Statistical characteristics and predictability of particle formation events at Mace Head

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[1] The seasonal characteristics of coastal nucleation events at the Mace Head Atmospheric Research Station, resulting from exposure of macroalgae to the atmosphere, were analyzed for a 2-year period from August 2002 to July 2004. Nucleation events occurred on 58% of the days over the period. The seasonal variation of the number of event days and event duration show a clear cycle, with maximum values in spring and autumn and the minimum values in the winter season. The nucleation events typically start ~75 min prior to the occurrence of the low-tide mark and the event start time is correlated (r = 0.75) to the low-tide height. The intensity of the events, as determined by the peak particle concentrations achieved, is also positively correlated with the amount of tidal areas exposed to ambient air, as determined by the tidal height, and solar radiation. A nucleation potential index (NPI) was developed as a tool to provide a predictive capability for event prediction at Mace Head. The index was derived from normalized tidal height, solar radiation intensity, and wind direction and was compared with the occurrence of nucleation events from the database. The result shows that Mace Head particle formation events can be quite well predicted with a threshold probability of 50%.

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1. Introduction

[2] Our global climate is affected by both natural and anthropogenic aerosols. Aerosols affect the global radiation budget [*Houghton et al.*, 2001], directly through scattering incoming solar radiation [*Charlson et al.*, 1992] and indirectly through modulation of cloud albedo [*Twomey*, 1974, 1991; *Charlson et al.*, 1987]. Secondary aerosol formation is frequently observed over urban areas [*Zhang et al.*, 2004; *Stanier et al.*, 2004], coastal environments [*O'Dowd et al.*, 2002b], the marine boundary layer [*Clarke et al.*, 1998], boreal forests [*Makela et al.*, 1997; *Kulmala et al.*, 2001], Antarctica [*Koponen et al.*, 2003], and Arctic areas [*Wiedensohler et al.*, 1996; *Pirjola et al.*, 1998; *Wiedensohler et al.*, 1996]. These secondary produced particles play an important role in global climate and atmospheric chemistry processes because they can grow to radiatively active sizes through coagulation and condensation processes [*O'Dowd et al.*, 2002b; *Pirjola et al.*, 2002]. A modeling study [*Pirjola et al.*, 2002] showed that the CCN concentration can be increased by up to 100% due to coagulation between particles and condensation of low volatility vapors following coastal nucleation events.

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[3] Coastal particle formation events are frequently observed at Mace Head, located on the west coast of Ireland [*O'Brien et al.*, 2000; *O'Dowd et al.*, 2002b]. The most intensive field measurement study on coastal particle formation events was the project on New Particle Formation and Fate in the Coastal Environment (PARFORCE), which was conducted over the 2-year period from 1998 to 1999, and included two intensive field campaigns in September 1998 and June 1999 at the Mace Head Atmospheric Research Station [*O'Dowd et al.*, 2002b]. It was reported from the campaign that nucleation events occur on 90% of days and particle number concentration often exceeded 10^6 cm^{-3} [*O'Dowd et al.*, 2002a].

[4] Hygroscopic studies of recently formed particles (mobility diameter 8–20 nm) revealed low growth factors (1.0–1.1) and led to a hypothesis that biogenically emitted iodine oxide is the most likely species inducing the growth of particles to detectable sizes [*Vakeva et al.*, 2002]. A transmission electron microscopy and energy-dispersive X-ray study by *Makela et al.* [2002] also confirmed that iodine compounds are found both in the recently nucleated coastal particles with diameter below 10 nm and was linked to seaweed emissions of iodocarbon gases. It is most likely that biogenic releases of iodine containing compounds from coastal tidal areas trigger coastal particle formation either *via* self nucleation, or *via* condensing processes onto stable clusters [*Kulmala et al.*, 2000, 2002] of which formation rates can be stimulated through ternary nucleation theory. Numerous laboratory experiments [*Burkholder et al.*, 2004; *McFiggans et al.*, 2004; *Yoon et al.*, 2004; *Jimenez et al.*,

2003; *Hoffmann et al.*, 2001] were conducted supporting the coastal iodine-aerosol formation relationship.

[5] It has been demonstrated that meteorological conditions also influence coastal nucleation events [*de Leeuw et al.*, 2002]. They investigated influences of tidal height, solar radiation, and micrometeorological variables on coastal nucleation events and concluded that such events are most probable under conditions of drying coastal biota under direct solar radiation where the increased heat flux is thought to help increase biogenic gas emissions.

[6] Although there have been a number of intensive 4–5 weeks field campaigns to investigate the physicochemical characteristics of coastal nucleation events, there is still a lack of a longer-term picture of the characteristics of coastal nucleation measurements. In this study, seasonal variations of nucleation events at Mace Head are investigated using aerosol number size distribution and total concentration data acquired using scanning mobility particle sizers (SMPS) and a bank of condensation particle counters (CPC) over a 2-year period from August 2002 to July 2004. Along with the results of seasonal variation in nucleation events, the frequencies and strength of these events versus tidal height and meteorological conditions are also presented. In addition, we present a simple but quantitative method to predict the Mace Head particle formation events by using tidal height, solar radiation, and wind data.

2. Data

2.1. Site and Instruments

[7] A series of CPCs and SMPS aerosol measurements are operationally deployed at the Mace Head atmospheric research station to identify and characterize the nucleation events. The Mace Head Atmospheric Research Station is located on the west coast of Ireland (53°20′N, 9°54′W) [*Jennings et al.*, 2003], about 50–100 m from the tidal zone. The station is located on a peninsula almost totally surrounded by biologically fertile tidal areas. Only a small sector from 20° to 40° is facing nontidal areas and the wind direction sector between 180° and 300° is from the open North Atlantic Ocean [*Kleefeld et al.*, 2002; *Jennings et al.*, 1997].

[8] The CPC bank is composed of a standard TSI CPC 3025 with a particle diameter size cutoff of 3 nm, a TSI CPC 3010 modified to have a cutoff diameter of 6 nm, and a normal TSI CPC 3010 with a 10 nm cutoff diameter. Data acquisition time resolution for the CPC bank was set to 1 Hz. The CPC3025 was diluted by a factor of 17.4 to avoid saturation of the CPC total number concentration in excess of 100,000 cm^{-3}. The dilution system which was used for the CPC 3025 is discussed in detail by *Yoon et al.* [2005]. It was possible to detect number concentration of ultrafine particles up to 1.7×10^6 cm^{-3} with the aid of this calibrated dilution system. The counting efficiencies of these CPCs and the stability of the dilution chamber were regularly monitored and verified.

[9] The Mace Head SMPS consists of a TSI differential mobility analyzer (DMA, model 3071) combined with a TSI CPC 3010. This SMPS was set to scan particle number size distributions from 10 to 230 nm with a time resolution of 120 s. We also deployed another SMPS system for nanometer size particles (nano-SMPS), which is a combination of a TSI nano-DMA (model 3085) and a TSI CPC 3025. The nano-SMPS was set to scan from 3.5 nm to 15 nm equivalent voltages, and each scan took 30 seconds. Closed loop flow configurations for sheath and excess air [*Jokinen and Makela*, 1997] were used for both SMPSs. A flow ratio between sheath and sample air was maintained at 10 to 1 (10 to 1 L min^{-1} for SMPS and 15 to 1.5 L min^{-1} for a nano-SMPS, respectively). Both the CPC and SMPS/nSMPS measurements operated from August 2002 to July 2004 and from these measurements an aerosol number concentration and size distribution database for the period was developed. In addition, meteorological data, such as wind speed and direction, relative humidity, air temperature, air pressure, etc., were also recorded for the same period from the Mace Head 22 m tower.

2.2. Definition of Nucleation Events and Their Characteristics

[10] The background particle number concentration in Mace Head is usually around 500 cm^{-3} when the air mass has a clean maritime origin. During a nucleation event, the number concentration increases dramatically, and the maximum value can easily reach the order of 10^6 cm^{-3}. A nucleation event in this study is defined as the formation of 3–10 nm diameter size particles reaching concentrations of 1000 cm^{-3} or greater. The occurrence of a nucleation event for each day was identified from the validated CPC, SMPS, and nano-SMPS database. Because of mechanical failures and instrument calibration periods, some days lack either CPC or SMPS data and consequently data were not available for all days. Only days with sufficient data to identify the occurrence of events were selected from the 2-year database. Event starting and ending times are determined by evaluating the particle concentration in the 3–10 nm size range from the CPCs, or through cross checking CPC 3025 against nano-SMPS total number concentration data. Maximum particle number concentrations during events were mostly obtained from CPC 3025 data, but the integrated total numbers from nano-SMPS measurements were also used occasionally when CPC 3025 data were not available.

[11] An example of a typical nucleation event at Mace Head is presented in Figure 1. Figure 1 shows a time series of particle number concentration measured by the diluted TSI CPC 3025 on 5 April 2004 (day of year 96). The gray sinusoidal line in Figure 1 represents the tidal height variation through the day while the two vertical lines indicate the event starting and ending times. It should be noted that tidal height is expressed in terms of meters above zero where 0 m corresponds to the lowest tide level possible. Mean water height is approximately 3 m and maximum water height (high tide) on this day is just over 5 m. On this day, the nucleation event started at 0914 UT and lasted for 5 hours and 37 min until 1451 UT. The background aerosol number concentration lay between 300 and 500 cm^{-3}, and the maximum number concentration during the event was 598,900 cm^{-3}. The tidal height when the nucleation event started was calculated to be 1.5 m, and the lowest height during this day of 0.9 m was found around noontime.

3. Results

3.1. Seasonal Characteristics of Nucleation Events

[12] The validated data set was extensive enough to identify the occurrence of nucleation events for 660 days

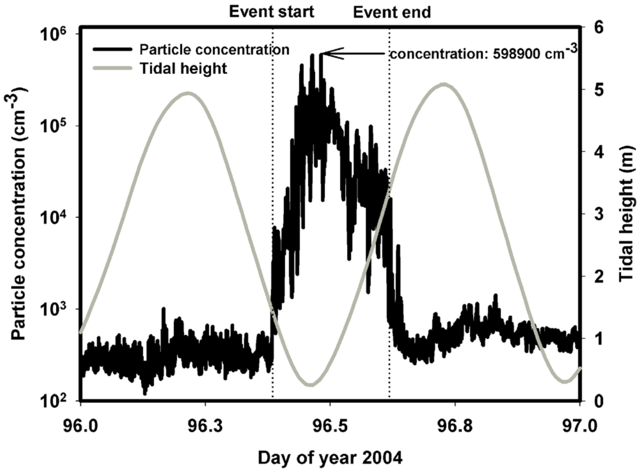

Figure 1. An example of a particle formation event measured by the ultrafine condensation particle counter (TSI CPC 3025) at Mace Head on the 5 April 2004 (day of year 96). The black line indicates the total particle concentration measured by the CPC with a dilution ratio of 17.4, and the gray line corresponds to tidal height. The vertical dotted lines correspond to event starting and ending times.

during the 2-year period. The percentile ratios of these days for each month are shown in Figure 2. In general, it was possible to identify the occurrence of an event on more than 90% of the total number of days, except for August where it was only possible on 71% of the days.

[13] In addition, 450 days can be categorized as clean air mass cases (68% of the total of 660 days). For this analysis, the clean air cases were selected when air mass arriving at Mace Head were within the sectors satisfying the nucleation event types I and II defined by *O'Dowd et al.* [2002a]. In their study, the air mass trajectory regimes reaching Mace Head were categorized into four types according to different distances from the tidal areas and the number of tidal zones encountered. Type I corresponds to clean marine air mass cases with a wind direction from the south to northwest in which the tidal zone encountered is located about 100 m from the measurement station. Type II also corresponds to clean air masses, but airflow is from the northwest to north sector passing over several tidal areas between 100 m and 10 km. Type I and II events are always clean marine events without recirculation of returning continental air. Type III events are typically characterized by modified maritime or continentally influenced air advecting over a tidal region 2–3 km east-to-southeast from the station. Finally, type IV correspond typically to null events and polluted air not advecting over any tidal zone prior to arriving at Mace Head. While type IV events are characterized as null events on the basis that there is no advection over tidal source regions, it should be noted that there can be nonevents also under type I. II, and III conditions even though there is advection over tidal source regions.

[14] In total, 383 nucleation events were detected from all types of air masses (58% of the total of 660 days). When considering clean air mass cases only, 254 events were found among 450 days or 56% of the total clean air mass days. The monthly number of event days for the 2-year period is shown in Figure 3. Table 1 shows the accumulated number and percentile ratio of event days for each month. Table 1 also includes the results of nucleation events for clean air mass cases at Mace Head. The total number and percentage of events days show clear seasonal variations, with maxima in spring and autumn and minima in winter. This clear seasonality is likely to be related to the activity of marine biota. A study by *Savoie et al.* [2002] during the Atmosphere/Ocean Chemistry Experiment (AEROCE) campaign, also showed that the concentration of marine-biota activity related species, such as nonsea salt sulfate and methanesulfonate (MSA) in the particle phase, have a seasonal cycle at Mace Head showing high values in spring-autumn and a lower value in the winter season.

[15] Total accumulated monthly hours of nucleation events are also analyzed and are shown in Figure 4. In total, 1853 hours of nucleation events were observed at Mace Head during the 2-year period. When only type I and II are considered for the same period, 1235 hours of events were recorded under clean marine air mass cases. For all event types (referring to the black curve in Figure 4), the

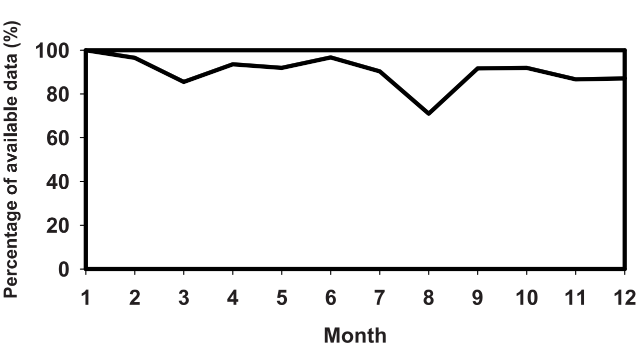

Figure 2. Monthly percentile ratio of measurement days to decide on the occurrence of a nucleation event.

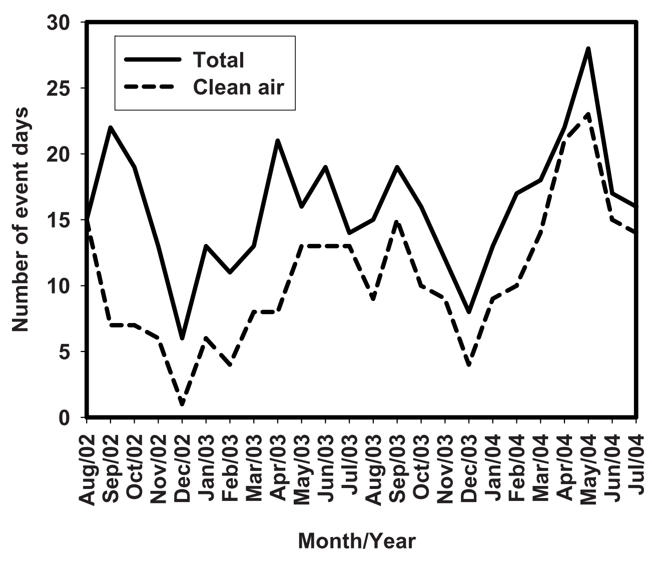

Figure 3. Monthly number of particle formation event days from August 2002 to July 2004. The black curve is for all cases of air mass origin, and the dashed curve is for clean marine air masses only.

Table 1. Number and Percentile Ratio of Nucleation Event Days Identified From the 2-Year Data Set

	All Air Mass Case		Clean Air Mass Case	
Month	Number of Days	Percentile Ratio	Number of Days	Percentile Ratio
1	26	42	15	37
2	28	51	14	52
3	31	59	22	65
4	43	76	29	82
5	44	76	36	72
6	36	62	28	60
7	30	54	27	53
8	30	69	24	65
9	41	74	22	73
10	35	61	17	56
11	25	48	15	40
12	14	26	5	21

maximum monthly accumulated event hours was found in May to be 129, and a secondary peak was found in September to be 92 hours. The value for the summer season is relatively lower than for spring and autumn. For example, a total of 67 hours of nucleation events were recorded in July. The accumulated duration hours for winter time show the lowest values, 27 hours for December and 45 hours in January. For only type I and II cases (refer to gray line in Figure 4), the maximum event hours is also found in May with 108 hours, but a secondary peak in autumn season is not evident, compared with the case where all air mass types are considered. The value for December is only 8 hours for the clean marine air case, showing the same pattern as the case with all event cases. The lower contribution of clean air events to the total number of events for months 9–10 is most likely due to the more frequent occurrence of high-pressure weather systems during these months, thus bringing more continental air to Mace Head; hence the number of clean marine events would be reduced.

[16] Figure 5 shows the mean daily nucleation event duration of hours for each month. These monthly data also show similar seasonal variations to the results of the number of events regardless of event types, having peaks in spring and autumn (5.8 hours in May and 5.2 hours in October), and a minimum in winter (3.4 hours in January). Data for the summer season also show lower values than for the spring/autumn cases, for example, 4.5 hours in July. The total averaged event duration was calculated to be 4.5 hours when all event types are considered.

3.2. Nucleation Events and Tide

[17] The starting times (the time of first detection of 3–5 nm particles) was noted for all of the 383 events. In addition, the tidal height for the Galway Bay area as a function of time of day was calculated, and it was possible to determine the time when the tidal height was at its lowest. The relationship between these two data sets for all event types and marine air cases are shown in Figure 6. In general, nucleation starts close to the time of lowest tide height and nucleation events were observed to start as early as 0400–0500 UT, and as late in the day as 1700–1800 UT. Correlation coefficients (r) for the two cases were derived from the relationship between event start time and low-tide time. This value was 0.75 for all air mass types and 0.82 for clean air mass cases, respectively. Nucleation events usually started prior to the low-tide time with the exception of some early morning events. It has been postulated that the source of the precursor gas driving particle formation is the exposed macroalgae [*O'Dowd et al.*, 2002c; *Jimenez et al.*, 2003; *O'Dowd et al.*, 2002b] and that as the tide level approaches the lowest tide height, there has been sufficient time for the already exposed algae to dry and undergo significant stressing to release the required iodine vapors to produce nucleation. The data set shows that nucleation starts 75 min on average before the low-tide height occurrence for all events and 64 min on average before low-tide height for clean marine events. This could be due to differing source strengths in the regions air advected over under nonmarine and marine conditions in additional to variations in distance traveled from the source regions to the measurement station.

[18] The maximum particle number concentrations were determined for all nucleation events. In addition, the lowest

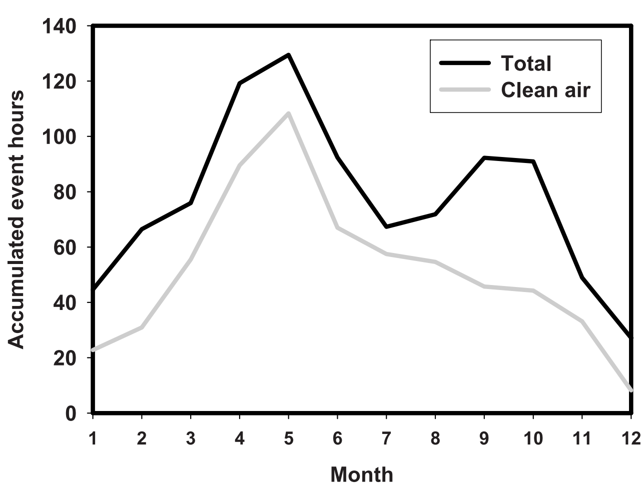

Figure 4. Monthly accumulated duration of event hours, for all event cases (black curve) and clean air mass cases only (gray curve).

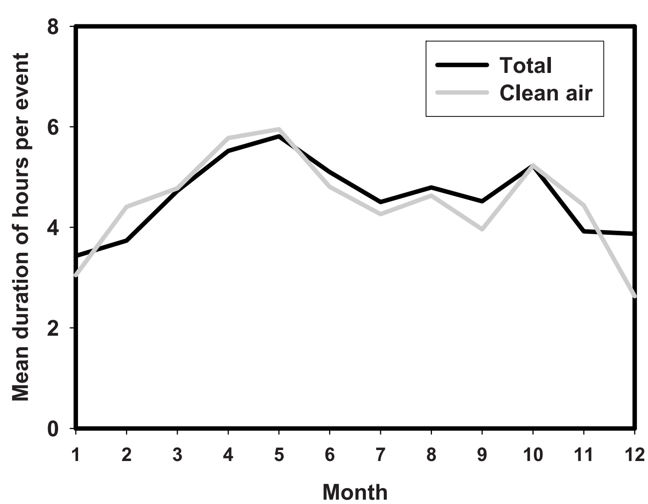

Figure 5. Monthly averaged duration of nucleation event hours per event. All types of event cases are shown by the black line, and clean air mass cases are shown by the gray line.

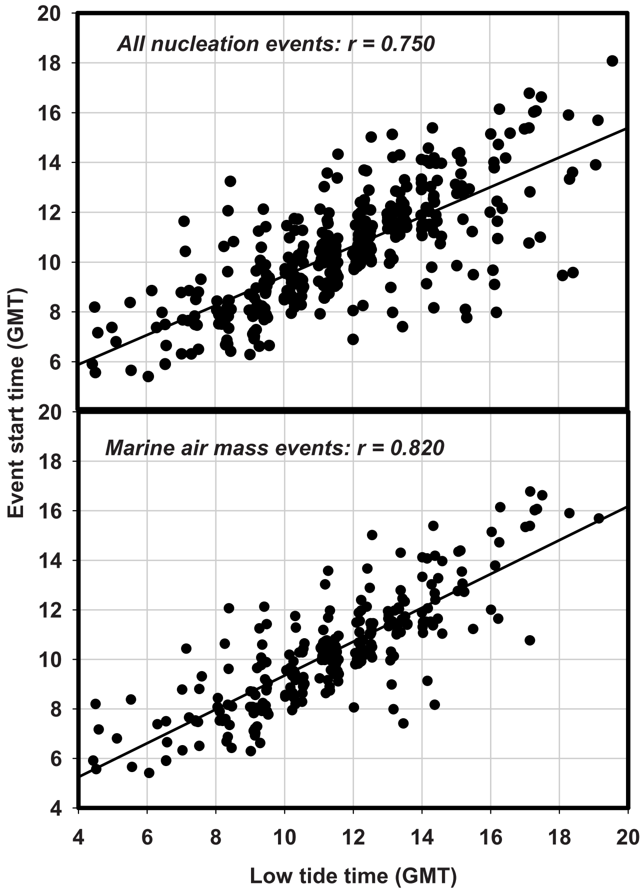

Figure 6. Relationship between the lowest tide time and nucleation event start time for (top) all event types and (bottom) clean air mass cases only. Correlation coefficients are represented by r, and the first-order regression lines are shown by the black lines.

tidal heights (i.e., tidal amplitude, corresponding to exposure area) for the event days were also obtained. This relationship is shown in Figure 7. The lowest tidal heights were categorized into bins with 0.25 m resolution for this analysis. The average and standard deviation values of the maximum particle number concentration were calculated for each height bin. Though the standard deviation values are high for each tidal height bin, a negative correlation between the two variables is found. The amount of exposed tidal area is closely related to the tidal height, as a lower tidal height means wider exposure of the tidal area to the atmosphere and to solar radiation. As a result, the low-tide height can be used as an indicator of the amount of exposed tidal area, and in turn, an indicator of the concentration of emitted precursor gases. It can be inferred from this analysis that nucleation events tend to be stronger as the area of exposure of the tidal zone increases.

3.3. Predictability of Nucleation Events From Tidal Height, Solar Radiation, and Wind Direction

[19] As discussed in sections 3.1 and 3.2, coastal nucleation events are strongly influenced by the availability of precursor gases and solar radiation. The concentration of precursor gases is related to the width of tidal zone exposed to the atmosphere, and in turn, tidal height. In addition to these two factors, the origin of air masses arriving at the measurement location is also particularly important, especially at Mace Head [*O'Dowd et al.*, 2002a] since this also determines which tidal areas the air is exposed to. Attempts to predict the occurrence of particle formation events at Mace Head by relating them to the occurrence of three factors, that of solar radiation, tidal height and wind direction, are described in this section. The intensity of clear sky solar radiation at ground level at Mace Head was normalized for each day and averaged over a 30 min period. This normalization was done by applying 0 for nighttimes and 1 for the solar radiation at noontime and scaling the radiation level to each time step. Tidal heights were also normalized to the same time resolution. As the lowest tidal height at Mace Head corresponds to the maximum exposure of tidal area, the lowest 2-year tidal height value of −0.006 m at 1230 UT on 20 March 2003 was normalized to 1, and 0 was scaled for the maximum height of 5.56 m (1730 UT, 7 October 2002) for the 2-year period from August 2002 to July 2004.

[20] A frequency analysis of daily wind directions at Mace Head was performed to find out prevailing wind directions for the 2-year period. In this analysis, 16 wind direction segments were used and the raw wind direction data has a time resolution of 1 min. The distribution of daily wind direction is shown in Figure 8. The most frequent prevailing daily wind direction segment was found to be the south-southwest (202.5° ± 11.25°) with 100 of the 730 days from this wind section. The least frequent wind direction sector was the north-northeast (22.5° ± 11.25°) with only 10 days from the 2-year data set.

[21] Figure 9 shows the probability of occurrence of a nucleation event for each wind direction segment. The probability values for each direction were found by dividing the number of observed nucleation events for a specific wind direction sector by the total number of occurrences of the wind direction. Wind and nucleation occurrence data with a time resolution of 30 min were used, and as a result, there are 48 time slices for 1 day (35,040 cases for the

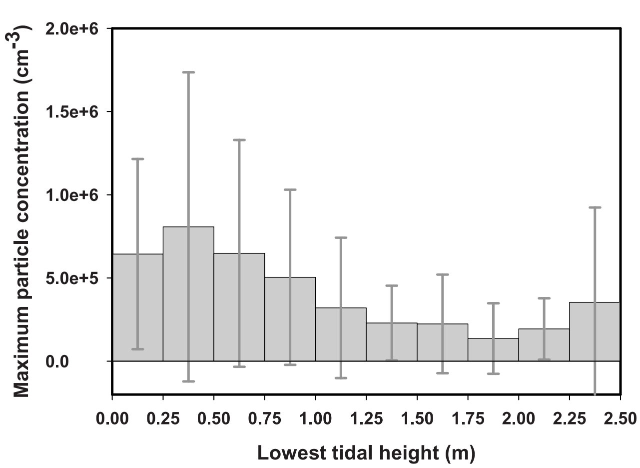

Figure 7. Relationship between the lowest tidal height and the maximum particle number concentration during events. The plot shows the average of maximum concentration ±1 SD for each tidal height bin.

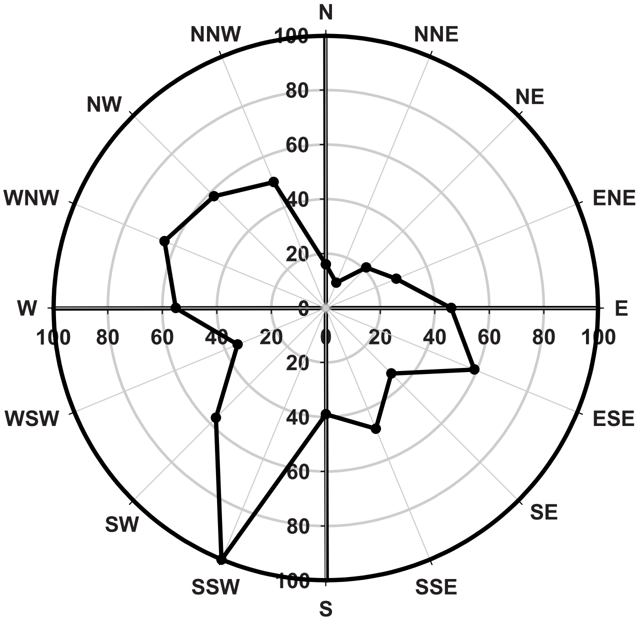

Figure 8. Frequency distribution of daily wind direction at Mace Head for the 2-year period from August 2002 to July 2004.

2-year period). The maximum probability of occurrence of an event was 0.214 when the wind direction was 0° ± 11.25° (N). The smallest value of 0.030 was found when the wind direction was 248° ± 11.25° (WSW). These wind direction-related nucleation probability data were normalized for each of the 16 reference directions, by scaling 1 for the value of 0.214 for the wind direction north and 0 for 0.030 for WSW.

[22] A nucleation event potential index (NPI) for every 30 min for the 2 years was derived. Two kinds of NPI (NPI-I and NPI-II) were obtained. The NPI-I was obtained by multiplying normalized tidal height by daily normalized solar radiation. The NPI-II was calculated by multiplying the scaled wind direction factor by the NPI-I. Then occurrences of nucleation events with the same time step were analyzed in terms of index values, and these results are shown in Tables 2 and 3. Table 2 shows the probability of occurrence of events in terms of NPI-I. For example, if NPI-I is higher than 0.8, it can be interpreted that the occurrence of nucleation events at Mace Head is more than 62% likely. According to this analysis with NPI-I, the 50% probability threshold of a nucleation event has an index value of 0.75. When the scaled wind direction factor is added, NPI-II, the absolute values become smaller; for example, an index value of 0.5 gives a 70% probability of an event. In this case, the 50% threshold point is found at a NPI-II value of 0.43 (refer to Table 3).

[23] Derived NPI-I and NPI-II values were compared with the occurrence of real-time nucleation events for the 2 years of data. Figure 10 shows examples of this comparison. The horizontal lines in Figures 10a–10c correspond to the 50% probability according to both NPI-I and NPI-II. The occurrence of particle formation is defined as 0 for a nonevent and 1 for an event. Results for the period from 29 March (day of year, DOY 88) to 7 April 2003 (DOY 97) are presented in Figure 10a. There were 10 successive days with nucleation events during the period. The values for NPI-I and -II are also high for this period. The NPI-I values are close to or slightly higher than the 50% threshold for this period, while the NPI-II values are higher than the 50% threshold, and nucleation events were observed. Figure 10b shows a second example for a period from 18 March (DOY 78) to 26 March 2004 (DOY 86). For this period, the NPI-I values are also close to or slightly higher than the 50% threshold similar to Figure 10a, but a nucleation event was not observed on DOY 78 and 80 mainly due to unfavorable wind direction conditions which contribute to the low NPI-II value. By contrast, nucleation events occurred on DOY 82, 83, and 84 though NPI-I values are small. This is probably because of favorable wind direction conditions which made the NPI-II values higher than the 50% threshold. The effects of wind direction on the occurrence of nucleation events are more clearly seen in Figure 10c, which presents results from 13 June to 21 June 2004 (DOY 165–173 of 2004). Nucleation events did not occur for the first 4 days (DOY 165–168) during the period due to low NPI-II values. By contrast, events occurred without exception for the next

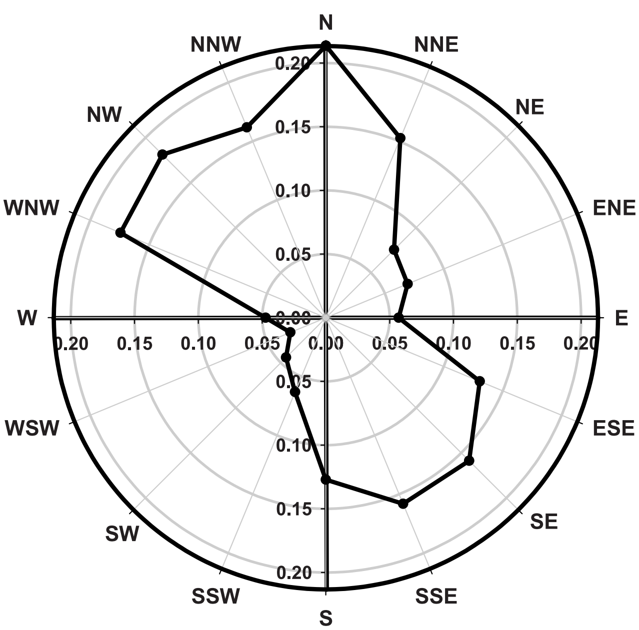

Figure 9. Probability of occurrence of a nucleation event for each wind direction sector for the period.

Table 2. Percentile Probability of Occurrence of Nucleation Events According to Index I Derived From Cloudless Solar Radiation Intensity and Tidal Height

Value[a]	Number of Nonevents	Number of Events	Total Number of Cases	Ratio of Event Cases, %
Below 0.1	20,860	611	21,471	2.8
0.1–0.2	2,632	324	2,956	11.0
0.2–0.3	2,414	367	2,781	13.2
0.3–0.4	1,980	400	2,380	16.8
0.4–0.5	1,283	399	1,682	23.7
0.5–0.6	928	392	1,320	29.7
0.6–0.7	646	395	1,041	37.9
0.7–0.8	421	440	861	51.1
Over 0.8	224	372	596	62.4

[a]Scaled daily solar radiation times tidal height.

Table 3. Percentile Probability of Occurrence of Nucleation Events According to Index II Derived From Cloudless Solar Radiation Intensity, Tidal Height, and Wind Direction

Value[a]	Number of Nonevent	Number of Events	Total Number of Cases	Ratio of Event Cases, %
Below 0.1	26,112	1,222	27,334	4.5
0.1–0.2	2,034	561	2,595	21.6
0.2–0.3	1,201	453	1,654	27.4
0.3–0.4	677	458	1,135	40.4
0.4–0.5	291	380	671	56.6
Over 0.5	245	577	822	70.2

[a]Scaled daily solar radiation times tidal height times normalized wind factor.

5 days (DOY 169–173), which are mainly dominated by high NPI-II values. It is shown that both NPI-I and -II indices can be used to predict Mace Head nucleation events satisfactorily with a probability of 50%, but the NPI-II is found to be a more useful tool because of the nature of the topography around Mace Head. The Mace Head Atmospheric Research Station is surrounded mostly by tidal areas, but the widest tidal zones are located to the northwest and north of the measurement site. As a result, it is plausible to also consider wind direction (NPI-II) when predicting nucleation event probabilities at Mace Head.

[24] This analysis is not meant to provide a detailed linkage to all key processes associated with coastal nucleation events at Mace Head since there are too many complex processes to incorporate into a complete predictive system. Such processes include detailed micrometeorological parameters, vapor emissions, photolysis and oxidation rates, condensation rates to preexisting aerosol sinks, which in turn can be dominated by wind-driven sea spray production. Further, the gas-phase iodine cycle is extremely complex with many open questions relating to actual iodine vapor pathways to iodine oxide aerosol remaining the subject of current debate; while quantification of the nucleation mechanism and the formation of aerosol particles from nucleated embryos still remains a challenge to elucidate. In addition, none of these processes are measured or quantified

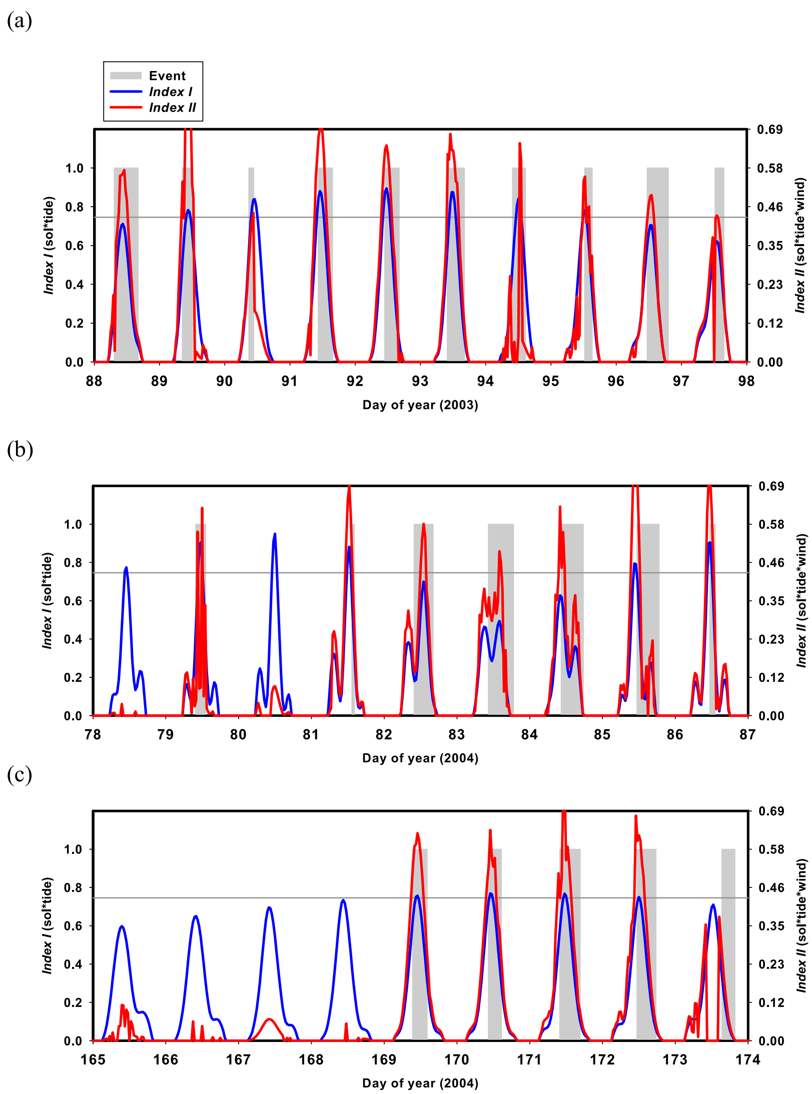

Figure 10. Relationship between nucleation events and indices NPI-I and -II (index I in blue; index II in red) for the periods (a) from 29 March to 7 April 2003, (b) from 18 March to 26 March 2004, and (c) from 13 June to 21 June 2004.

on an operational basis at Mace Head and therefore cannot be incorporated into a predictive tool. Nevertheless, what this analysis does demonstrate is that from three operationally measured environmental conditions at Mace Head, a simple predictive tool can be effectively utilized to predict coastal nucleation events to an acceptable level of predictive accuracy.

4. Summary and Conclusions

[25] The formation of nanometer diameter sized particles at a coastal environment has been analyzed for a 2-year period from August 2002 to July 2004 at the Mace Head Atmospheric Research Station. Coastal nucleation events occur all year-round usually coinciding with low tide and sunlight. It is shown that nucleation events occurred on 58% of the total days. Events occurred on more than 70% of the days in May and September, but only on 26% of days in December. Event duration time also showed a similar seasonal cycle, 5–6 hours in May and October and only 3.4 hours in January. The seasonal variation of the number of event days and event duration hours show a clear cycle, with larger values in spring and autumn, and the lowest in the winter season. It was found that the number of event days for summer is relatively lower than for spring/autumn. This seasonal variation is mainly due to the amount of precursor gases emitted from marine algae during low tide.

[26] A nucleation event is more likely when tidal height at Mace Head is closer to the low-tide level which results in increased emissions of nucleation precursor gases into the atmosphere. A correlation analysis of the nucleation start time and tidal height results in a correlation coefficient of 0.75. It was found that on average, nucleation starts 75 min prior to the low-tide mark. The analysis suggests that 75 min prior to low tide, there is sufficient biota exposed and that sufficient water has evaporated from the tidal beds [*de Leeuw et al.*, 2002] to promote a sufficient flux of precursor gases to promote nucleation at a detectable rate. The peak particle concentration is also negatively correlated with low-tide height indicating that the more area that is exposed, the stronger the event is likely to be, although the dependency is less clear in the relationship between event start time and tide height. A nucleation potential index (NPI) was employed as a tool to provide a predictive capability of nucleation events at Mace Head. The NPI-I, which was derived only from tidal height and solar radiation data, could predict daily probability of nucleation events for days with a favorable wind direction condition. Though nucleation events occur regardless of wind directions at Mace Head, the probabilities of event occurrence differ for each direction because of various topographical characteristics around Mace Head. For example, the probability of a nucleation event is highest when the wind direction is in the west-northwest-to-north sector because the air mass arriving at the measurement station has passed over a wide range of biologically fertile tidal areas. When this probability function according to wind direction was added to the NPI-I and defined as NPI-II, the daily nucleation events can be better predicted for a threshold of 50% probability.

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