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Classifying aerosol type using in situ surface spectral aerosol optical properties

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Discussion of individual cluster results from multivariate cluster analysis

Cluster 1 includes MLO, SUM, BRW, ALT and SPL and is characterized by stations with low aerosol loadings (small σ_{sp}), medium to small aerosol particles (1< SAE<2), and AAE values with a range of 0.5 < AAE < 1.5. All of the stations in Cluster 1 are either Arctic stations (BRW, ALT, and SUM) or remote mountaintop stations (MLO and SPL). The median optical properties of the cluster correspond with an aerosol type of 'small particles low absorption' or 'BC dominated' according to Cappa et al. (2016). On the one hand, the inferred aerosol of 'small particles low absorption' type might agree well with what aerosols would be expected at remote locations such as these, since a few of these sites might measure sea salt (though we would expect sea salt to be a large aerosol), and most aerosols to reach these sites are well aged and could be coated due to the aging time (Jacobson, 2001). An aerosol type of 'BC dominated' would also not be surprising at these sites, given the potential for local aerosol source contributions from biomass or fossil fuel burning, though the generally low scattering coefficients at these sites indicate the presence of anthropogenic aerosols is infrequent.

Cluster 2 includes AMY and GSN, the two coastal stations located in South Korea, and is characterized by high aerosol loadings (high σ_{sp}), small aerosol particles (SAE~1.5) and BC dominated aerosols (AAE~1.2). Existing aerosol classification schemes would designate the median optical property values of this cluster as fossil fuel burning aerosols (BC) and/or biomass burning aerosols (BrC) (Cazorla et al., 2013; Lee et al., 2012; Russell et al, 2010; Yang et al., 2009). Given the location of these sites within a highly polluted region and occasional local biomass burning, the inferred aerosol types make sense. The relatively low SAE compared to the sites in Cluster 3 can be attributed to the contribution of sea salt aerosol.

Cluster 3 is comprised of ARN, SGP, FKB, BEO, LLN, PVC, APP, BND and KPS- all of which are continental polluted stations, with the exception of PVC and ARN, which are marine polluted sites characterized by both oceanic air masses and continental polluted air masses. The stations in Cluster 3 have the highest SAE values of all the sites (SAE~2) and thus the smallest particles, and meet the optical property thresholds for fossil fuel and biomass burning aerosols observed in previous studies (Cappa et al., 2016; Cazorla et al., 2013; Clarke et al., 2007; Lee et al., 2012). What separates Cluster 2 from Cluster 3 is the slightly higher SSA value for sites in Cluster 2, which may indicate a stronger sea salt aerosol signal at the Cluster 2 marine polluted sites that is not present at most sites in Cluster 3 (except ARN). Additionally, aerosol loading at sites in Cluster 2 is substantially higher (factor of 2-3) than at the sites in Cluster 3.

Cluster 4 contains NIM and PGH, both characterized by very high aerosol loadings (high σ_{sp}), low SAE (SAE~0.5), indicating large particle, and AAE~1.3. The median optical signature of the aerosol at these monitoring stations is representative of dust aerosol (Costabile et al., 2013; Cazorla et al., 2013; Lee et al., 2012; Yang et al., 2009; Russell et al., 2010; Bahadur et al., 2012) potentially mixed with a black and/or brown carbon (Cappa et al., 2016). Previous studies report that high loadings of dust aerosol are found at both PGH (Kotamarthi, 2013) and NIM (Osborne et al., 2008), validating the inferred aerosol type of this cluster. NIM also experiences some biomass burning aerosol (Osborne et al., 2008; MacFarlane et al., 2009). PGH experiences fresh biofuel burning daily as well as seasonal pollen and large bioaerosol from the surrounding forest, so it is not surprising that these sites get designated as 'Mixed Dust/BC/BrC'.

Cluster 5 is comprised of just one monitoring site, CPR. Although it is typically undesirable to have a cluster with only one member, the algorithm placed CPR in its own cluster for many choices of 'k' clusters, indicating that indeed it is unique enough to be in its own cluster. Cluster 5 doesn't have interquartile spread on the values since it only has one station. Cluster 5 (CPR) is characterized by the highest AAE value (AAE median = 2.00), the smallest SAE value (SAE median = 0.28), and a high particle loading. Both marine aerosols and African dust event aerosols have been measured at Puerto Rican sites (e.g., Denjean et al., 2016; Propsero et al., 2014; Kalashnikova and Kahn, 2008; Reid et al., 2003), as well as occasional anthropogenic aerosol (Allan et al., 2008) biomass burning aerosol and volcanic aerosol. Based on median optical properties, the CPR aerosol falls into the dust or 'Mixed Dust/BC/BrC' regime suggested by previous classification schemes (Cappa et al., 2016, Costabile et al., 2013; Cazorla et al., 2013; Lee et al., 2012; Yang et al., 2009; Russell et al., 2010; Bahadur et al., 2012), though knowledge of the CPR site suggests the station would also measure sea salt.

Monitoring stations CPT, GRW, PYE, THD and WLG make up Cluster 6. The cluster is defined by high SSA values (SSA~0.95), low σ_{sp} and moderate SAE (SAE~0.96) and AAE (AAE~1.12) values. It is worth noting that the AAE values at stations in this cluster have a large spread, and GRW only fits the criteria of this cluster with its high SSA, but is otherwise anomalous in its optical properties. The optical property values fall within the bounds of multiple aerosol type thresholds suggested by various studies, and thus the aerosol type will be considered mixed. The majority of stations within this cluster (CPT, GRW, PYE, THD) are remote marine sites that may receive occasional dust, biomass burning and/or pollution events amid typical sea salt particle measurements, so a mixed dominant aerosol type might be expected. A slight outlier to this group is WLG, which is a remote mountaintop site. Although sea salt particles are very unlikely to be a constituent of the WLG aerosol, WLG does experience strong dust and pollution events, depending on the season and wind direction (Kivekäs et al., 2009). The main difference between Clusters 5 and 6 is the higher AAE and lower SAE values in Cluster 5 (CPR), indicating a stronger presence of large dust particles and smaller influence of BC at CPR compared to the locations in Cluster 6.

Table S5. Median optical property values at 24 monitoring stations in the NOAA Federated Network, both filtered (σ_{sp} > 1 Mm^{-1} and σ_{ap} > 0.5 Mm^{-1}) values and non-filtered

Station	# data points filtered (unfiltered)	SAE (lq, uq) Filtered (σ_{sp}>1 Mm^{-1})	AAE (lq, uq) Filtered (σ_{ap}>0.5 Mm^{-1})	SAE (lq, uq) Unfiltered	AAE (lq, uq) Unfiltered	σ_{sp} (Mm^{-1}) Filtered (>1 Mm^{-1})	σ_{ap} (Mm^{-1}) Filtered (>0.5 Mm^{-1})	σ_{sp} (Mm^{-1}) Un-filtered	σ_{ap} (Mm^{-1}) Un-filtered
ALT	1648 (6266)	**1.27** (1.05, 1.43)	**0.86** (0.79, 0.95)	**1.18** (0.81, 1.45)	**0.85** (0.65, 1.00)	**9.69** (8.16, 12.11)	**0.75** (0.63, 0.90)	**3.44** (1.31, 7.85)	**0.17** (0.06, 0.53)
AMY	8914 (8933)	**1.57** (1.36, 1.75)	**1.22** (0.94, 1.42)	**1.57** (1.36, 1.75)	**1.22** (0.94, 1.42)	**107.72** (61.81, 189.54)	**8.72** (5.53, 13.44)	**107.50** (61.69, 189.2)	**8.70** (5.51, 13.44)
APP	15547 (16201)	**2.11** (1.94, 2.26)	**1.20** (0.87, 1.48)	**2.10** (1.93, 2.26)	**1.21** (0.87, 1.48)	**24.46** (14.59, 38.17)	**2.13** (1.38, 3.19)	**23.58** (13.53, 37.33)	**2.06** (1.3, 3.13)

ARN	8237 (8605)	**1.37** (0.97, 1.70)	**1.32** (1.16, 1.50)	**1.34** (0.90, 1.68)	**1.31** (1.14, 1.49)	**26.10** (16.7, 40.73)	**3.15** (1.83, 5.04)	**25.89** (16.50, 40.28)	**3.03** (1.64, 4.91)
BEO	5775 (9397)	**1.87** (1.44, 2.07)	**1.31** (1.05, 1.55)	**1.75** (1.09, 2.04)	**1.34** (1.07, 1.59)	**22.64** (11.52, 40.04)	**1.94** (1.07, 3.21)	**9.55** (2.61, 27.8)	**0.89** (0.25, 2.34)
BND	15257 (15523)	**2.01** (1.84, 2.17)	**1.15** (0.93, 1.34)	**2.01** (1.84, 2.17)	**1.15** (0.92, 1.34)	**33.06** (19.90, 55.14)	**2.69** (1.58, 4.17)	**32.58** (19.45, 54.67)	**2.65** (1.53, 4.13)
BRW	2612 (16574)	**1.17** (0.78, 1.52)	**0.99** (0.89, 1.10)	**1.00** (0.54, 1.50)	**0.87** (0.51, 1.06)	**10.47** (7.87, 15.97)	**0.73** (0.60, 1.00)	**6.05** (2.62, 10.89)	**0.18** (0.06, 0.36)
CPR	5744 (14314)	**0.28** (0.17, 0.54)	**2.00** (1.16, 2.65)	**0.31** (0.19, 0.52)	**1.38** (0.89, 2.35)	**35.32** (24.33, 50.22)	**1.01** (0.71, 1.5)	**21.15** (14.63, 34.03)	**0.37** (0.14, 0.84)
CPT	3158 (12599)	**0.67** (0.34, 1.14)	**1.12** (0.97, 1.31)	**0.22** (0.05, 0.59)	**1.15** (0.81, 1.57)	**21.31** (13.76, 29.79)	**1.14** (0.73, 2.45)	**17.27** (11.6, 24.74)	**0.17** (0.04, 0.51)
FKB	5543 (5593)	**1.80** (1.59, 1.95)	**1.07** (0.98, 1.16)	**1.80** (1.59, 1.95)	**1.07** (0.98, 1.16)	**32.37** (18.12, 57.77)	**5.75** (3.17, 9.96)	**32.24** (17.98, 57.81)	**5.7** (3.10, 9.90)
GRW	7960 (1419)	**-0.12** (-0.34, 0.19)	**0.62** (0.31, 0.85)	**-0.13** (-0.36, 0.16)	**0.51** (0.10, 0.80)	**30.73** (19.37, 47.42)	**0.84** (0.64, 1.29)	**25.96** (16.4, 41.30)	**0.56** (0.36, 0.90)
GSN	10731 (10998)	**1.51** (1.29, 1.70)	**1.21** (1.03, 1.34)	**1.51** (1.28, 1.70)	**1.21** (1.02, 1.34)	**61.85** (37.92, 106.47)	**4.59** (2.70, 7.40)	**60.45** (36.37, 104.95)	**4.49** (2.59, 7.32)
KPS	8923 (8923)	**2.06** (1.90, 2.19)	**1.39** (1.24, 1.60)	**2.06** (1.90, 2.19)	**1.39** (1.24, 1.60)	**45.11** (25.27, 90.90)	**6.27** (3.61, 12.02)	**45.11** (25.27, 90.90)	**6.27** (3.61, 12.02)
LLN	8294 (12333)	**1.94** (1.82, 2.08)	**1.11** (0.97, 1.25)	**1.90** (1.70, 2.07)	**1.10** (0.93, 1.25)	**24.02** (11.81, 40.00)	**2.39** (1.20, 4.56)	**12.09** (3.07, 33.97)	**1.23** (0.31, 3.27)
MLO	2351 (17174)	**1.40** (0.85, 1.76)	**1.42** (1.08, 1.89)	**1.45** (0.67, 1.95)	**1.25** (0.35, 1.99)	**9.38** (4.88, 18.39)	**0.85** (0.64, 1.19)	**2.03** (0.71, 7.36)	**0.1** (0.01, 0.3)
NIM	4527 (4530)	**0.32** (0.14, 0.64)	**1.66** (1.46, 1.22)	**0.32** (0.14, 0.64)	**1.66** (1.46, 1.22)	**91.02** (50.67, 185.24)	**9.25** (5.68, 16.05)	**91.02** (50.67, 185.24)	**9.25** (5.68, 16.05)
PGH	4079 (4172)	**0.75** (0.53, 0.92)	**1.03** (0.88, 1.22)	**0.76** (0.52, 0.93)	**1.03** (0.88, 1.22)	**126.31** (66.48, 232.01)	**8.14** (4.52, 126.31)	**122.96** (63.02, 229.36)	**7.95** (4.30, 15.79)

PVC	4990 (7300)	**2.15** (1.64, 2.50)	**0.99** (0.68, 1.25)	**1.84** (1.21, 2.38)	**0.94** (0.60, 1.24)	**16.08** (10.19, 27.87)	**1.10** (0.75, 1.82)	**16.16** (9.86, 28.80)	**1.41** (0.42, 16.16)
PYE	481 (3856)	**0.98** (0.53, 1.29)	**0.50** (0.30, 1.52)	**0.88** (0.46, 1.53)	**0.17** (-0.47, 0.56)	**40.00** (26.59, 59.97)	**0.69** (0.58, 1.00)	**22.98** (11.87, 40.04)	**0.18** (0.07, 0.33)
SGP	14610 (15430)	**1.77** (1.43, 2.06)	**1.30** (1.05, 1.51)	**1.77** (1.42, 2.06)	**1.30** (1.05, 1.52)	**26.75** (16.06, 42.27)	**2.31** (1.41, 3.42)	**25.48** (14.70, 41.09)	**2.21** (1.27, 3.33)
SPL	8509 (14562)	**1.69** (1.24, 2.03)	**1.37** (1.22, 1.51)	**1.71** (1.26, 2.05)	**1.34** (1.18, 1.48)	**11.50** (7.79, 17.70)	**0.93** (0.69, 1.35)	**7.03** (3.05, 12.94)	**1.01** (0.33, 7.03)
SUM	462 (16558)	**1.93** (1.62, 2.07)	**1.04** (0.93, 1.16)	**1.89** (1.36, 2.31)	**0.92** (0.57, 1.18)	**8.06** (6.27, 11.58)	**0.64** (0.55, 0.81)	**0.7** (0.24, 1.73)	**0.06** (0.02, 0.13)
THD	5283 (11229)	**0.96** (0.62, 1.43)	**1.43** (1.14, 1.70)	**0.86** (0.53, 1.36)	**1.31** (0.91, 1.65)	**21.51** (13.09, 34.56)	**0.94** (0.68, 1.4)	**14.78** (8.23, 25.37)	**0.47** (0.25, 0.90)
WLG	6494 (6569)	**1.10** (0.72, 1.35)	**1.37** (1.22, 1.54)	**1.10** (0.72, 1.35)	**1.37** (1.22, 1.54)	**42.19** (20.08, 101.06)	**3.01** (1.67, 6.16)	**41.52** (19.71, 100.22)	**2.95** (1.64, 6.07)

Table S6. Classifications of trajectory clusters at each station

Station Name	Trajectory Cluster Number	Trajectory Cluster Classification
ALT	1	Remote Marine
ALT	2	Continental Arctic
AMY	1	Continental Polluted
AMY	2	Continental Polluted
AMY	3	Marine Polluted
APP	1	Continental Polluted
APP	2	Continental Polluted

ARN	1	Continental Polluted
ARN	2	Marine Polluted
ARN	3	Marine Polluted
BEO	1	Continental Polluted
BEO	2	Continental Polluted
BND	1	Continental Polluted
BND	2	Continental Polluted
BRW	1	Remote Marine
BRW	2	Continental Arctic
CPR	1	Remote Marine
CPR	2	Remote Marine
CPR	3	Remote Marine
CPR	4	Remote Marine
CPR	5	Polluted Marine
CPT	1	Remote Marine
CPT	2	Remote Marine
FKB	1	Continental Polluted
FKB	2	Continental Polluted
GRW	1	Remote Marine
GRW	2	Remote Marine
GSN	1	Continental Polluted
GSN	2	Polluted Marine
KPS	1	Continental Polluted
KPS	2	Continental Polluted
LLN	1	Continental Polluted
LLN	2	Continental Polluted
MLO	1	Polluted Marine
MLO	2	Polluted Marine

MLO	3	Polluted Marine
NIM	1	Continental Dust/Polluted
NIM	2	Continental Dust
NIM	3	Continental Dust
PGH	1	Continental Dust/Polluted
PGH	2	Continental Dust/Polluted
PYE	1	Remote Marine
PYE	2	Remote Marine
PYE	3	Remote Marine
PVC	1	Continental Polluted
PVC	2	Continental Polluted
PVC	3	Polluted Marine
SGP	1	Continental Polluted
SGP	2	Continental Polluted
SPL	1	Continental Polluted
SPL	2	Continental Polluted
SUM	1	Continental Arctic
SUM	2	Continental Arctic
THD	1	Remote Marine
THD	2	Marine Polluted
THD	3	Marine Polluted
WLG	1	Continental Dust
WLG	2	Continental Dust/Polluted
WLG	3	Continental Polluted
WLG	4	Continental Dust