Figure S1: Average ΔMAC values observed after evaporation/redissolution of SOA+AS solutions at different rotary evaporator bath temperatures. Despite the weak dependence of the evaporation-induced increase in *MAC* on the evaporation temperature, we conducted all of the experiments at 50°C in order to accelerate the evaporation process. Based on this dependence, we can expect that evaporation of room temperature samples would result in ~30% higher *MAC* relative to the values reported in the main manuscript.

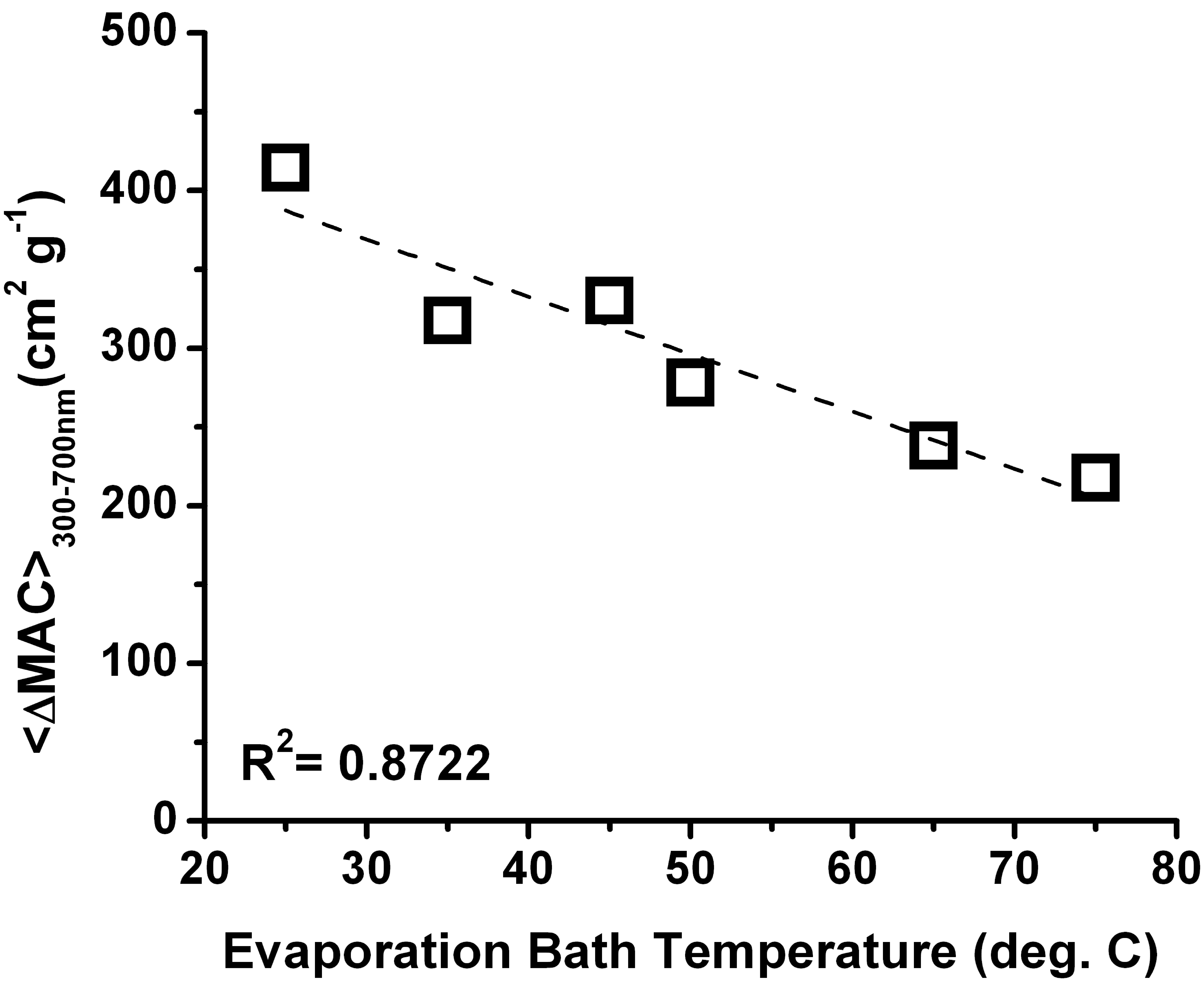

Figure S2: Absorption spectra of the LSOA+AS solution after evaporation/redissolution recorded as a function of time after the redissolution step. This experiment demonstrates that the chromophore is stable with respect to the hydrolysis on the time scale of hours. If anything, the absorption slightly increased with time indicating that the reaction was still going on after the redissolution. (For the results reported in the main text, we typically took the UV/Vis spectra within minutes after the redissolution). The stability of the chromophores is significant as carbonyl-imine equlibria in water containing ammonia are shifted towards carbonyls, and the imine products are generally not stable with respect to hydrolysis, especially under acidic conditions. The nitrogen-containing products become stable with respect to hydrolysis only when nitrogen becomes part of a heterocycle.

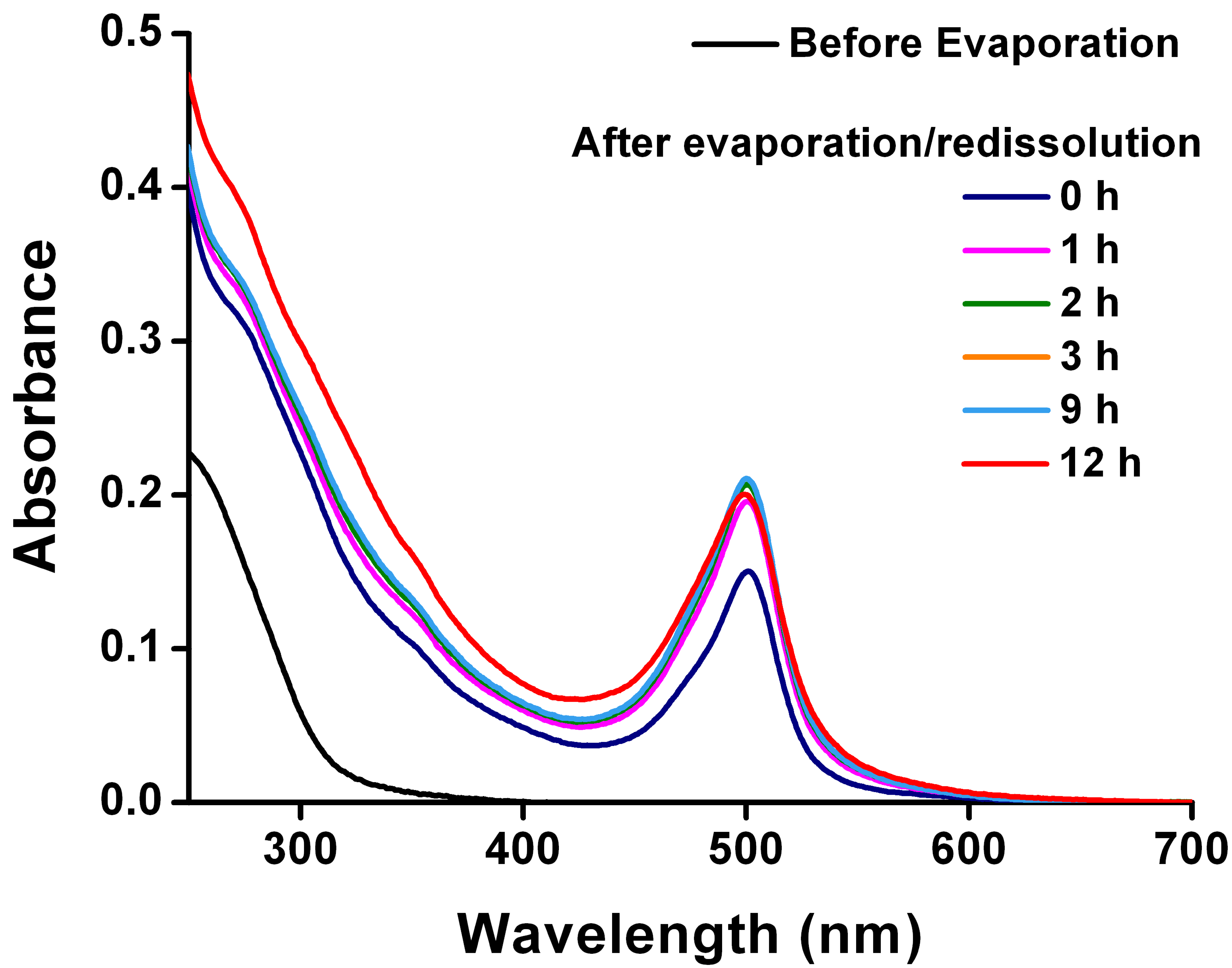

Figure S3: Absorption spectra of the SOA extract before and after evaporation in the presence of ammonium sulfate and the simplest amino acid glycine. The shift in the position of the 500 nm band provides indirect evidence of incorporation of nitrogen in the chromophores' structures.

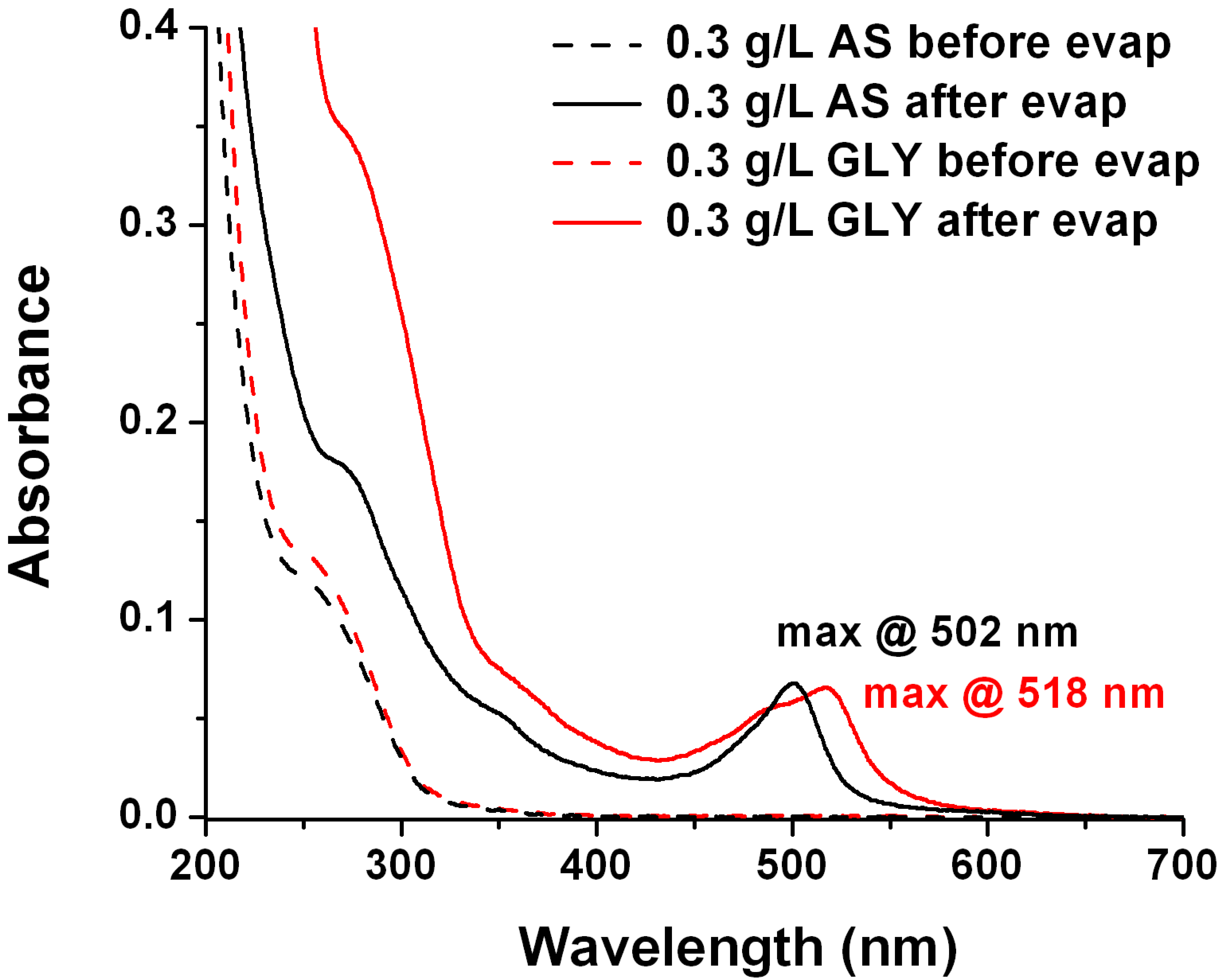

Figure S4: A photograph of the evaporated/redissolved samples of AS+SOA obtained at different SOA:AS mass concentration ratios (values stated on the vials). The corresponding absorption spectra are shown in Figure 2 in the main text. The left-hand vial corresponds to the evaporated/redissolved solution of SOA without any AS added (no color change). The right-hand vial corresponds to the evaporated/redissolved solution of SOA mixed with glycine; the corresponding spectrum is shown in Figure S3.

Figure S5. UV/Vis spectra of samples obtained by evaporations of SOA + sulfuric acid solution with the initial pH =2 and redissolving the red-brown residue in either water (H_2O), acetonitrile (ACN), or tetrahydrofuran (THF). The absorbance was converted into *MAC* using Eq. (1) in the main text. The evaporation residue could not be completely redissolved in water, and this is reflected in the smaller *MAC* of the reconstituted H_2O solution. THF and ACN dissolved the residue more completely, resulting in larger *MAC* values. However, it is likely that the dissolution was still incomplete based on the significant difference in the wavelength dependence of *MAC* in ACN and THF. This behavior can be contrasted with the SOA+AS case, where the residue that had formed during evaporation could be fully redissolved in water.

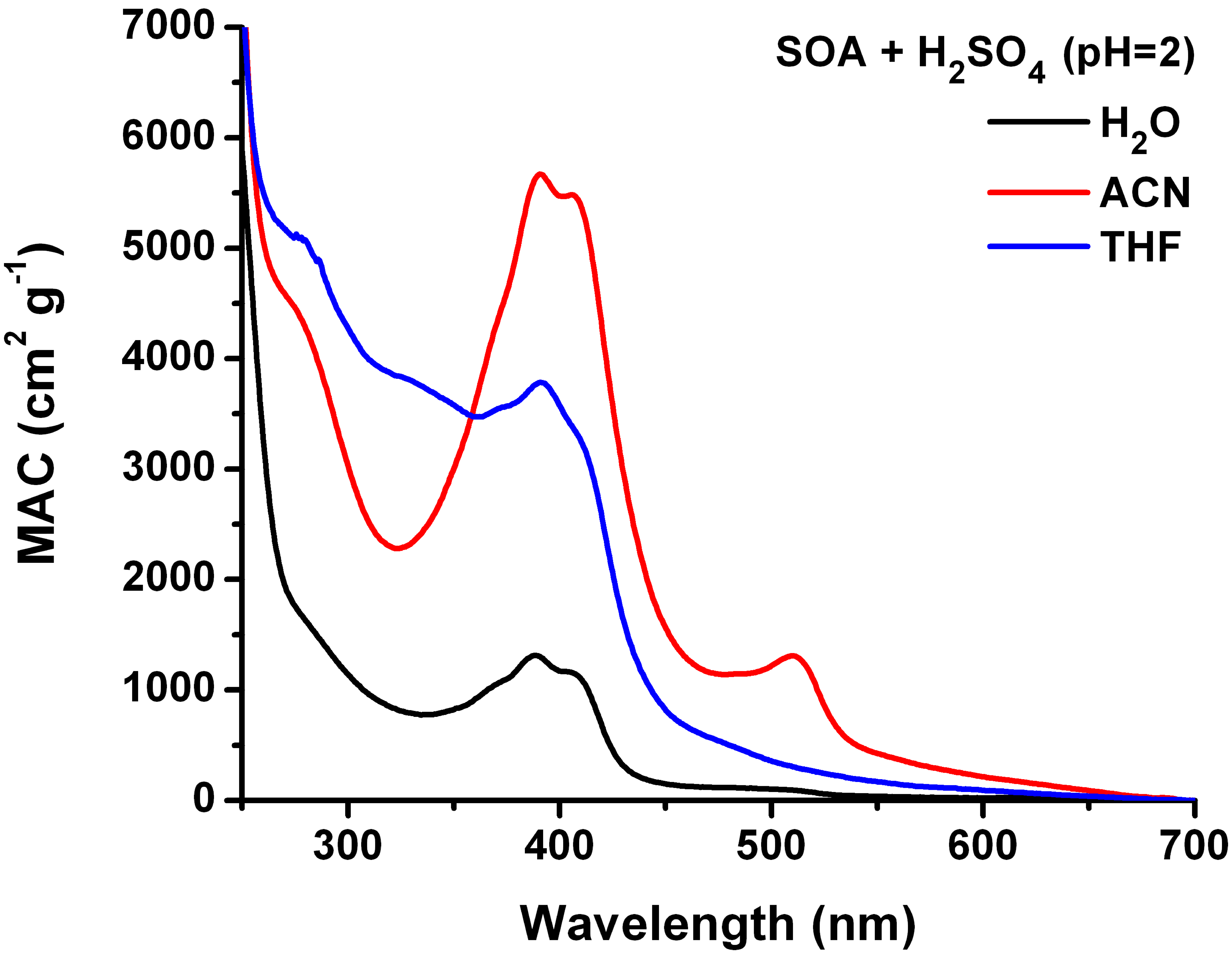

Figure S6: Absorption spectra of the SOA + sulfuric acid solution (initial pH = 2) after evaporation/redissolution in water, recorded as a function of the time after the redissolution. This figure proves that the water-soluble fraction of the chromophores is stable with respect to the hydrolysis. Just like in the case with the SOA+AS evaporated/redissolved samples (see Fig. S2), the absorption continues to increase slowly following the redissolution, suggesting that secondary chemistry is still taking place.

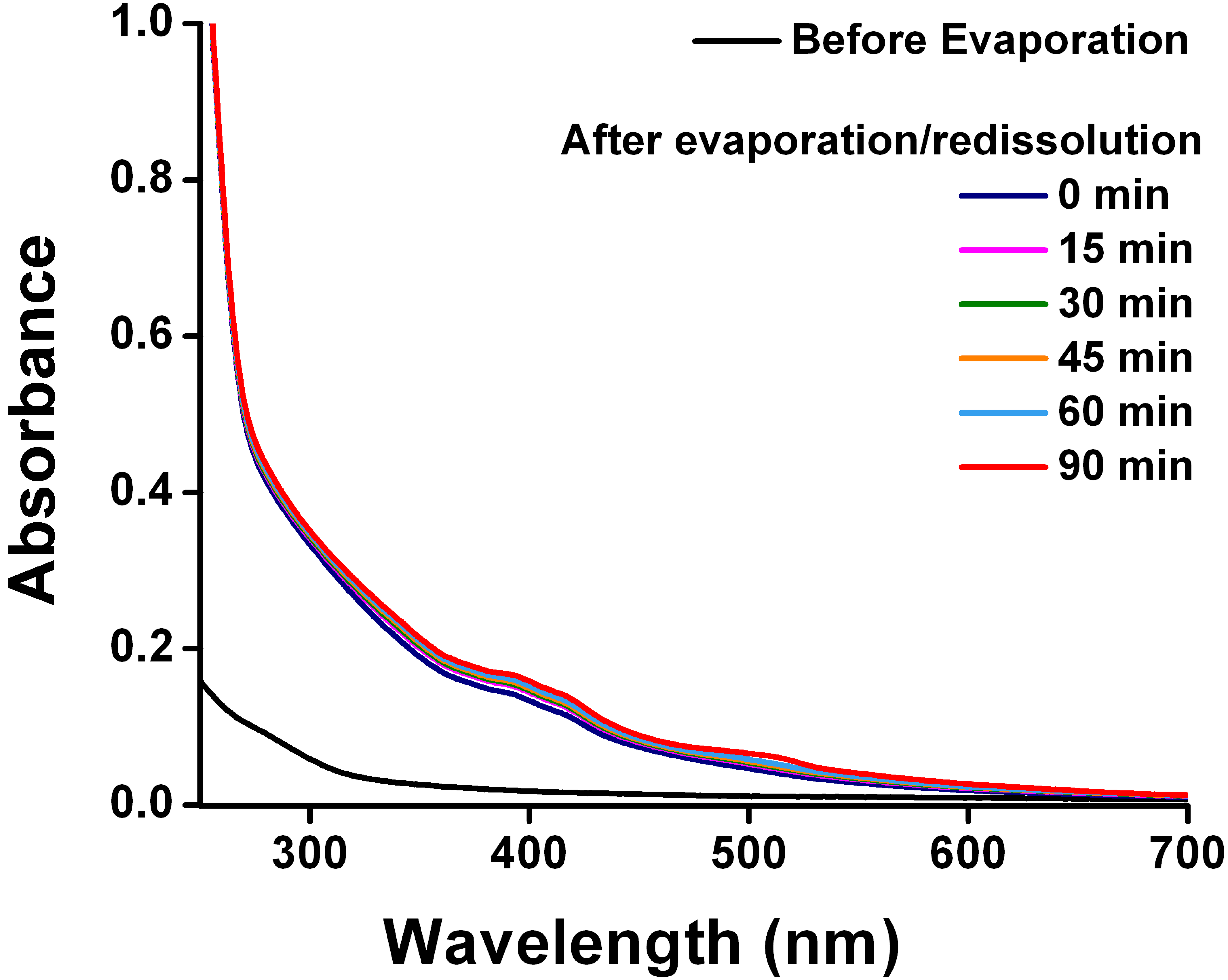

Figure S7. UV/Vis spectra of samples obtained by evaporations of aqueous solutions SOA + sulfuric acid and SOA + methanesulfonic acid with the initial pH =2 and redissolving the residue in acetonitrile (ACN), which is a better solvent, according to Figure S5.. Sulfuric and methanesulfonic acid produce very similar peaks but they are smaller in the methanesulfonic acid case, and the peaks are slightly shifted. We take it as an indication that the $–OS(O)_2OH$ and $–OS(O)_2CH_3$ groups modulate both the solubility and the optical properties of the chromophores.

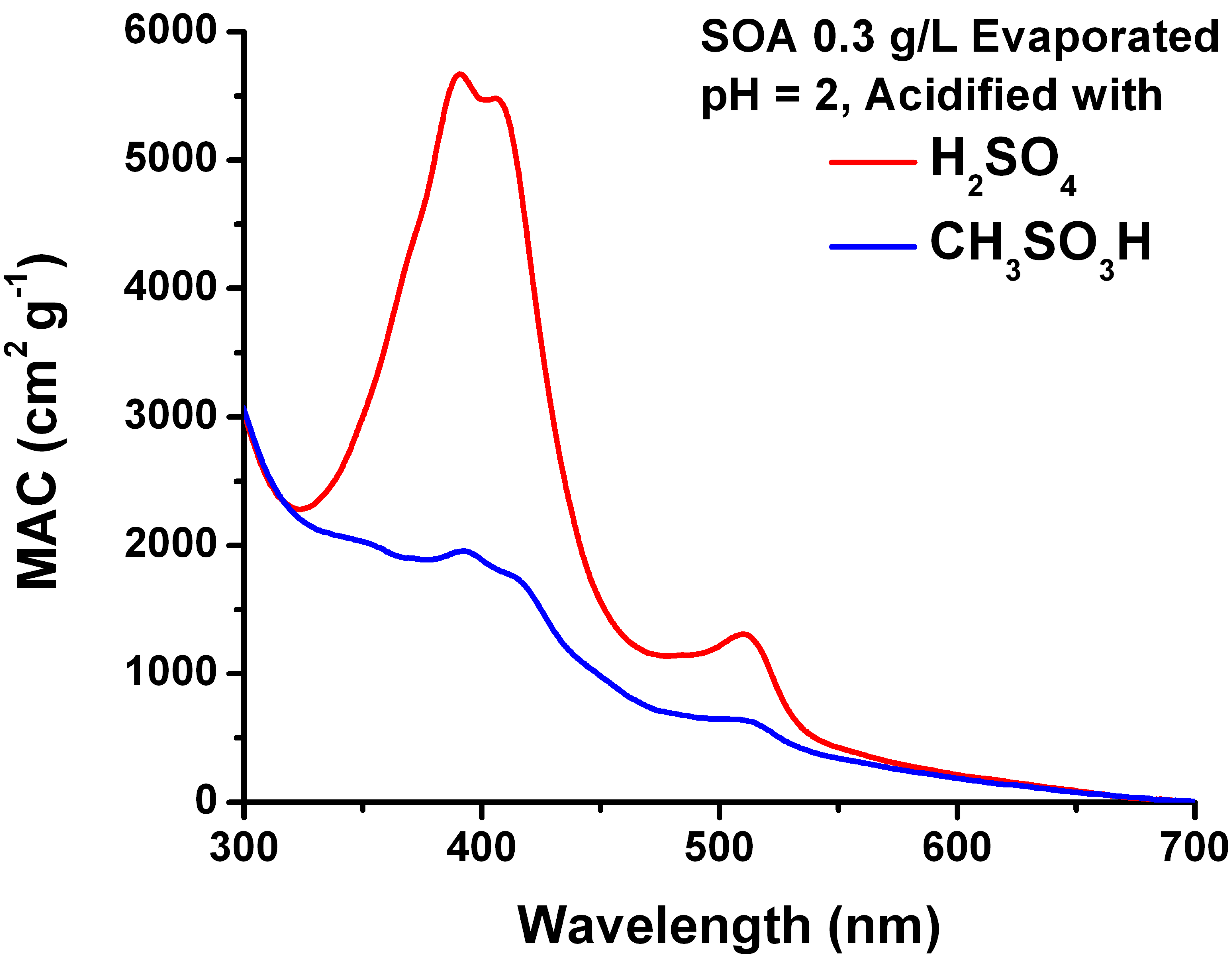

Table S1. The list of nitrogen containing ions detected by positive ion mode ESI-MS in the SOA + ammonium sulfate solutions before and after evaporation/redissolution. The formulas for the corresponding neutral compounds can be obtained from the positive ions by subtracting one hydrogen atom.

SOA , nothing added, evaporated

m/z	C	H	O	N
352.2816	19	39	3	1
494.2087	17	35	15	1

SOA + AS, evaporated

m/z	C	H	O	N
360.1413	17	23	6	1
390.1604	13	27	12	1
392.1522	14	27	10	1
448.2017	16	33	13	1
450.1822	15	31	14	1
460.2026	17	33	13	1
462.2174	17	35	13	1
464.1971	16	33	14	1
478.2135	17	35	14	1
480.1921	16	33	15	1
484.2532	24	37	9	1
488.1975	18	33	14	1
490.2134	18	35	14	1
492.2290	18	37	14	1
506.2078	18	35	15	1
506.2438	19	39	14	1
508.2235	18	37	15	1
520.2600	20	41	14	1
522.2388	19	39	15	1
532.2593	21	41	14	1
534.2390	20	39	15	1

SOA + AS, evaporated (continued)

m/z	C	H	O	N
536.2550	20	41	15	1
538.2342	19	39	16	1
548.2549	21	41	15	1
548.2910	22	45	14	1
550.1763	22	31	15	1
550.2341	20	39	16	1
550.2709	21	43	15	1
499.2316	19	37	17	1
552.2128	20	41	16	1
552.2497	10	16	7	2
277.1031	21	24	10	2
465.1504	20	40	9	2
475.2621	19	32	13	2
497.1977	20	32	14	2

Table S2. The list of sulfur containing ions detected by negative ion mode ESI-MS in the SOA + sulfuric acid solutions before and after evaporation/redissolution. The formulas for the corresponding neutral compounds can be obtained from the negative ions by adding one hydrogen atom.

SOA + H_2SO_4 (pH = 4)				
m/z	C	H	O	S
285.0285	8	13	9	1
299.0441	9	15	9	1
315.0389	9	15	10	1
455.1234	17	27	12	1
467.1230	18	27	12	1
469.1385	18	29	12	1
471.1177	17	27	13	1
483.1537	19	31	12	1
485.1330	18	29	13	1
499.1489	19	31	13	1
501.1279	18	29	14	1

SOA + H_2SO_4 (pH = 4), evaporated				
m/z	C	H	O	S
251.0594	9	15	6	1
265.0387	9	13	7	1
267.0543	9	15	7	1
279.0542	10	15	7	1
281.0335	9	13	8	1
281.0698	10	17	7	1
285.0285	8	13	9	1
295.0493	10	15	8	1
297.0648	10	17	8	1
299.0441	9	15	9	1
311.1686	17	27	3	1
439.1279	17	27	11	1
451.1275	18	27	11	1
467.1230	18	27	12	1
469.1385	18	29	12	1
481.1385	19	29	12	1
485.1330	18	29	13	1

SOA + H_2SO_4 (pH = 2)				
m/z	C	H	O	S
271.0127	7	11	9	1
285.0284	8	13	9	1
299.0441	9	15	9	1
315.0389	9	15	10	1
327.0388	10	15	10	1
329.0546	10	17	10	1
331.0337	9	15	11	1
441.1072	16	25	12	1
455.1227	17	27	12	1
457.1030	16	25	13	1
459.1176	16	27	13	1
467.1233	18	27	12	1
469.1028	17	25	13	1
469.1387	18	29	12	1
471.1178	17	27	13	1
481.1394	19	29	12	1
483.1180	18	27	13	1
483.1547	19	31	12	1
485.1337	18	29	13	1
487.1126	17	27	14	1
487.1492	18	31	13	1
497.1341	19	29	13	1
499.1127	18	27	14	1
499.1488	19	31	13	1
501.1288	18	29	14	1
503.1446	18	31	14	1
513.1276	19	29	14	1
513.1653	20	33	13	1

SOA + H_2SO_4 (pH = 2), evaporated				
m/z	C	H	O	S
223.0280	7	11	6	1
225.0437	7	13	6	1
235.0280	8	11	6	1
237.0436	8	13	6	1
239.0229	7	11	7	1
249.0438	9	13	6	1
251.0229	8	11	7	1
251.0593	9	15	6	1
253.0386	8	13	7	1
263.0231	9	11	7	1
263.0593	10	15	6	1
265.0385	9	13	7	1
267.0179	8	11	8	1
267.0543	9	15	7	1
277.0387	10	13	7	1
279.0543	10	15	7	1
281.0335	9	13	8	1
285.0284	8	13	9	1
293.0337	10	13	8	1
295.0491	10	15	8	1
297.0285	9	13	9	1
299.0441	9	15	9	1
311.0441	10	15	9	1
313.0233	9	13	10	1
327.0388	10	15	10	1
377.0218	10	17	11	2
423.0961	16	23	11	1
463.1280	19	27	11	1
465.1075	18	25	12	1
467.1233	18	27	12	1
469.1028	17	25	13	1