Characterisation of Atmospheric Properties during MAPTIP

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SUMMARY

Aerosol particle size distributions were measured during the MAPTIP experiment at Hr.Ms. Tydeman. Extinction coefficients were calculated using a Mie routine. The analysis shows that the TNO local MPN aerosol model accurately predicts the aerosol extinction for data in its range of validity. The applicability of the MPN model decreases outside its limits for geographical region and environmental parameters. The model is compared to the Navy Aerosol Model. The chemical composition of the aerosol during MAPTIP is discussed in terms of geography and environmental parameters.

1. INTRODUCTION

The performance of electro-optical systems is affected by suspended aerosol particles that scatter and absorb electromagnetic radiation. In this way, the atmospheric aerosol is in part responsible for the degradation of the radiance contrast between a target and its natural background, as viewed by an infrared sensor. For correct assessment of the effective range of electro-optical systems in a given meteorological scenario it is necessary to understand and to predict the effects of the atmosphere on propagation of electromagnetic radiation. Hence, there is a need for accurate atmospheric models that can be used in propagation prediction codes.

Presently, the Atmospheric Transmission/Radiance computer code LOWTRAN[1] is the primary tool for this assessment. This code uses the Navy Aerosol Model,[2,3,4] NAM, to predict the transmission in the marine atmosphere along horizontal paths at shipboard levels (around 10 m). NAM is based on an extensive data set including a variety of geographical locations and performs well in open-ocean conditions at mid-latitudes in low to moderate wind speeds. Nevertheless, discrepancies have been observed in sub-tropical regions, high wind conditions and in coastal areas with polluted continental air masses.[5,6,7] Apparently, the influence of a variety of nearby local sources is not well handled by NAM.

A local aerosol model has been developed for the polluted North Sea, based on aerosol size distributions as measured during the HEXMAX experiment[8] in October - November 1986 at the Meetpost Noordwijk (MPN), a platform located at 9 km off the Dutch coast. The MPN aerosol model calculates the aerosol extinction in two steps. First, the aerosol size distribution is calculated from meteorological parameters, including wind direction.[9] The wind rose between 110 - 340° is partitioned in various sectors based on industrial, rural continental and predominantly marine influences. Subsequently, an approximated Mie-algorithm is used to calculate the aerosol extinction from the size distribution. The model predicts the aerosol extinction to within a factor of 2.0 (68% confidence limit), which is a factor of 2 better than the generally used NAM model.[5] The effect of wind direction on the aerosol extinction has been ascribed to the relative contribution of aerosol from marine and continental origin.[10] Data on the spatial variability of aerosol and other environmental parameters, at various distances from the coast, are required to validate the MPN aerosol model and to ascertain the geographical limits of its applicability.

The MAPTIP (Marine Aerosol Properties and Thermal Imager Performance) experiment,[11] conducted between October 11 and November 5, 1993, yielded new data sets on aerosol particle size distributions and environmental parameters in the area of Meetpost Noordwijk (MPN). During MAPTIP, the oceanic research vessel Hr.Ms. Tydeman was made available by the RNL Navy. One of the tasks of Hr.Ms. Tydeman was to monitor environmental conditions and their spatial variations. Standard meteorological parameters such as wind speed, wind direction, air temperature, relative humidity, pressure and water temperature were measured continously. In addition, aerosol particle size distributions were recorded and impactor samples were taken at several times to determine the aerosol chemical composition. The operational sequence of MAPTIP was chosen such that Hr.Ms. Tydeman was able to collect data both near MPN and at various locations a day sailing from MPN.[12] Figure 1 presents the composite sailing schedule of Hr.Ms. Tydeman.

The present contribution focusses on the aerosol extinction, calculated from the particle size distributions measured at Hr.Ms. Tydeman. The experimental extinction values are compared to predictions by NAM and the local MPN aerosol model. We show that the local MPN model provides an

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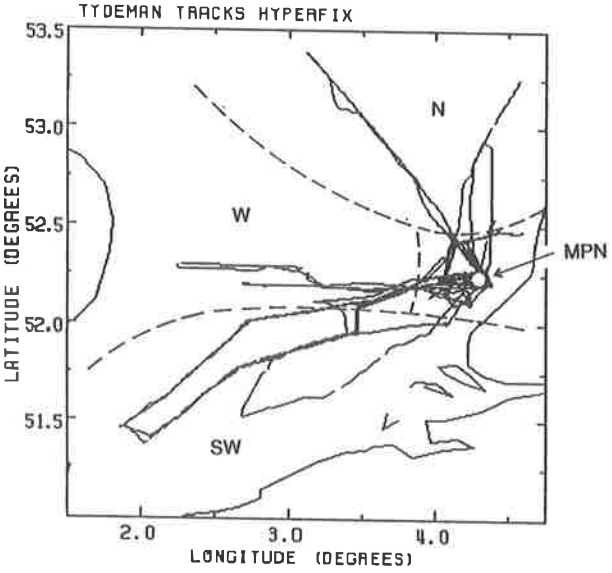

Figure 1: Composite sailing schedule of Hr.Ms. Tydeman. The position of the MPN tower is indicated by the arrow. The dashed lines define the regions used in the assessment of spatial variability of the aerosol extinction.

accurate estimate of the aerosol extinction close to the MPN tower, but that the accuracy is reduced further from the coast. The aerosol extinction and aerosol chemical composition is discussed in terms of environmental parameters.

2. EXPERIMENTS

2.1 Aerosol sampling

Two Knollenberg optical particle counters (Particle Measuring Systems, Boulder, Colorado) were used to measure particle size distributions in the 0.16 - 32 µm diameter range, that is, an ASAS 300 A active probe and a CSAS 100 HV classical probe. The probes were mounted on the roof of the bridge of Hr.Ms.Tydeman, at a height of approximately 15 m above the mean water level. The aspiration tubes were manually pointed into the wind. The probes were operated unattended and could thus be used 24 hours per day. Down times occurred due to rain, maintainance, data processing and storage, probe failure, etc.

The data were stored and digitized in the data acquisition system and transferred to a personal computer, via an IEEE interface at preset time intervals. Particle size distributions dN/dD ($\mu m^{-1}cm^{-3}$) were calculated and stored together with meteorological parameters obtained from Hr.Ms. Tydeman standard instrumentation and TNO operated sensors. The size distributions were averaged over 10-min periods and polynomials of degree 1 and 5 were fitted to the datapoints in *log (dN/dD)* versus *log (D)* space.

Aerosol samples for elemental analysis were collected in a continuous forced air flow inside a wind tunnel, which directed itself continuously towards the wind. The wind tunnel was mounted next to the PMS-probes, at a height of approximately 16 m above the mean water level. The aerosols were separated by a Batelle cascade impactor (cut off diameters 0.25, 0.5, 1, 2, 4, 8 and 16 µm) and collected on quarz reflector holders. Aerosol collection took place while Hr.Ms. Tydeman was anchored and sample times were typically 11.5 hours. The samples were subsequently analyzed at the University of Antwerp (UIA) by the Total-Reflection Xray Fluorescence (TXRF) Technique[11] to yield the elemental composition in each size bin.

2.2 Postprocessing

The experimental aerosol size distributions were used to compute extinction coefficients at 5 wavelengths (10.6, 4.0, 1.064, 0.6943 and 0.55 µm), with an upgraded version of the FEL-TNO Mie code.[13,14] This code yields an exact solution for each size, and uses a Simpson integration method to integrate over the particle size distribution until a specified accuracy (0.01) is obtained. Since extinction was not measured directly at the Hr.Ms. Tydeman, we consider those extinctions to be the best estimate of the actual aerosol extinctions during the experiment.

The NAM model predicts an aerosol extinction based on *u24*, the average wind speed in the previous 24 hours, u_{10N}, the wind speed at 10 m height in neutral conditions, the relative humidity and the visibility. The local MPN aerosol model uses u_{10N}, the wind direction, the relative humidity and the ASTD (air-sea temperature difference) to calculate the aerosol extinction. To supply the input parameters for the models, the values of the wind speed u measured at 22.6 m above mean sea level were converted to u_{10N}. Corrections were made using *Liu et al.*[15] and the shallow-water expression for the drag coefficient C_{DN} that applies to the MPN area.[8,16] Subsequently, *u24* was calculated from the u_{10N} values. Because no direct measurements were made of the visibility at 0.55 µm at Hr.Ms. Tydeman, we have calculated the visibility VIS from the extinction coefficients α at 0.55 µm obtained with the exact Mie code: VIS = 3.915 / α.

2.3 Validation

The data set of the PMS probes was validated prior to the analysis. In the last week of the experiment, only a partial aerosol spectrum (0.75 - 32 µm) could be recorded during nights due to failure of the ASAS probe. The lack of data on smaller diameters (0.16 - 0.75 µm) gave rise to systematic underestimation of the slope of the Junge fit compared to records which contained the full size range (0.16 - 32 µm). Therefore, the records with partial spectra have been removed from the data set.

A heavy fog developed in the morning of Sunday 31 October. The fog lasted untill the evening of Tuesday 2 November, but was most dense on Sunday. The fog was accompanied by high humidity, which could give rise to a slight supersaturation. Under these conditions the

hygroscopic aerosols may be activated. They grow very rapidly in size and behave as cloud droplets. Consequently, they cannot be described by equations that apply to subsaturated aerosol. We decided to study the fog event on Sunday 31 October as a case study.

Small numbers of data records were removed from the data set because they were polluted by the diesel exhaust from the stack or because Hr.Ms. Tydeman made short port calls to Scheveningen and IJmuiden. Incidentally, records were rejected for other reasons. A total of 1518 data records (76%) was available for the analysis.

The NAM model could be applied to 1254 data records. The remaining records lacked one or more of the environmental input parameters required for the NAM calculation (mostly relative humidity). The local MPN aerosol model could be applied to only 794 data records, because wind direction data of the Hr.Ms. Tydeman were not always available. In the future, the data set will be completed with wind direction data from the MPN tower, for the periods that Hr.Ms. Tydeman was in the vicinity of MPN.

3. ANALYSIS

3.1 Assessment of the performance of aerosol models

The performances of NAM and the local MPN aerosol model were assessed from logarithmic scatterplots of the extinction predicted by either model, versus the extinction as calculated from the experimental aerosol size distributions. For these plots, the standard deviation $\sigma_{y=x}$ of the data points with respect to the line Y = X (ideal model performance) was calculated. The value of $\sigma_{y=x}$ is a measure for the factor F to within the model predicts the experimental aerosol extinction (68% confidence limit): $\sigma_{y=x} = \log F$.

3.2 Validation of the local MPN aerosol model

The local MPN aerosol model predicts the aerosol extinction at the MPN tower for wind directions between 110 and 340 °N. [9] Therefore, we decided to use only those data records for the validation that had been collected in the vicinity of the MPN tower, i.e., in a region centered around MPN with a radius of roughly 40 km. This subset was then further reduced by imposing the restriction of a wind direction between 110 and 340 °N during data collection. Unfortunately, during the MAPTIP campaign the prevailing wind direction was east and only a limited amount of data records was recorded in wind directions between 110 and 340 °N. As a consequence, the subset for validation of the MPN model is rather small. Data records are only available for the wind sectors 110-155 °N and 240-310 °N. Table 1 presents the performance of the MPN model and, for comparison, the performance of NAM. In addition, the perfomances of the NAM and MPN models obtained previously for the HEXMAX data set (on which the MPN model is based) are listed.[5]

Table 1: Performance of the local MPN aerosol model and NAM in the vicinity of the MPN tower. The row labelled *N* indicates the number of data records available in each wind sector.

	Wdir	*110-340*	*110-155*	*240-310*
	N	*72*	*31*	*39*
λ = 10.6 µm	MPN	2.2	2.8	1.7
	NAM	2.0	2.1	3.2
λ = 4.0 µm	MPN	1.9	2.6	1.4
	NAM	3.9	4.3	3.5
λ = 1.064 µm	MPN	2.6	3.5	2.2
	NAM	3.1	1.9	4.9
$\Sigma\lambda$	MPN	2.2	2.9	1.7
	NAM	2.9	2.6	3.8
$\Sigma\lambda$ HEXMAX	MPN	2.0		
	NAM	4.1		

Table 1 shows that overall the MPN model predicts the aerosol extinction within a factor of 2.2, which is close to the performance of 2.0 reported earlier.[5] Although the present validation is based on a rather small dataset, this is encouraging. It proves that for an arbitrary aerosol sample the MPN model prediction is accurate, within its limits of application. The MPN model predicts the aerosol extinction more accurate than the NAM model, which supports the suggestion that the predictions from general models such as NAM must be carefully interpreted for a polluted coastal area.

The MPN model performs better in the 240-310 °N wind sector. This may reflect that for the original formulation of the model more aerosol data was available for this wind sector (224 hours of sampling) than for the 110-155 °N wind sector (28 hours of sampling),[9] which leads to a reduced accuracy for the latter wind sector. The MAPTIP data set with predominantly easterly winds provides a tool to increase the performance of the MPN model in the 110-155 °N wind sector and to extend the model to wind directions smaller than 110 °N.

3.3 Spatial variations

One of the TNO objectives in MAPTIP was to assess the geographical limits of the validity of the local MPN aerosol model for the North Sea. To this end, Hr.Ms. Tydeman made various treks over the North Sea. To assess the spatial variability, the aerosol data set was partitioned into 4

subsets (cf. Figure 1): a) the vicinity of MPN ; b) the region west of MPN; c) the region north of MPN; and d) the region southwest of MPN.

During the treks of Hr.Ms. Tydeman, only a small amount of data was collected while the wind direction was between 110 and 340 °N (37 data records total). However, these data show that the accuracy of the MPN model decreases when Hr.Ms. Tydeman is not in the vicinity of the MPN tower. The decrease is not surprising, since the transport of aerosol from sources on the continent and in the UK is only taken into account for the immediate vicinity of the MPN tower by virtue of its dependence on wind direction and wind speed. The partitioning in wind direction was made in the first place to account for different source regions. Of course also the fetch, and thus aerosol transport, changes with wind direction. The transport mechanism is further affected by wind speed, as explained by *Van Eijk and De Leeuw*.[9] However, to generalize the MPN model, the empirical equations must be extended with physical descriptions of the transport processes. The Hr.Ms. Tydeman data set has been collected for this purpose.

The limited data set offers few possibilities to assess the spatial variability of the aerosol extinction. To increase the data set it is necessary to extend the MPN model to wind directions smaller than 110 °N and larger than 340 °N. As a zero-order approximation, we have extended the wind sector labelled 'North Sea' (310 - 340 °N) to 310 - 030 °N and the sector 'rural region of Holland' (110 - 155 °N) to 030 - 155 °N.[9] In this way, the MPN model covers the full wind rose.

The performances of the extended MPN and the NAM models for the various regions are presented in Table 2. The table shows that the extended MPN model predicts the aerosol extinction in the vicinity of the MPN tower to within a factor of 3.7, compared to a factor of 2.2 for the original MPN model (cf. Table 1). Clearly, the zero-order extension is too crude and must be refined in the future to take explicitly into account the aerosol advected from Amsterdam (and its harbor) and the industrial area near IJmuiden.

Table 2 supports the conclusion mentioned above that the accuracy of the MPN model decreases outside the vicinity of the MPN tower. The prevailing easterly winds prompt the MPN model to include a large amount of aerosol of continental origin and relatively few aerosol of marine origin, due to the limited fetch. Further away from the coast, the contribution of continental aerosol decrease due to removal processes. As a consequence, the aerosol extinction in this region decreases, especially for the visible and near-IR wavelengths. On the other hand, the contribution of marine aerosol will increase with increasing fetch, leading to larger aerosol extinction, especially for the far-IR wavelengths. The net effect depends on their relative importance. We found that the MPN underestimates the aerosol extinction, in particular for the far-IR wavelengths. This suggests that the effect of the increased concentration of marine aerosol dominates the effect of the decrease in concentration of continental aerosol.

The above reasoning explains why the MPN model prediction is less accurate for the region 'west' compared to the region 'MPN'. For the regions 'north' and 'southwest', other factors are of importance too. The average wind speed encountered by Hr.Ms. Tydeman while in the regions 'north' and 'southwest' was some 4 m/s more than in the regions 'MPN' and 'west'. The wind speed in combination with the

Table 2: Performance of the NAM and extended MPN models for various geographical regions and for the total data set. The column labelled *N* gives the number of data records for each model in each region.

Region	Model	N	10.6 μm	4.0 μm	1.064 μm	Σλ
All	MPN	794	4.1	5.6	4.5	4.7
	NAM	1254	1.9	3.2	2.2	2.4
MPN	MPN	531	3.2	4.2	3.7	3.7
	NAM	888	2.0	3.4	2.3	2.5
West	MPN	63	3.6	6.2	2.6	3.9
	NAM	103	1.9	3.7	3.1	2.8
North	MPN	81	5.6	7.8	5.5	6.2
	NAM	108	1.6	2.0	1.7	1.8
SW	MPN	119	7.8	12.5	9.5	9.8
	NAM	153	1.7	3.3	1.7	2.1

larger fetch must have resulted in an increased concentration of marine aerosol in these regions, as compared to the prediction by the MPN model. Hence, the MPN model substantially underestimates the aerosol extinction in the regions 'north' and 'southwest' resulting in a poor performance. The increased contribution of aerosol of marine origin in the regions 'north' and 'southwest' is also reflected in the performance of the NAM model. This model yields better results for the more marine conditions encountered in these regions. This supports our earlier conclusion[9] that in air masses with a large marine component NAM performs well, even in coastal areas.

3.4 Atmospheric stability

The MAPTIP experiment was characterized by unstable thermal stratifications, with an average ASTD (air-sea temperature difference) of approximately -4 °C. In very unstable conditions, the values of the ASTD reached -9 °C, whereas values of +1 °C were observed in neutral and stable conditions. The effect of the ASTD on the performance of the NAM and MPN aerosol models is assessed in Table 3.

Table 3: Performance of the NAM and MPN aerosol models for very unstable thermal stratification (ASTD < -4 °C), and other thermal stratifications (ASTD > -4 °C).

	ASTD	*> -4 °C*	*< -4 °C*
	N	*415 / 719*	*380 / 536*
λ = 10.6 µm	MPN	3.5	4.7
	NAM	1.7	2.3
λ = 4.0 µm	MPN	5.1	6.0
	NAM	2.5	4.3
λ = 1.064 µm	MPN	3.7	5.2
	NAM	2.0	2.5
$\Sigma\lambda$	MPN	4.1	5.3
	NAM	2.1	2.9

The table shows that the performance of the aerosol models is reduced during very unstable thermal stratification. For the MPN model, this probably reflects that the model is not correctly parameterized for these extreme conditions. The original formulation was based on a data set with ASTD values in the range of -4.5 to +3 °C. The present results show that the ASTD dependence cannot be extrapolated to very unstable conditions.

The reduced performance of the NAM model may be attributed to the meteorological conditions during MAPTIP. In unstable conditions, rising thermals and associated downward motions of colder air parcels cause efficient mixing of the aerosol throughout the boundary layer resulting in a reduced concentration of aerosols at deck level. Opposed to this effect, the whitecap coverage which relates to the aerosol production, becomes larger.[17] During MAPTIP, the production of marine aerosol was low due to the limited fetch in the predominantly easterly winds. Consequently, the NAM model overestimates the amount of aerosol and yields an aerosol extinction that is too high.

3.5 Chemical composition

Impactor samples have been collected during MAPTIP to determine the composition of the aerosol as a function of size and geographical location. Table 4 presents dates and locations of the various samples. The aerosol was collected in 7 size bins, and for each size bin the concentrations of 18 elements in the aerosol have been determined. For the analysis, we have selected 4 elements that are characteristic for aerosol from anthropogenic sources (Cu, Zn, Pb, Se) and 3 elements that are characteristic for marine aerosol (Ca, Sr, Cl). Figures 2 and 3 present the mass distributions. The figures show clearly that the smaller aerosol is predomi-

Table 4: date and location of impactor samples. The asterisk denotes a time on the following day.

Sample	Date	Time	Region
A	24 Oct	01.00 - 12.25	southwest
B	26 Oct	19.15 - 07.30*	MPN
C	28 Oct	20.20 - 07.45*	west
D	30 Oct	07.30 - 19.20	southwest
E	1 Nov	18.30 - 06.30*	MPN

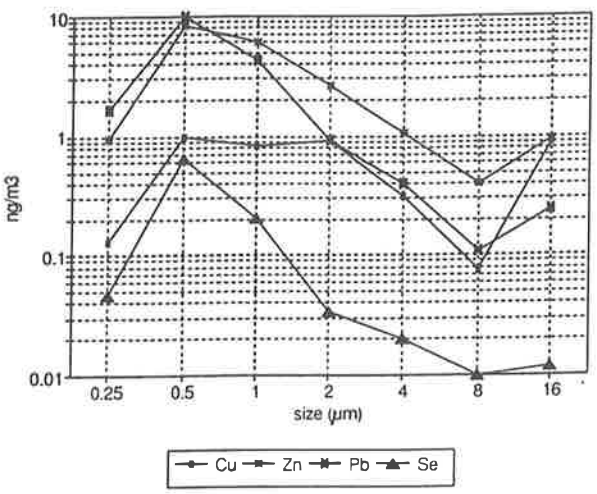

Figure 2: Average mass distribution of elements characteristic for aerosols from anthropogenic sources

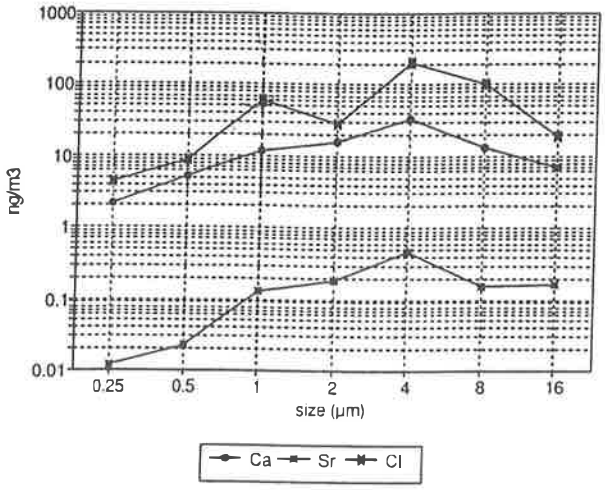

Figure 3: Average mass distribution of elements characteristic for aerosol of marine origin.

Table 5: elemental concentration of aerosol in ng/m^3

	B+E mpn	A+C+D	A+B mpn	C+D+E
Cu	5.1	3.6	1.0	6.3
Zn	15.9	24.7	4.0	31.0
Pb	17.4	17.3	4.7	25.8
Se	0.63	1.15	0.08	1.52
Ca	52.5	114	112.5	74
Sr	0.48	1.18	1.34	0.61
Cl	277.5	52.9	1023	32.3

Table 6: elemental concentration of aerosol in ng/m^3 for consecutive impactor samples

	B mpn	A	E mpn	C+D
Cu	0.7	1.3	9.4	4.8
Zn	3.1	4.9	26.9	33.8
Pb	7.88	1.56	27.0	25.2
Se	0.09	0.07	1.17	1.70
Ca	74	151	31	96
Sr	0.75	1.92	0.21	0.81
Cl	531	1515	24	36.5

nantly of anthropogenic origin, whereas the larger aerosol is of marine origin. This result justifies our approach in the local MPN aerosol model to treat the aerosol at the North Sea as a mixture of smaller aerosol of continental origin and larger aerosol of marine origin.[9]

Table 5 presents results of the elemental analysis of the aerosol samples. For each impactor sample (A-E) the elemental concentrations have been summed over all size bins to yield total concentration per impactor sample. In table 5, we present the average total element concentration for the impactor samples collected near MPN (B+E) and for the samples collected at some distance of the coast (A+C+D). For the latter samples, the fetch was larger in the easterly winds. This should result in a larger concentration of 'marine' elements (Ca, Sr and Cl) and in a smaller concentration of 'anthropogenic' elements (Cu, Zn, Pb and Se). The table shows that the expected behaviour is only observed for 3 of the 7 elements (Cu, Ca and Sr). This is due to a change in the general conditions, which is explained below. If we compare individual impactor samples that were collected consecutively, the results are better in line with the expectation.

Thus, for the assessment of spatial variability we may compare sample B (near MPN) to sample A (southwest), and sample E (near MPN) to samples C+D (west, southwest). The comparison is shown in table 6. The 'marine' elements (Ca, Sr, Cl) now all show the expected increase in concentration for a location at some distance of the MPN tower. For the 'anthropogenic' elements, the expected decrease in concentration for a location at some distance from MPN is only observed in 50% of the cases. Apparently, more data is needed for a firm conclusion about the spatial variability of 'antropogenic' aerosol.

The aerosol content, both physically and chemically, changed markedly during the MAPTIP experiment. This resulted in a large change in visibility in the afternoon of 28 Oct. Prior to this time the visibility was good (mostly larger than 10 nMi), whereas afterwards the visibility did not exceed 10 nMi and heay fog events occurred. The change in visibility could not be associated with a change in wind direction, i.e. with a change in origin of air masses. The change in visibility was confirmed by independent measurements of the visibility at MPN, and by manual observations at Hr.Ms. Tydeman. As mentioned previously, the prevailing wind direction was east during the whole campaign. However, the event changed the chemical composition of the aerosol drastically. This is shown in table 5, where we compare the impactor samples collected before (A+B) and after (C+D+E) the change in visibility. At this time, any explanation is speculative. It is possible that in haze and fog the deposition rate of aerosol of anthropogenic sources is reduced, because the particles are trapped in the fog droplets and stay aloft. This would result in the observed increase in concentration after 28 Oct. The reduction of aerosols of marine origin may be related to the wind speed and wave height. Before 28 Oct, the wind speed varied between 5 and 20 m/s and the wave heights were in

the range 50-150 cm. Afterwards, the wind speed did not exceed 10 m/s and the wave height was typically 40 cm.

4. CONCLUSION

We have presented an analysis of the aerosol extinction, as calculated from aerosol size distributions measured by Hr.Ms. Tydeman during the MAPTIP campaign. The local aerosol model developed previously for the vicinity of the MPN tower was found to yield accurate estimates of the aerosol extinction for data in its range of validity. However, the MPN model in its present form cannot be used for other geographical regions, nor for environmental conditions that are outside the specified range. We intend to use the MAPTIP data set for extension of the MPN model to other environmental conditions (easterly winds, unstable stratification) and other geographical regions (by taking into account the transport of aerosol).

The analysis of the aerosol extinction could not be completed without taking into account the specific conditions of a coastal area, such as limited fetch and contributions of aerosol of continental origin. The development of local models is fruitful, because these models can yield more accurate results than general models, such as NAM. This supports our feeling that there is a need for models, which take into account the physics of aerosol production and transport in the coastal zone.

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