## **Supplemental Methods**

## Purification of 3'-hydroxy-ε,ε-caroten-3-one, 3-hydroxy-β,ε-caroten-3'-one, and ε,ε-carotene-3,3'-dione

The extract from the reaction mixture with lutein was dissolved in dichloromethane and passed through a silica column equilibrated with dichloromethane. The eluate was developed into 3'-hydroxy- $\varepsilon$ , $\varepsilon$ -caroten-3-one and  $\varepsilon$ , $\varepsilon$ -carotene-3,3'dione on a silica-TLC plate using dichloromethane:ethyl acetate (80:20, v/v) as the solvent system. The oxidation products recovered from the TLC band were finally purified by reverse-phase HPLC on a Wakopak Navi C30-5 column (100 x 250 mm; Wako Pure Industries, Osaka, Japan), with methanol containing 0.05% ammonium acetate as a mobile phase. 3-Hydroxy- $\beta$ , $\varepsilon$ -caroten-3'-one and  $\varepsilon$ , $\varepsilon$ -carotene-3,3'-dione were also obtained from the reaction mixture with zeaxanthin in the same manner as described above.

## Purification of 3'-hydroxy-β,ε-caroten-3-one

The extract from the reaction mixture with lutein was dissolved in dichloromethane and developed on a silica-TLC plate using dichloromethane:ethyl acetate (80:20, v/v) as the solvent system. The oxidation product recovered from the TLC band was purified by normal-phase HPLC on a cyanopropyl column of Inertsil CN-3 (10 × 250 mm; GL Sciences, Tokyo, Japan) attached to an Inertsil CN-3 guard column (7.6 × 30 mm) with the following mobile phase at a flow rate of 5 mL/min: *n*hexane:dichloromethane:methanol (85:15:0.15, by vol.). The eluate was finally purified by reverse-phase HPLC on a Wakopak Navi C30-5 column (100 x 250 mm) with methanol:ethyl aceate (94:6, v/v) as a mobile phase.

## Purification of β,β-caroten-3-one and β,ε-caroten-3'-one

The extract from the reaction mixture with  $\beta$ -cryptoxanthin was developed on a silica-TLC plate using dichloromethane:ethyl acetate (95:5, v/v) for  $\beta$ , $\beta$ -caroten-3-one and dichloromethane:ethyl acetate (90:10, v/v) for  $\beta$ , $\epsilon$ -caroten-3'-one as the solvent system. The oxidation products recovered from the TLC band were purified by reverse-phase HPLC on a Wakopak Navi C30-5 column (100 x 250 mm) with methanol:ethyl acetate (70:30, v/v) containing 0.05% ammonium acetate as a mobile phase.