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Visibility in the Netherlands during New Year's fireworks: The role of soot and salty aerosol products

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ABSTRACT

The visibility on New Year's nights in the Netherlands is low during stagnant weather. This is due to the scattering and absorption of light by the aerosol-smoke from the fireworks. We made an assessment of the responsible aerosol-species. The investigation took place during the New Year's night of 2009. Measurements were made at a regional site in the centre of the country away from specific local sources. An Integrating Nephelometer measured the light-scattering by the inherent compounds after removal of water from the aerosol by drying the air. The actual light-scattering was determined in an open-air scatterometer; it was a factor of five higher than the "dry" value. The difference in actual and "dry" light-scattering can only be explained by water–uptake of the salty hygroscopic components of the aerosol. This hypothesis is substantiated by measurements of the composition of the aerosol. The size-dependent concentrations of the salty ionic species were determined on-line with a MARGA-"sizer". These components were for a large part in particles in the size range that most effectively scatter light. The "dry" light-scattering was exerted by the inorganic salt components and the sooty carbonaceous material alike. However, the salty products from the fireworks are hygroscopic and take up water at the high relative humidities occurring that night. This explains the fivefold larger light-scattering by the wet ambient aerosol as compared to that by the dry aerosol in the integrating nephelometer. The visibility, which is the inverse of the open-air scattering, is thus indirectly governed by the salty products of the fireworks due to their uptake of water. Under stagnant weather conditions during New Year's nights in the Netherlands both the aerosol concentrations and the relative humidity are high; this implies that the ionic species govern the low visibilities in general, be it via their uptake of water.

1. Introduction

The fireworks during New Year's night in the Netherlands are rather unique because a better part is ignited in the streets. This gives rise to high concentrations of aerosol-smoke at street level and especially during stagnant weather this is accompanied by low visibilities. The visibility is reduced by the scattering and absorption of visible light. The present study was a sequel to an investigation in the previous year which showed that half of the aerosol consisted of sooty material with the rest inorganic salts (ten Brink et al., 2017).

The visibility may not be affected in the same way by the mentioned two types of compounds, because the scattering is very sensitive to the size of the particles. Furthermore the salts are hygroscopic and could take up water by which the aerosol particles grow in size and scatter more light. We investigated this hypothesis by assessing the relation between the size/composition of the aerosol and the visibility; this relation is briefly introduced in the next subsection. The light-absorption by the soot was independently tested.

1.1. Visibility, light-scattering and hygroscopic growth

The visibility is defined as the maximum distance at which objects are discernible; it is known as visual range in aviation. The visual range is reduced because aerosol particles scatter the light on its way from the object to the observer. This distance is less at higher aerosol concentrations. Originally the visibility was determined with a long-path transmissometer and the light-reduction measured was extrapolated to the distance at which the light intensity would decrease to a few percent of the original value. Nowadays another method is used. The light scattered by the aerosol is measured with a scatterometer. The scattering is quantified via the light-scattering coefficient, which is a measure for the relative decrease in intensity of the light per meter. Its

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unit is m^{-1}. The relation between visibility and measured light-scattering (coefficient) is standardised (KNMI, 2005) via the following inverse relation:

$$Vis = 3.0 / b_{scat} \quad (1)$$

with visibility (Vis) in meters and the light-scattering coefficient (b_{scat}) in the unit m^{-1}. In the manual the Meteorological Optical Range is the official term for the visibility.

Equation (1) is described in detail in the classical textbook on aerosol science “Smoke, dust and haze” (Friedlander, 2000). There a full chapter is devoted to the light-scattering by aerosol particles; the book also describes the approach of assessing the contribution by the various aerosol compounds to the light-scattering. First, the light-scattering coefficient is determined in a so-called integrating nephelometer. Integrating means that all scattered light is collected and not at a given angle for which the signal is a complex function of particle size. Before the air arrives in the measuring section of the instrument it is heated to remove the water from the aerosol. The reason is that the aerosol-water would otherwise contribute to the light-scattering, while its concentration is not determined.

Graphs in textbooks (e.g., Hidy, 1984) provide the values for the scattering coefficient as a function of mass at a given aerosol size. The following feature is also explained there. A rather steep maximum occurs at a particle size that corresponds to the wavelength of light, which for visible light is around a diameter of 0.50 μm. When compounds are preferentially present in aerosol of this size they contribute most to the light-scattering. This is the reason that we specifically measured the mass concentration of the compounds in the size range of 0.2–2 μm. This approach was also followed in an earlier study to assess the contribution of the major components of ambient aerosol to the light-scattering (ten Brink et al., 1997). To put this in further perspective: a $PM_{2.5}$ mass concentration of 23 μg m^{-3} corresponded to a light-scattering coefficient of 1.0×10^{-4} m^{-1} (ten Brink et al., 1996).

The actual light-scattering in the atmosphere is exerted by particles that contain water. This water is taken up by hygroscopic inorganic salt compounds like ammonium nitrate, even at a moderate relative humidity of 60%. The water uptake by the hygroscopic compounds of ambient aerosol and its effect on the light-scattering has been studied in a special set-up in our laboratory in which the relative humidity can be varied (ten Brink et al., 2000; Veefkind et al., 1996). The average increase in light-scattering was a factor of two at an RH of 85%. Also the decrease in light-scattering when the air is dried was assessed (Dougle et al., 1998). This combined expertise was used in the evaluation of the present measurements.

The mass concentrations of the hygroscopic ionic compounds were determined as a function of size with the automated on-line MARGA-“sizer” (ten Brink et al., 2007). The size classification is discussed in the next section. The operating principle of the MARGA itself is summarised here. At the inlet steam is injected into the sampling air. This condenses onto the aerosol particles that grow to droplets, which are subsequently collected in a cyclone. The soluble fraction of the compounds dissolves in the water. The drain from the cyclone is fed to an ion-chromatograph for direct on-line chemical analysis of the dissolved ions. An important aspect is that the method representatively collects the semi-volatile ammonium nitrate. This is important because its concentration was high and it strongly contributes to the light-scattering at the measuring site. This contribution has to be subtracted from the total light-scattering to obtain the light-scattering exerted by the aerosol from the fireworks.

2. Experimental

2.1. Campaign

The measuring campaign was part of a project to assess the size and composition of the aerosol in the Netherlands in a number of months in the year 2008. The study was extended for an extra day for the present investigation of the aerosol during New Year's night of 2009. The measurements were performed in the CESAR-observatory at Cabauw in the centre of the Netherlands, between the cities of Utrecht and Rotterdam. The aerosol at the site originates from a mix of fireworks, instead of being from a specific type of fire-crackers at a street location. Furthermore visibility data were available.

Fog was forecast for New Year's night, but did not develop. The weather was stagnant though with a light wind from a north to easterly direction with velocities in the class of 1 m s^{-1}. These and other hourly data, including those on relative humidity (RH) and visibility, were obtained from the standard instrumentation of which the data are given in tables of the national network (KNMI, 2009) for the site.

Note. The RH that night was in excess of 95%. With an uncertainty in hygrometers of 5%, this means that the actual value can be anywhere between 95% and 100%. The associated extra light-scattering by water taken up by hygroscopic components of the aerosol has an uncertainty of more than an order of magnitude. Reversely, the larger value of the open-air light-scattering measured as compared to the “dry” light-scattering is a better measure. The five-fold increase found in section 3.1 is due to the uptake of water by the hygroscopic particles and occurs when the RH is 97–98%. This uncertainty is substantially less than that of the tabulated RH.

2.2. Instrumentation

2.2.1. Light-scattering and visibility

An Integrating Nephelometer (TSI 3563) was used to measure the aerosol light-scattering. In this instrument the sampling air is dried by a heated inlet to a reference low relative humidity of 40%; at this humidity the water content is negligible and the aerosol is called “dry” (ten Brink et al., 2000). The Integrating Nephelometer was located on a measuring platform at a height of 60 m. This means that the aerosol as encountered there is not of a local origin and thus representative for an average more regional aerosol. The instrument provides data with a 5 min resolution, with zeroing occurring every tenth time-step.

Values for the directly measured visibility were obtained from the data tabulation of the national meteorological network (KNMI, 2009); it is termed visual range in that database. The data are the average and maximum and minimum values in an hourly interval. The visibility is derived from an open-air light-scattering instrument (KNMI, 2005).

2.2.2. Composition as a function of size; the MARGA-sizer

Measurements of the concentration of the major ions were made with the MARGA-sizer. This instrument was located inside the meteo-tower of the CESAR site at Cabauw. Due to space restrictions the instrument needed a long sampling line. The advantage was that the air in the line is warmed up and that the aerosol is dried and thus size-classified in the dry state.

The MARGA-sizer measures the ionic components as a function of size and has been described before in detail with regards to monitoring of the standard inorganic species in the Netherlands, viz. nitrate, sulphate, ammonium (ten Brink et al., 2009; ten Brink et al., 2007). Its long-term performance was compared with that of a standard Aerodyne Aerosol Mass Spectrometer at the site (Mensah et al., 2012). We will summarise the main characteristics of the size-classification of the MARGA-sizer because of its importance for the light-scattering. Furthermore the novel detection of the cations is discussed.

The size-classification occurs via a set of parallel impactors. The impactors have cut-off diameters of 2.0, 0.1, 0.56, 0.32 and 0.18 μm. Aerosol particles behind these impactors have diameters smaller than these respective values. A total cycle of sampling and analysis takes 2 h. The characteristics of the impactors were checked after the campaign in the lab. The measured cut-off diameters of the impactors were close to the factory value. However, a constant fraction of around 7% of the