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SHORT COMMUNICATION

Dense Fog in the Netherlands: Composition of the Nuclei that Contribute Most to the Droplet Number Concentration

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ABSTRACT

Dense fogs, with a visibility of less than 200 m, form a traffic hazard. Usually, models describing their formation use observations at the Cabauw super-site in the Netherlands for evaluation. A key parameter is the number of fog droplets and thus the number of aerosol particles on which the fog droplets form, the so-called fog nuclei (FN). No observational data are available for this key microphysical feature. An assumption is that this number scales with the concentration of the hygroscopic aerosol component sulfate. However, in the Netherlands nitrate and organics are the more important components of the total aerosol and thus possibly also of the FN. This short communication provides the first actual data via measurements with an aerosol mass spectrometer—AMS—for a period with dense fog events observed in November 2011. The aerosol in the relevant size range was composed of about half of the hygroscopic ammonium nitrate/sulfate. The other half consisted of organics; the low O/C ratio indicated that these compounds are rather hydrophobic; the hygroscopicity factor kappa of this mix was estimated at 0.3. This value implies that the activation diameter (the lowest diameter of the FN) was at least 150 nm. The mass distribution was converted into a number distribution which showed a sharp decrease as a function of size for diameters above this threshold. This result implies that the vast majority of the FN have diameters to the activation diameter. These smallest FN contained ammonium nitrate as the major hygroscopic compound. Currently, data for other dense fogs are evaluated to search for a possible generality of this finding.

Keywords: AMS; Ammonium nitrate; Organics; Hygroscopicity factor; Activation diameter

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1. Introduction

Fogs form a traffic hazard, especially those with a visibility of less than 200 m, known as dense fog [1]. Fog formation initiates when the relative humidity (RH) surpasses saturation, leading to the condensation of excess water vapor onto aerosol particles. These particles, known as fog nuclei (FN), serve as the nuclei around which fog droplets form. The size of these fog nuclei, denoted as the "activation" diameter (Da), determines their ability to catalyze fog droplet formation. This diameter is governed by both supersaturation and composition of the aerosol particles [2]. While the precise measurement of supersaturation remains challenging, it is worth noting that for dense fogs a supersaturation of 0.1% is an upper limit [3].

In modeling efforts of fog occurrence, there is a major uncertainty in the number of aerosol particles serving as nuclei for the fog droplets that in turn impact the visibility. These FN are assumed to consist of sulfate [4] and ammonium sulfate in particular [5]. The latter authors modeled the formation of fog occurring at the central meteorological site of Cabauw in the Netherlands, which serves as a reference point in general for evaluating fog models [6,7]. The choice for sulfate seems questionable for the Netherlands because the mass concentration of ammonium nitrate and organics in the total aerosol is higher than that of ammonium sulfate [8–10] and this might also be the case for those particles that contribute most to the number of FN and thus droplets.

The chemical composition of fog droplets has been measured for several decades (PoValley Italy: [11] Central California [12] a.o.). The results highlight that ammonium nitrate and sulfate are the most abundant hygroscopic compounds. Nitrate concentrations there were often higher than sulfate. However, the chemical composition of fog droplets is biased due to aqueous chemical reactions and aerosol washout [13]. In order to improve forecasts of fog occurrences, the FN numbers and chemical composition need to be known. Moreover, since Cabauw data are used for the evaluation of the fog models, FN measurements at this site are deemed essential.

Here we present the first experimental information on the composition of the FN with the specific aim to assess the composition of the FN that governs the number of fog droplets. Data stem from a measuring campaign in November 2011 that coincided with an extensive period of dense fog in the Netherlands. Since the majority of the fogs occur in the winter season and especially in mid-fall in the Netherlands [13], an investigation in November seems quite representative.

2. Materials and methods

The national observatory of CESAR-Cabauw in the center-west of the Netherlands was the site at which the measurements were performed. The site is representative of the regional aerosols [9]. Data came from a campaign performed in November 2011. Assessment of dense fog events was done using the visibility data at the site [15]. The criterion for dense fog is a visibility of less than 200 m [16] in at least two consecutive hours to ensure the maturity of these fogs. November 2011 was characterized by the occurrence of several periods of dense fog with a total of 86 hours of these. This is a unique situation because the number of hours with dense fog a year is on average limited to around a hundred hours [4].

An Aerosol Mass Spectrometer (AMS, Aerodyne Research Inc.) measured the non-refractory (material vaporizing around 600 °C under close-to-vacuum conditions) chemical compounds as a function of size. These are the major aerosol compounds in the Netherlands such as ammonium nitrate sulfate and organics [9]. The AMS measures in the size range of 80 to 2000 nm vacuum aerodynamic diameter; a full description of the present AMS is given by Schlag [9]. It should be noted that the vacuum aerodynamic diameter is 1.6 times larger than the mobility diameter [9] in which the FN is expressed in the models.

Air is sampled from a height of 60 m and travels through a tube ending in a manifold in the basement. The air is dried with Nafion dryers to a relative humidity of less than 40% [17]. The air residence time in the tube is sufficient for the complete drying of the droplets so that the fog nuclei are left when the air arrives within the AMS. There is standard a PM10 cut-off impactor at the inlet to remove coarse dust but during fog, it will also remove droplets. For that reason, we did not consider aerosol characteristics during fog events. As a proxy, we used the properties of the aerosol prior to the fog. Air masses after dissipation are most often from a different origin [18] and we did, thus, not consider these as proxy for the aerosol during an event.

3. Results and discussion

As mentioned in the previous section, an AMS continuously monitored the aerosol composition as a function of size at Cabauw from the 7th until the 31st November 2011. During that time seven events of dense fogs occurred from the 13th to the 23rd with a total number of 86 hours of dense fogs with one event almost continuously from the 19th to the 22nd. We assume in the first data evaluation that the FN covers a size-range similar to that of the cloud nuclei (150 to 450 nm) diameter range observed in an earlier research campaign on the composition of cloud nuclei [10] using a supersaturation of 0.1%. The mass concentration of the four major components in this "FN/CCN"-range before or in the few hours of a break in the long fog event is shown in **Table 1**. Prior to all these events, nitrate is clearly the most important compound of the aerosol with concentrations larger than 2 $\mu g.m^{-3}$ except for one event (1.1 $\mu g.m^{-3}$ on 18 November). Sulfate concentrations remain low (< 0.75 $\mu g.m^{-3}$) for all events.

Table 1. Mass concentration of the indicated compounds in aerosol in the "CCN" diameter range of 150–450 nm related to the dense fog events mentioned in the first column.

Starting time	Duration (hrs)	NH_4 ($\mu g.m^{-3}$)	NO_3 ($\mu g.m^{-3}$)	SO_4 ($\mu g.m^{-3}$)	Org ($\mu g.m^{-3}$)
13-11-2011 16:00	13	0.5	2.3	0.5	1.8
17-11-2011 03:00	4	0.3	2.2	0.7	1.5
18-11-2021 20:00	5	0.2	1.1	0.3	0.7
19-11-2011 18:00	28	0.4	2.1	0.35	1.8
20-11-2011 00:00	31	1.45	2.3	0.55	1.45
23-11-2011 05:00	4	0.45	2.5	0.75	1.6

Source: Schlag [9].

To estimate the FN size range for the events, we made use of the total mass distribution as a function of size. Using an average density of 1.6 $g.cm^{-3}$ [9] and assuming the sphericity of each particle, the mass size distribution was translated to a number size distribution (**Figure 1**) consistent with the spectrum measured with Scanning Mobility Particle Sizers during previous campaigns [18,20]. Virtually all particles have diameters less than 500 nm, as seen in **Figure 1**.

According to Petters and Kreidenweis [2] and based on the chemical composition given in **Table 1**, the aerosol hygroscopicity factor is estimated around 0.3 corresponding to an activation diameter (Da) of 150 nm assuming a supersaturation of 0.1%. The actual supersaturation is probably overestimated as observed during the ParisFog study [19]. At a supersaturation of 0.05%, the activation diameter Da would be larger (around 220 nm) leading to a decrease in the FN concentration. Therefore, the FN concentration estimated using a supersaturation of 0.1% is the highest threshold.

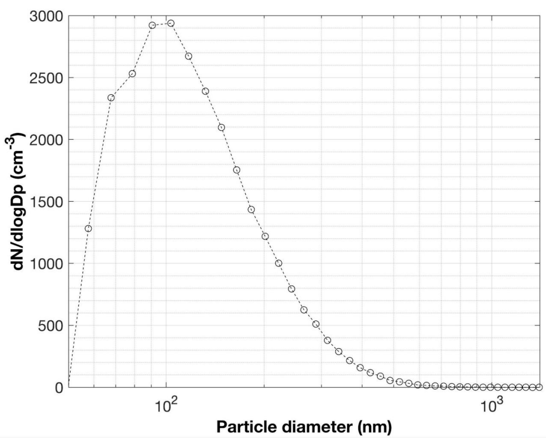

Figure 1. Number concentration of aerosol as a function of size deduced from the mass distribution of the main components on 13 November 2011 before the start of the first dense fog in the series; see **Table 1**.

As can be seen in **Figure 1** the number of particles with a size larger than the activation diameter (of 150 or 220 nm) rapidly decreases with size. This means that the most numerous FN are those with a diameter close to the activation diameter. Those FN contained ammonium nitrate as the dominant hygroscopic compound.

The finding of a prominence of nitrate for the number of droplets in dense fog is currently being further evaluated with data from other events; we already noticed that the chemical data are less detailed but there is information on the microphysics, like droplet and CCN number concentrations and their relation with the composition of the FN that determine those figures. We also suggest using our approach for an evaluation of similar AMS data for fogs in the Po-Valley [22].

4. Conclusions

In a series of dense fog in November 2011 at the CESAR-Cabauw super site (the Netherlands), the composition of fog nuclei as a function of size was determined. This is supplementary information to the abundant macrophysical data base there used for the evaluation of fog models [1,5]. We found that in the smallest most numerous FN that contribute most to the droplet number, ammonium nitrate was the major hygroscopic compound. Whether this finding is of a general nature has to be assessed in other dense fogs that we are currently evaluating.

Author Contributions

Conceptualization, H.tB., P.S. and S.C.; methodology, H.tB.; formal analysis, H.tB. P.S.; investigation, H.tB. and S.C.; writing-original draft preparation, H.tB. and S.C.; writing-review and editing, H.tB. and S.C. All authors have read and agreed to the published version of the manuscript.

Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data are available on request.

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