Supplementary Information

Highly stable and efficient α-phase FA-based perovskite solar cells prepared in ambient air by strategically enhancing the interaction between ions in crystal lattices

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Figure S1. PL intensity of 0 MASCN perovskite film fabricated in glovebox.



Figure S2. XRD pattern of 0 MASCN perovskite film fabricated in glovebox. The δ -phase is not observed in this condition, confirming that water molecular in ambient is the main reason for the formation of δ phase.



Figure S3. Differential scanning calorimetry of MASCN powder.



Figure S4, (a)-(e) Tauc plots of perovskite films with the increase of MASCN from 0, 25, 50, 75, and 100 mol% respectively.



Figure S5 (a), (b) and (c) Top view SEM images of perovskite prepared with 25 MASCN, 75 MASCN and 100 MASCN, respectively. (d) The calculated size of perovskite grains derived from 0-100 MASCN.



Figure S6 J-V characteristics of the best PSC in forward and reverse scans.



Figure S7 Statistical box figures of (a) *PCE*, (b) *Voc*, (c) *Jsc*, (d) *FF* of the devices fabricated with different amounts of MASCN.



| RH | PCE (%) | Voc (V) | <i>Jsc</i> (mA cm ⁻²) | FF (%) |
|----------|---------|---------|-----------------------------------|--------|
| Glovebox | 13.67 | 0.95 | 22.41 | 63.96 |
| 30±5% | 16.63 | 1.03 | 22.18 | 72.58 |
| 50±5% | 18.07 | 1.04 | 23.34 | 74.11 |
| 70±5% | 18.01 | 1.02 | 24.04 | 73.31 |
| 90±5% | 16.75 | 0.98 | 23.58 | 72.05 |

Figure S8 *J*-*V* characteristics of the 50 MASCN PSCs fabricated in different conditions in forward scan.



Figure S9 Surface temperature of the resulting devices under one sun (100 mW cm⁻²) continue illumination without optical filter.



Figure S10 *J-V* curves of the resulting devices before and after light soaking at one sun illustration (100 mW cm⁻²). (a) with MASCN, and (b) without MASCN.

| sample | A ₁ | τ_1 | A ₂ | τ ₂ | | | |
|----------|----------------|----------|----------------|----------------|--|--|--|
| | | (ns) | | (ns) | | | |
| 50 MASCN | 0.43 | 38.09 | 0.57 | 179.76 | | | |
| 0 MASCN | 0.95 | 2.68 | 0.05 | 7.16 | | | |
| | | | | | | | |

Table S1. Fitting data of the TRPL of the perovskite with 0 MASCN and 50 MASCN.