This work presents a valuable yearlong ToFACSM dataset for organic and inorganic source apportionment in Athens. It provides valuable information for the source of organic and inorganic source in Athens. The manuscript is generally well written, and falls well within the scope of AMT. Thus, it is of value to be published. After said that, some modifications are necessary to clarify what values the work add.

Major comments:

This manuscript exhibits valuable measurements for organic and inorganic source apportionment, while it lacks focus. It may be helpful to strengthen how this dataset is useful for environmental studies or climate studies. Besides, it would be better to provide some implications on what values this dataset adds compared to previous studies. In summary, the authors should provide more discussions to strengthen the values of the dataset. Besides just providing values, it is also valuable to clarify why these values are valuable.

Minor comments:

Abstract, The method has been proposed in previous studies, but you applied it in a yearlong investigation in Athens. Thus, "new methodology" is not proper.

Line 41. Brown carbon is also organic aerosol, while it absorbs light from ultraviolet to visible region.
Line 46. “Organic fraction usually comprises the greatest fraction of ambient aerosol”, is it correct for all the regions?

Line 48 “Secondary” should be “Secondary organic aerosols”.

Line 51 -52 “aerosol forming” should be “aerosol-forming”.

Line 53. “SOAs are the dominant form of organic aerosols”, is it really correct? Please provide the references.

Line 54 – 58, please provide references to verify your clarifications.

Please check the English grammar. Some examples are shown in the following.

Line 58 “in agriculture etc” should be “in agriculture, etc”

Line 60 “are also released in” should be “are also released into”.

Line 82 you said “Although source apportionment studies on organic aerosols for long periods have been prevailing in recent years covering a wide range of different sites, a long period of combined organic and inorganic source apportionment has not yet been published.”, but in line 70 you also say “Previous studies on particulate matter source apportionment in Greece have mainly focused on inorganic datasets”. I know you mean that the aims of this work is to combine organic and inorganic source apportionment for a yearlong investigation. However, the logic of the sentence should be re-arranged.

Line 82 – 84: This sentence is too long.

Line 85 – 86 : It would be better to re-write this sentence as two sentences. “one on the combined” should be “another on the combined”

Line 93 “member of” should be “a member of”. 
Line 96 “North east” should be “Northeast”.

Line 107 “afterwards” should be “afterward”.

Line 108 “principle of” should be “the principle of”.

Figure 3: the axis of different subfigures are too close.

Line 576: is “OA” “OOA”?

Line 596 – 599: This sentence is too long. In addition, in the discussion, some specific discussions on how the dataset adds should be added.