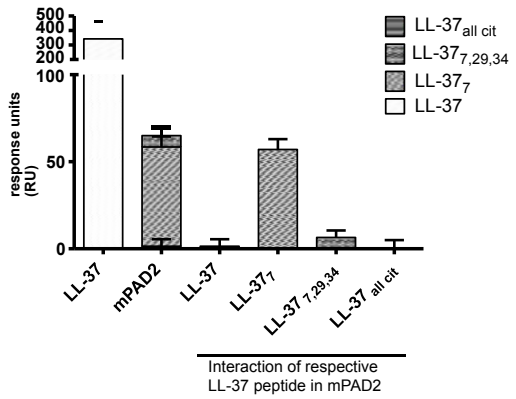
A.



B.

Peptide	mPAD2	mPAD4
LL-37	1%	49%
LL-37$_7$	77.5%	35%
LL-37$_{7,29,34}$	20%	14%
LL-37$_{All\ Cit.}$	1.5%	2%

C.



D.

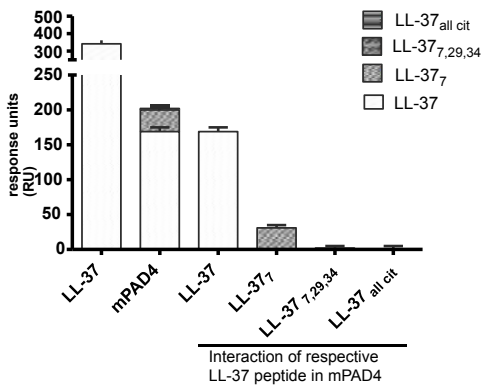


Figure S1. The efficiency of DNA binding by synthetic native and differentially citrullinated forms of LL-37 (LL-37$_7$, LL-37$_{7,29}$, LL-37$_{7,29,34}$, and fully citrullinated LL-37) estimated with gel retardation assay and binding preference of DNA fragment with synthetic native or modified forms of LL-37 in mPAD2/mPAD4.

(**A**)Synthetic oligonucleotide (CpG) was mixed with either synthetic native and modified forms of LL-37 at 1:5 (DNA:peptide) molar ratio and incubated at room temperature for 10 min. The complex formation was determined by running the complexes on 1.5% agarose gel. The composition of mixtures (mPAD2 & mPAD4) was established based on amino acid sequence analysis of LL-37 using automated Edman degradation (**B**). Surface plasmon resonance characteristics of the interaction between the mixtures of LL-37 analog with the DNA. To determine the interaction of the peptide mixtures resembles the catalytic modification of LL-37 by PAD2 (mPAD2 - **C**) and PAD4 (mPAD4 - **D**) (the mixture composition see Table - **B**) with DNA, the maximal binding levels (Rmax) of the mixtures were determined in the concentration range from 50 nM to 5 μM. The Rmax values of the mixtures were compared with the quota determined separately for the mix components at the same concentration level. Response units (RU) were used to describe the amounts of coupled DNA.

A.

B.

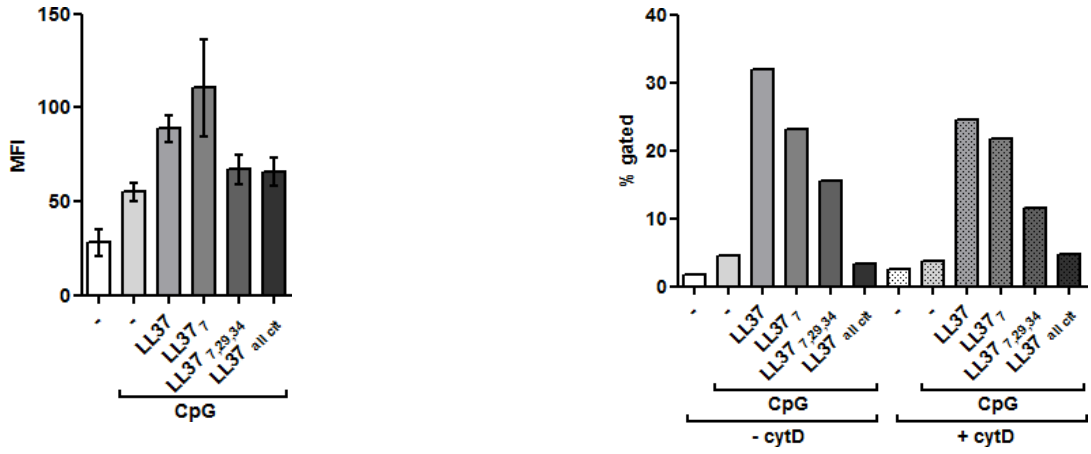


Figure S2. The efficiency of the CpG binding and uptake accompanied by LL-37 or its modified forms was determined by flow cytometry analysis.
Isolated pDCs were incubated with FiTC-CpG alone (final concentration 3μM) or FiTC-CpG with synthetic native and modified forms of LL-37 (final concentration 28.8 μg/mL) for 15 mins before the cells were washed with PBS for 3 times and analysed by FACS (**A**). Deposition of the cell membrane was established by preincubation of pDCs with Cytochalasin D for 15 min (**B**). Data are represented as mean fluorescent intensity (MFI) of gated cells or percentage of FiTC positive cells, respectively.

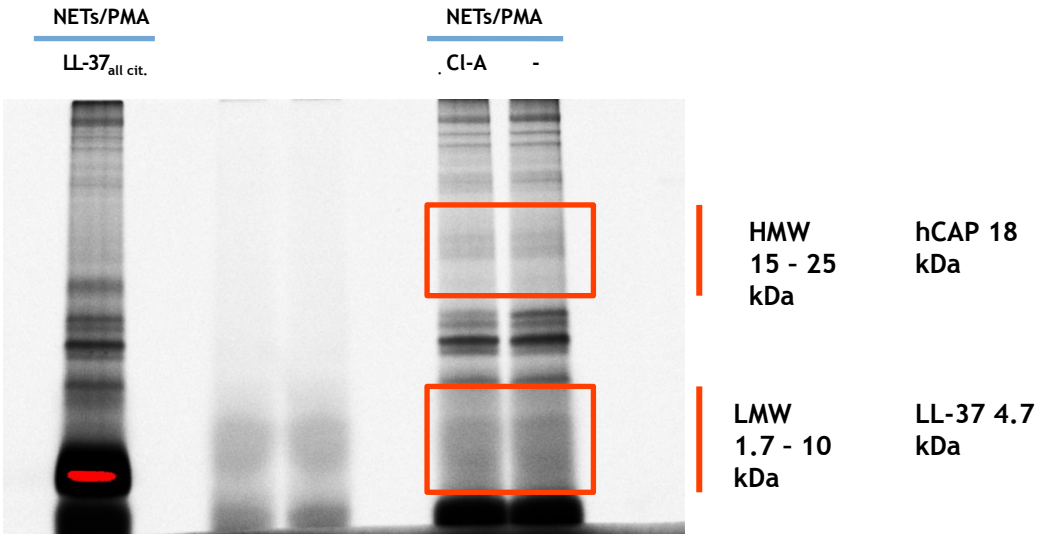
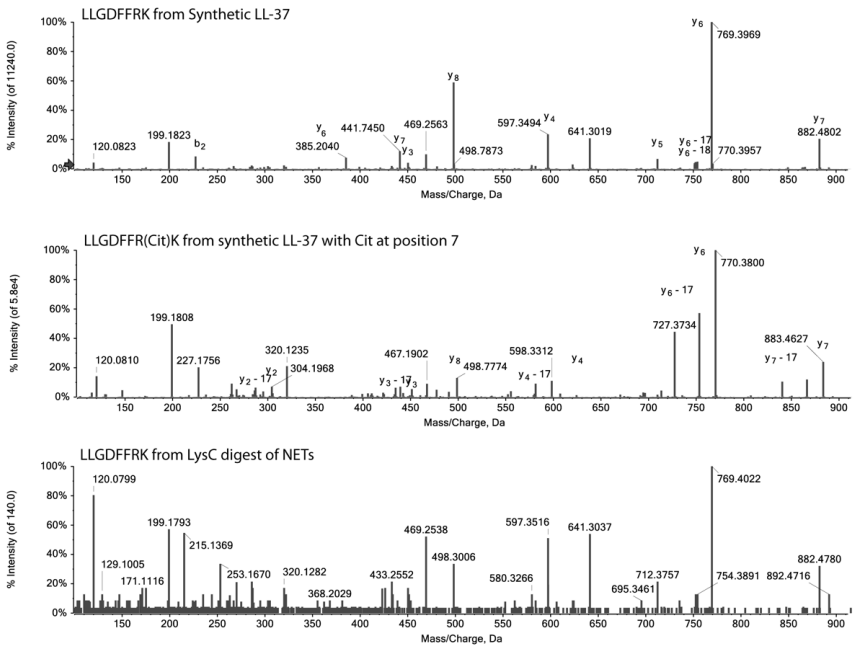


Figure S3. The profile of citrullinated proteins in PMA induced NETs in the presence (Cl-A) and absence (-) of PAD inhibitor visualized by the rhodamine-phenylglyoxal (Rh-PG) labelled probe.
NETs supplemented with synthetic fully citrullinated LL-37 ($LL\text{-}37_{all\ cit}$) was used as the control of citrullinated LL-37 migration in the gel – field in bracket.

A.



B.

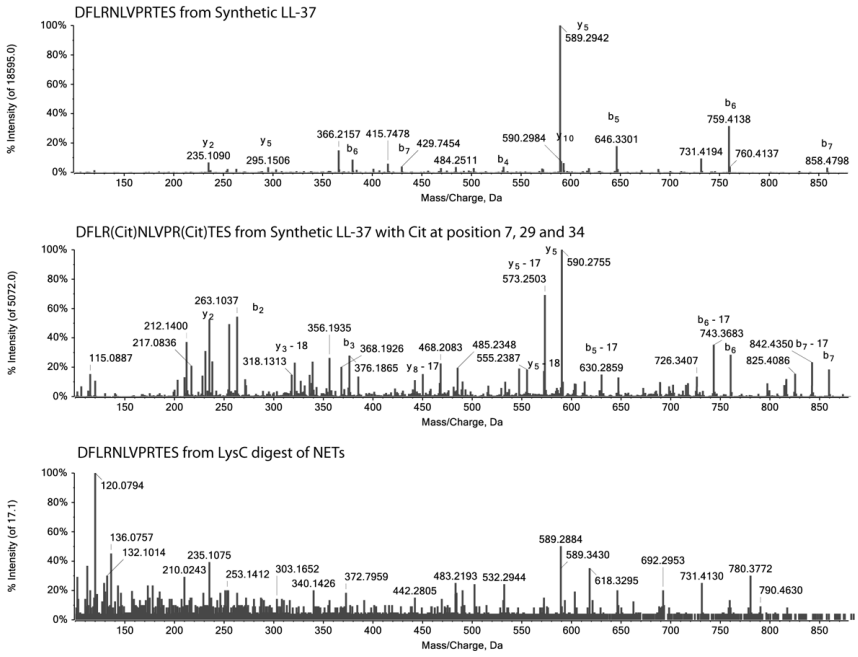


Figure S4. MSMS data of LysC generated N- and C-terminal peptide of LL-37. Three different variants of synthetic LL-37 were analyzed. Unmodified, Citrullinated at R7 or all Arg residues citrullinated.

(**A**) MSMS spectra of the N-terminal peptide LLGDFFRK in unmodified (Top) and modified version (middle). In a NETs sample digested with LysC the unmodified version was detected. (**B**) MSMS spectra of the C-terminal peptide DFLRNLVRPTES in unmodified (Top) and modified version (middle). In a NETs sample digested with LysC a weak signal of the unmodified version was detected. These data shows the additional internal fragmentation and neutral loss associated with citrullination.