We are grateful to the referees for their insightful comments which helped to improve the manuscripts substantially. We provided point-by-point responses to the referee's comments below where our responses are in blue.

Referee 2:

The study of Lin et al. analyses the PM1 spatial and chemical variation in Ireland using ACSM and AE33 measurements. PM1 spatial variation is very important since a lot of sources are specific to different locations across Europe and are insufficient characterized. Chemical Online measurements offer the opportunity to assess with high accuracy the time evolution of atmospheric aerosol chemical composition. The paper is well-written, making extensive use of the available literature and the results are visualized in an appropriate way. New information is presented in the study related to the main PM1 sources in Ireland during wintertime.

Response: We thank the referees for the positive feedback. Below, we provide point-to-point response to the referee's comments.

As general remark I recommend that Mace Head measurements and discussion to be threated separately in another study since no data are available in the same time period and BC is assumed to be the constant between 2013 and 2016 wintertime, without scientific evidence.

Response: We agree that the assumption for the constant BC concentration between 2013 and 2016 lacks scientific evidence. In the original version, we tried to compare the BC source apportionment with AE-33 across Ireland. However, as both reviewers pointed out, large uncertainty was associated with the usage of the 2016 BC data to infer 2013 BC data in the original version. Therefore, to reduce confusion, we have replaced the AE-33 data in Jan 2016 with MAAP data in Jan 2013 in the revised version. Also in the revised version, we tend to keep the Mace Head data as part of this comparison study rather than being treated in another study because we believe the comparison of the OA source apportionment between Carnsore Point at the east coast and Mace Head at the west coast of Ireland are very insightful for the characterization of the spatial variation of NR-PM_1 and OA sources despite their measurements in different years.

Line 15-20 (pp1) Why average concentration for PM 1 in Dublin is comparable with average in Birr? (the value for Dublin is almost double than Birr).

Response: Corrected. It now reads, "Birr, a small town in the midlands area of Ireland with a population <1% of that in Dublin, showed an average PM_1 concentration (4.8 μg m^{-3}, ranging from <0.5 to 63.0 μg m^{-3} in December 2015) around half of that (56%) in Dublin"

Line 25-30 (pp3-4) For ACSM should be included all calibration coefficients determined during the campaign measurements, for all sites.

Response: We have added a supplementary Table for all the calibration coefficients for all sites in the revised text. In the text, "…Ionization efficiencies (IEs) and relative ionization efficiencies (RIEs) for sulfate and ammonium were determined through the calibration with ammonium nitrate and ammonium sulfate following the procedure described by Ng et al. (2011), and the IEs and RIEs at each site were provided in Table S1…"

Table S1. Ionization efficiencies (IE) and relative ionization efficiencies (RIE) obtained through the calibration with ammonium nitrate and ammonium sulfate.

	Dublin	Carnsore Point	Birr	Mace Head
IE	3.24e-11	3.28e-11	3.24e-11	4.47e-11
RIE (sulfate)	0.96	0.74	0.96	0.56
RIE (ammonium)	7.58	6.74	7.58	4.76

Line5-10 (pp 4) Is the CE 1 applied after comparison with SMPS for all sites during the same weather conditions?

Response: Actually, only the ACSM deployed in Dublin was compared with the co-located SMPS during the winter of 2016. Because the same ACSM instrument was also deployed in Birr (December 2015) and Mace Head (January 2013) during wintertime with similar weather conditions, CE of 1 was also applied to the ACSM dataset at these two sites. For the ACSM deployed at Carnsore Point, the same magnitude of increase in the concentrations of PM1 as that Dublin during 5-6 December 2016 (Fig. 5 and 6) confirmed the application of CE=1 was also physically meaningful at Carnsore Point.

We have clarified this in the revised text, it now reads, "For all ACSM measurements, a collection efficiency (CE) of 1 was applied for all the measured species. This CE was validated against a collocated scanning mobility particle sizer (SMPS) which shows the sum of the calculated ACSM volume and black carbon (BC) volume correlated well (r = 0.96 and slope = ~1) with the SMPS volume (size ranged from 14.6 nm to 685.4 nm) at the sampling site in Dublin during the winter of 2016 (Lin et al., 2018). Note that the same ACSM was also deployed in Birr during December 2015 and Mace Head during January 2013 under similar weather conditions and thus a CE of 1 was also applied for the datasets at these two sites. For the ACSM at Carnsore Point, the similar magnitude of increase in PM_1 in continental air masses (See Sect. 3.2) confirmed that the application of CE of 1 for the Carnsore Point dataset was physically meaningful. Also, note that a CE of 1 provided a lower limit for all ACSM-measured mass concentration."

Did you used SMPS volume concentration? What size range was used for SMPS set-up? Indeed, the CE did not affect the relative contribution of nonrefractory PM1, but if BC is included in PM1, the relative contribution of BC is dramatically modified, my suggestion is to argue more why CE 1 was chosen.

Response: Yes, the SMPS volume concentration was used with the size ranging from 14.6 nm to 685.4 nm. Also, see the response for the previous comment.

Chapter 3 (pp 5-6) Is not clear what are the final *a* values chosen and the correlation values with BC tracers for final solutions. Please clarify these.

Response: We have added more details on how the final *a* values were chosen and added correlation values for the BC tracers for the final solutions. In the revised text, it now reads, "High *a* values (e.g., 0.3-0.5) or a loose constraint led to potential mixing between these heating-related factors especially when their time series showed temporal co-variation (i.e., all showed higher concentration at night). At high *a* values, the mixing between these factors was evidenced by the sudden drop of correlation coefficient between the time series of the peat factor and BC_{wb} with R dropping from 0.82 at an *a* value of 0.3 to the R of 0.47 at an *a* value of 0.4 (Fig. S5) while the correlation between the corresponding profile of e.g., peat also

dropped from 0.96 to 0.90 confirming the mixing between peat and other factors (e.g., wood). In contrast, a lower *a* value (e.g., 0-0.2) reduced mixing and improved the separation by tightly constraining their individual profiles. As shown in Fig. S5, at an *a* value of 0.1, the time series of peat showed a good correlation with BCwb (R=0.88) while the profile of peat was also tightly correlated with the reference peat profile (R=0.99). Therefore, an *a* value of 0.1 was chosen as the most optimal ME-2 solution."

Fig. 5 insufficient explained e.g. the altitude of air masses, number of days used for the model
Response: We have now added more information regarding the HYSPLIT model. It now reads, "…The back trajectories (BTs) on the right panel was calculated using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT; Stein et al. (2015)). The BTs were calculated for an arrival height of 500 m at the length of 72 h, and were every 6 h during continental air masses during 5-6 December (top right) and every 12 h during marine air masses during 22-27 December (bottom right)."

References:
Lin, C., Huang, R.-J., Ceburnis, D., Buckley, P., Preissler, J., Wenger, J., Rinaldi, M., Facchini, M. C., O'Dowd, C., and Ovadnevaite, J.: Extreme air pollution from residential solid fuel burning, Nature Sustain., 2018.
Ng, N. L., Herndon, S. C., Trimborn, A., Canagaratna, M. R., Croteau, P. L., Onasch, T. B., Sueper, D., Worsnop, D. R., Zhang, Q., Sun, Y. L., and Jayne, J. T.: An Aerosol Chemical Speciation Monitor (ACSM) for routine monitoring of the composition and mass concentrations of ambient aerosol, Aerosol Sci. Technol., 45, 780-794, 2011.
Stein, A. F., Draxler, R. R., Rolph, G. D., Stunder, B. J. B., Cohen, M. D., and Ngan, F.: NOAA's HYSPLIT Atmospheric Transport and Dispersion Modeling System, Bull. Am. Meteorol. Soc., 96, 2059-2077, 10.1175/bams-d-14-00110.1, 2015.