What Causes Observed Fog Trends: Air Quality or Climate Change?

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ABSTRACT

Fog is a situation when the visual range, which is the horizontal visibility, is reduced to less than 1000 m near the Earth’s surface by the presence of cloud droplets. Fog trend analyses are reported in the literature for hundreds of stations worldwide, the majority of which showing a considerable reduction of fog. Although fog is often associated with conditions at which cloud condensation nuclei had been activated at rH (relative humidity) > 100% and rapid growth had lead to the formation of fog droplets, this study focusses on urban air masses and conditions when rH is just below 100%. Mie scattering analysis shows that fog can form under such conditions and the reduction of the visual range is mainly caused by submicron aerosol particles which grow to diameters around 1 µm through deliquescence. The liquid water content itself is poorly correlated with the visual range. Assuming equilibrium conditions, both an increase of the air temperature and a reduction of the aerosol particle concentration lead to reductions of fog. In our example case, the increment for a temperature increase by 0.1°C had about the same effect as the reduction of aerosol concentrations by 10%. Care must be taken in projecting this result to actual conditions because the system is non-linear. However, physical evidence is presented which confirms that both climate change and an improvement of air quality are mechanisms that can contribute to the reduction of fog.

Keywords: Fog trends; Air quality; Air pollution; Visibility; Visual range.

INTRODUCTION

Fog as a physical phenomenon in the atmospheric boundary layer has various implications for atmospheric and environmental research as well as for humans’ life. Foggy conditions change the radiation transfer, leading to enhanced reflection of solar light at the fog layer’s surface and to a decrease of radiative surface cooling. Fog impacts ecosystem processes through the reduction of solar light, through the formation of liquid surfaces on the vegetation, and through deposition of nutrients and pollutants. The reduction of the horizontal visibility in fog exerts a huge influence on transport systems in air, on streets, and on open water. Further, the presence of fog may be very beneficial in cases when fog can be collected for freshwater production in some semi-arid, subtropical climates.

There is vast evidence in the scientific literature that the frequency of fog events and the intensity of fog exhibit significant changes over time. The literature analysis in the following paragraphs shows that the majority of stations worldwide show a decrease of fog. Changes of the regional climatic conditions such as the urban heat island (UHI) effect or changes of predominant regional circulation patterns may lead to increasing air temperatures and thus to a decrease of the relative humidity (rH) in the air. This argument is plausible as long as there are no feedback mechanisms overriding the sheer temperature effect. The change of the number of cloud condensation nuclei (CCN) in air has been mentioned as another potential cause of fog trends. However, the relevant physical processes behind this reasoning have not been discussed so far. Can, for example, a decrease of the number concentration of CCN cause a decrease of fog? If yes, how strong is this effect likely to be in comparison to a temperature increase? It is the scope of this study to answer these questions based on deliquescence and Mie scattering computations with various air masses. The concentrations of CCN and the temperature will be modulated to identify their impact on the visibility. Can global change cause a global decrease of fog?

Fog trends over several decades have been analyzed for over 1000 stations worldwide. For most stations, a significant decrease of fog has been identified, while some stations show no detectable trends or positive ones. The techniques to detect fog and to analyze trends on the basis of the respective data sets vary widely. In many cases, eye-observations of the visual range at certain times of the day are applied. The...
Horizontal visibility is typically classified but the employed classes vary from place to place or even over time within the observation period. In other cases, the visual range is measured with visibility meters that use the forward scattering of infrared light. The available data sets from all over the world are thus very heterogeneous. It is not the scope of this analysis to provide a full review of applied techniques, metrics, and statistical tools, but we will briefly overview the existing literature on fog trends. To simplify the wording, we state that “fog decreases” if the visibility (which is the visual range $x_v$) during a foggy situation increases or if the frequency of the occurrence of fog decreases. An “increase of fog” is defined correspondingly, both phenomena are “fog trends”.

Vautard et al. (2009) analyzed meteorological data of over 300 stations throughout Europe and found that fog decreased over the period of 30 years. A correlation with the emissions of sulfur dioxide was stated, suggesting a significant contribution of air quality improvements, although the physical processes behind that mechanism are not further discussed. Similarly, Muraca et al. (2001) analyzed fog occurrence throughout Canada and found that there was a strong decrease at most of over 184 stations in the time period from 1971 to 1999. The potential causes for the observed trends are not discussed. The results are only in partial agreement with those of Hanesiak and Wang (2005), who found a decrease of fog over the period from 1953 to 2004 only for the northeast, but not for the southwest of the Canadian Arctic. Mount Washington in the northeastern USA exhibits an increase of fog during the time period since the 1930s (Seidel et al., 2007). In the southeastern USA, 12 of 26 analyzed stations showed a significant decrease of fog over the period from the late 1940s until the early 2000s, while only two exhibited positive trends (Forthun et al., 2006). The possible causes of increasing temperatures, decreasing relative humidity, and aerosol composition and concentrations are discussed, however, no conclusive picture about the involved processes could be drawn. For the midwestern USA and based on a database from 1948 until 1996, Westcott and Isard (2001) and Westcott (2002) found a decrease of fog in the region around the Grates Lakes, whereas many stations in the States of Iowa, Missouri and Illinois showed increases. Fog trends in the Greater Los Angeles Area were analyzed by Witiw and LaDochy (2004, 2008) and LaDochy and Witiw (2007, 2012) for the time period 1948–2004. They identified a clear decrease of hours of dense fog, particularly for the stations near the Pacific coast (Fig. 1). Influences of the Pacific Decadal Oscillation index and the El-Niño Southern Oscillation could be found, these phenomena could however not explain the magnitude of the observed trends. Whether increasing temperatures are more important or the improvement of air quality, is discussed by the authors but could not be conclusively answered. Feedback mechanisms between air quality and local climate complicate the situation. In a more recent study, Williams et al. (2015) found that ongoing urbanization, specifically the influence of the UHI is responsible for the decrease of fog in southern California. A satellite data analysis showed that the fog frequency also decreased strongly in the California Central Valley (Baldocchi and Waller, 2014). The potential contributions of climate change, increase of air quality and agricultural practices are mentioned, however not analyzed in detail.

Araujo et al. (2001) and Goncalves et al. (2004, 2007, 2008) analyzed the development of fog frequency in the metropolitan region São Paulo over the time period 1933–2005 and detected a strong decrease, although there was an interim phase of increased fog in the late 1970s (Fig. 1).

**Fig. 1.** Longterm fog data at selected sites. Annual number of fog events in São Paulo (full black line, left axis, Goncalves et al., 2008), annual number of hours of dense fog at Los Angeles International Airport (bold grey line, left axis, Witiw and LaDochy, 2008), and average fog frequency (%) of 31 stations mainly in Pakistan and India with a significant trend (broken line, right axis, Syed et al., 2012).
is argued that an increase of the air temperature and the resulting decrease of the relative humidity is the most likely cause for the observed trend, which itself is caused by global change and an intensification of the UHI. For Argentina, an analysis of data from 16 stations between 1951 and 1990 showed no significant trend of fog for the rural sites, however, the urban sites of Buenos Aires show a strong decrease, which is discussed to be the effect of increasing urbanization and the increase of the UHI (Hoffmann and Nuñez, 2001).

In China, an analysis of data of over 600 stations over the time period 1961–2003 showed an overall remarkable downward trend of fog (Chen et al., 2006). The strongest decreases were observed for western China, northern China, the Yunnan Guizhou Plateau, and parts of Southeast China. However, rather small regions of increasing fog were also identified in eastern Sichuan, northern Hubei, northern China, and the lower reaches of the Yellow River. Temperature increases are identified by the authors as causes for the observed fog trends on the basis of statistical analyses. On the other hand, Han et al. (2015) hypothesize on the basis of urban data sets in the North China Plain covering the period 1980–2010 that their observed increase of fog is due to increasing emissions of air pollutants. Fu et al. (2014) found for the North China Plain that fog increased before 1995, was more or less stagnant from 1995 to 2003, and decreases significantly since then. They argue that the decrease of fog occurred mainly during the winter (heating period) and that an increase of air quality since 2003 is the main cause for the decrease of fog. A similar pattern with increasing fog in the 1970s and 1980s and a decreasing trend after 1999 was observed by Quan et al. (2011). Ding and Liu (2014) argue for an observed decrease of fog in China after 1990 that it is a decrease of rH that causes that trend.

Belorid et al. (2015) found significant decreases of fog at 10 of the 23 stations analyzed throughout South Korea. Mainly radiation fog was affected by the trend, and ongoing urbanization with a decrease of the nightly radiative cooling rates was identified as most likely cause. In Japan, decreasing fog trends are reported for several stations, ongoing urbanization (Yamashita and Tsuji, 2007) and systematic changes of the regional circulation patterns (Sugimoto et al. 2013) are discussed to have contributed or even caused the observed trends.

Al-Fenadi (2001) shows that in Libya, some stations show upwards trends and some other stations downwards trends, while the year-to-year variability is high at all sites. No analysis of possible causes is presented. Van Schalkwyk (2011) found a decreasing trend of fog at the airport of Cape Town, South Africa, for the period 1978–2008. Regional circulation patterns are discussed as possible causes, although no conclusive arguments are presented. The same applies for the McMurdo station on Ross Island, Antarctica (Lazzara, 2008) during the time period 1973–2007.

Amiranashvili et al. (1997, 1998, 2001, 2007) analyzed fog trends at 70 stations in Georgia for the time period 1936–1990. About half of the stations showed a decreasing trend of fog while five stations exhibited an increase of fog. The stations with a positive trend were all located at altitudes between 1000 and 2000 m above sea level (asl). Statistical analyses with meteorological parameters were done. Interestingly, the strongest correlation was found between a decrease of the number of fog days and an increase of aerosol concentrations.

Nomoto (2003) analyzed fog trends of over 50 stations in Thailand and the period 1951–1995. A decrease of fog was found throughout the country. Increases of the temperatures and decreases of humidity due to deforestation and urbanization were discussed as possible causes. In Pakistan, India, and Bangladesh, 82 sites, most located in urban areas, were analyzed for fog occurrence in the years 1976–2010 (Syed et al., 2012). Significant trends of fog were mostly detected in a blanket from Pakistan over India to Bangladesh running almost parallel to the Himalayan mountains. The trends in this region are positive (increasing fog, Fig. 1) and the authors discuss as possible causes land use change (e.g., irrigation), global change with varying temperature, humidity and regional circulation regimes, and atmospheric aerosol concentration changes. Rahimi (2012) analyzed data from over 100 stations in Iran for the period 1985–2005. Most observed trends were negative, only three stations exhibited positive trends in fog. Increasing temperatures were discussed as possible causes for the observed trends.

METHODS

Fog is a situation when the near-surface horizontal visibility is reduced by deliquesced particles (cloud droplets) to 1000 m or less. Whenever the relative humidity (rH) of an atmospheric air mass close to the Earth’s surface reaches over 100%, the conditions are favorable to form fog. In other words, the concentration of cloud condensation nuclei (CCN) is always sufficient to enable the formation of fog at supersaturations of only a few 10’s of a percent above 100%. In this study, we analyze rather specific situations of fog that may form at rH < 100%, when deliquescence of particles is intense enough to reduce the horizontal visibility to less than 1000 m. The conditions we study are somewhat at the verge between haze and fog. Haze describes conditions in the atmosphere with a reduction of the horizontal visibility to less than a few km, but not to less than 1 km, by dry particles or by deliquesced particles or both, and during conditions of high relative humidity, though clearly below 100% (Quan et al., 2011). In a given air mass, the conditions may change from haze to fog when the relative humidity increases to near 100% and above. The classic Köhler theory (e.g., Seinfeld and Pandis, 2006) describes the deliquescence of particles at rH below and above 100%. In this contribution, we refer exclusively to the hygroscopic properties of inorganic compounds in particles and droplets, although organic compounds may play an important role in droplet physics as well (e.g., Sorjamaa et al., 2004). The effects of organic compounds such as surfactants are much less well described than those of many inorganic compounds. Fig. 2 shows 7 examples of particles containing various amounts of ammonium sulfate. This salt is very hygroscopic and deliquescent, i.e., uptake of water, occurs at rH > 79.5% (Seinfeld and Pandis, 2006). Ammonium sulfate particles
Fig. 2. Köhler curves for (NH₄)₂SO₄ particles with dry diameters $D_{p,dry}$ of 0.03, 0.05, 0.075, 0.1, 0.15, 0.2, and 0.25 µm (from left to right) at 20°C. The relative humidity (rH) is shown as function of the respective wet diameters of particles, $D_{p,wet}$. The bold line interpolates rH between the respective critical diameters $D_{p,crit}$ of the droplets. It intersects an individual Köhler curve at its $D_{p,crit}$ and its rH of critical supersaturation (for example at $D_{p,wet} = 0.348$ µm and rH = 100.41% for the particle with $D_{p,dry} = 0.05$ µm, second line from left). Note that the intersection between the bold line and the Köhler curves of the two largest particles are outside the displayed range at $D_{p,wet} = 2.78$ µm and at $D_{p,wet} = 3.89$ µm, respectively. Any particle is non-activated as long as $D_{p,wet} < D_{p,crit}$.

would thus be typical CCNs. At rH = 98%, the diameters of the particles have already increased by factors between 2.4 and 3.1 for the particles shown in Fig. 2 and grow further with increasing rH as shown in Fig. 2. The growth rate (diameter change per change of rH) is larger for the larger particles. If supersaturation occurs, i.e., rH > 100%, particle sizes may reach their critical diameters $D_{p,crit}$, which corresponds to the maximum point of the individual particle’s Köhler curve and the respective level of critical supersaturation. It is evident from Fig. 2 that the larger particles require less supersaturation as compared to the smaller ones. If large particles are present, the activation may start at very low levels of supersaturation and rapid growth is initiated. Once some large particles reached their individual $D_{p,crit}$, their fast growth will lead to a decrease of rH in the gas phase and the internal mixing scales within the cloud will determine how long smaller particles will still be able to grow through condensation at higher rH’s within their respective microenvironments. The description of droplet dynamics, i.e., the concurrent growth of droplets of different sizes and chemical composition under such conditions of rH > 100% with time, requires a turbulent transport model for water vapor and phase transition. The application of such a model (e.g., Bott and Trautmann, 2002) is outside the scope of this contribution. We focus our view on equilibrium conditions at rH $\leq$ 100% and therefore exclusively to non-activated particles in a stable condition. We apply a simple box model in which no transport limitation of water vapor in the gas phase, or between the gas and liquid phases, was allowed. The model consists of three steps.

In the first step, the air masses under consideration are under closed conditions with respect to the aerosol number concentration and chemical composition. No collision and coagulation between individual particles occurred. We used the relative humidity (rH) as the dimension along which the diameters of individual droplets vary according to the Köhler theory. The temperature is set constant at 20°C, which means that the increase of rH is implicitly assumed to happen as a result of increasing water content in the box. As there is no simple solution to calculate the diameter of a droplet as function of rH, we produced look-up tables to assign a diameter to each particle at each rH of interest. The summation of all particles provides a size distribution of deliquesced particles at each rH.

Our analysis is based on five air masses. Two are taken from a textbook and a modelling guideline, and three were parameterized from literature information and represent simplified scenarios from urban environments. The SP30 case represents 30% of the “urban” scenario of Seinfeld and Pandis (2006), which means that 30% of the particles in each size mode of the respective Seinfeld and Pandis (2006) distribution are assumed to consist of (NH₄)₂SO₄, the remainder particles are assumed to be non-deliquescent and thus do not appear in Table 1 and Fig. 3. The R2013 air mass is taken from Rajesh (2013). Only the water soluble mode II is used and assumed to consist of (NH₄)₂SO₄, while mode I (soot) and mode III (insoluble compounds) are neglected. The London aerosol was estimated from data given by Harrison et al. (2012) for their Regents Park site. Modes I and II are set to consist by 60% of (NH₄)₂SO₄ and mode III consists by 20% of NaNO₃ and by 80% of NaCl, respectively. The remainder particles (40% of modes I and II) are assumed to be non-deliquescent and thus do not appear in Table 1 and Fig. 3. The São Paulo air mass was parameterized after data from Ynoue and Andrade...
Table 1. Parameterization of aerosol size distributions and compositions representing typical urban scenarios. The size distribution consist of up to three lognormal modes. Each mode is indicated with the number of particles \( N \) per cm\(^3\), the \( \log_{10} \) of the mode diameter in µm (\( \log D_m \)) and its standard deviation \( \log \sigma \). See text for air mass identification and more details. PM\(_{10}\) is the total mass of deliquescent particles with diameters up to 10 µm and assuming a particle density of 1.7 g cm\(^{-3}\). Note that only the water soluble part of the particles is parameterized and listed.

<table>
<thead>
<tr>
<th>air mass</th>
<th>mode I</th>
<th>mode II</th>
<th>mode III</th>
<th>PM(_{10})</th>
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</thead>
<tbody>
<tr>
<td></td>
<td>( N ) log ( D_m ) log ( \sigma )</td>
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</tr>
<tr>
<td>SP30</td>
<td>29790 –1.886 0.245</td>
<td>333 –1.854 0.666</td>
<td>10920 –1.301 0.337</td>
<td>30.7</td>
</tr>
<tr>
<td>R2013</td>
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<td>28000 –1.373 0.350</td>
<td>0 – –</td>
<td>35.5</td>
</tr>
<tr>
<td>London</td>
<td>3300 –1.523 0.220</td>
<td>3420 –1.046 0.220</td>
<td>3 0.000 0.210</td>
<td>14.9</td>
</tr>
<tr>
<td>São Paulo</td>
<td>5800 –1.398 0.350</td>
<td>420 –0.886 0.270</td>
<td>0.9 –0.222 0.380</td>
<td>14.8</td>
</tr>
<tr>
<td>China/Taiwan</td>
<td>19950 –1.398 0.34</td>
<td>200 –0.523 0.200</td>
<td>2.4 0.176 0.220</td>
<td>52.9</td>
</tr>
</tbody>
</table>

Fig. 3. Number size distributions (top panel) and mass size distributions (bottom panel) of the 5 air masses parameterized for this study (Table 1, see text for air mass identification and more details): SP30 (bold grey broken lines), R2013 (dashed-dotted lines), London (bold short-dashed lines), São Paulo (thin full lines), and China/Taiwan (grey bold full lines). The x-axes show the diameters \( D_{p, dry} \) of the dry particles in their non-deliquesced state. Note that only the water soluble part of the particles is parameterized and shown.

(2004). In their study, there was a lot of unidentified material and the relative contribution of ammonium sulfate is quite small. Therefore, mode I consisted by 10% of \( (NH_4)_2SO_4 \) and by another 10% of NaCl in our study. Mode II consists by 20% of \( (NH_4)_2SO_4 \) and mode III by 10% of NaNO\(_3\). The remainder (80% of modes I and II and 90% of mode III) are neglected in Table 1 and Fig. 3. Finally, we parameterized an air mass representative for the eastern mainland China region including Bejing, the larger Bejing area, the Pearl River Delta region, Shanghai, and western Taiwan. The aerosol distribution and composition was estimated after Tsai and Cheng (1999), Cheng and Tsai (2000), Guo et al. (2010), Yuan et al. (2012), Li et al. (2013) and Tao et al. (2014) for urban sites in mainland China and Taiwan. All three modes (Table 1) were set to consist by 29% of \( (NH_4)_2SO_4 \) and by 28% of NaNO\(_3\). Therefore, 43% of all particles do
not appear in Table 1 and Fig. 3. Number size distributions and mass size distributions of the five air masses are shown in Fig. 3. Note that the largest PM$_{10}$ concentration for the China/Taiwan air mass (52.9 µg m$^{-3}$) results from a small number (2.4 cm$^{-3}$) of relatively large (D$_m = 1.5$ µm) particles.

In step 1 of the model, we describe the increase of particle diameters with increasing rH (at rH > 79.5%). Dry particles with dry diameters D$_{p,dry}$ ≤ 10 µm (corresponding to PM$_{10}$) were employed. Again, equilibrium was assumed to be fully established at each rH. As a response to the increase of rH, the deliquescent particles became liquid droplets and grew significantly (see Fig. 2). The simulation was stopped when rH = 100% was reached. In addition, the growth of individual droplets was stopped when they had reached a wet diameter D$_{p,wet}$ of 100 µm, which occurred, for example, for (NH$_4$)$_2$SO$_4$ particles with D$_{p,dry}$ = 3.16 µm at rH > 99.9999% or for those with D$_{p,dry}$ = 10 µm at rH > 99.9326%.

The LWC [mg m$^{-3}$] of the air mass was computed from the volumes and number concentrations of droplets in the air mass as function of their size D$_{p,wet}$, and assuming a water density $\rho$ of 1 g cm$^{-3}$.

$$ LWC = \int_0^{100} V_{p,wet} \cdot N_{p,wet} \cdot \rho \cdot dD_{p,wet} $$  \hspace{1cm} (1)

In a second step, the reduction of the horizontal visual range $x_v$ as a result of Mie scattering of the droplets is computed. Whenever the resulting $x_v$ falls below 1000 m, the conditions are classified as fog according to the World Meteorological Organization’s (WMO) definition. Note that any impairment of $x_v$ due to Raleigh scattering is not considered in this contribution. Raleigh scattering may also result from air pollution such as aerosol particles and gases and reduces $x_v$, however, reductions to $x_v < 1000$ m through that process are quite unusual and not within the scope of this study.

We assumed that light absorption by water droplets is negligible, the imaginary refractive indexes of the droplets are zero, so that their scattering efficiencies equal the extinction efficiencies. The dimensionless Mie scattering efficiencies of individual droplets of D$_{p,wet}$ were computed for a light wave length of $\lambda = 550$ nm with the software provided by Scott Prahl at the website http://omlc.org/calc/mie_calc.html (last access 21 May 2015). Again, a lookup table was produced, this time between the droplet diameters D$_{p,wet}$ and the scattering efficiency $Q_{scat_{p,wet}}$. Note that there is a very pronounced maximum of $Q_{scat_{p,wet}}$ for droplets with diameters just above D$_{p,wet} = 1$ µm and that the scattering efficiencies of large droplets is much lower. We further assumed that the total scattering efficiency of all droplets in an air mass is the sum of individual scattering efficiencies of these droplets (Seinfeld and Pandis, 2006). From $Q_{scat_{p,wet}}$, the projected area $A_{p,wet} = \pi (D_{p,wet})^2$ of individual droplets and their number concentration in air $N_{p,wet}$ [cm$^{-3}$], the extinction coefficient $b_{ext}$ [m$^{-1}$] of the respective air mass was computed as

$$ b_{ext} = \int_0^{100} A_{p,wet} \cdot N_{p,wet} \cdot Q_{scat_{p,wet}} \cdot dD_{p,wet} $$  \hspace{1cm} (2)

Using $b_{ext}$, the visual range $x_v$ was computed with a common form of the Koschmieder equation (Seinfeld and Pandis, 2006) as

$$ x_v = \frac{3.912}{b_{ext}} $$  \hspace{1cm} (3)

With steps 1 and 2 of the model, we can describe the change of the particle (droplet) size distribution of a predefined dry aerosol size distribution and composition and the changes of LWC and $x_v$ as function of rH. In order to analyze the sensitivity of $x_v$ towards changes of the air temperature and aerosol concentration, we selected the original China/Taiwan air mass (Table 1) and the starting conditions $x_v = 1000$ m and 20°C (corresponding to rH = 99.0208%), to apply step 3 of the model. Now, the model box was, in addition to the conditions described in step 1, closed with respect to the total water content. In other words, the sum of LWC + water vapor concentration [mg m$^{-3}$] was kept constant. In this case, changes of rH were realized through changes of the temperature T. For an increase of T by a certain increment, rH decreases due to the slope of the water vapor saturation curve (Clausius-Clapeyron equation). The WMO version of the Magnus Formula was applied to approximate the Clausius-Clapeyron equation. If the temperature is increased, the rH decreases, consequently droplets evaporate and LWC decreases, which leads to an increase of rH, again. In the model, the new value of rH is looked up which represents the equilibrium between rH and LWC under the given total water concentration and the aerosol distribution. At this point, $x_v$ is higher than it was before the increase of T. Alternatively to the temperature change but still within step 3 of the model domain, the aerosol concentrations were changed by reducing or increasing the number concentrations within the entire size range of dry particles (columns N in modes I, II, and III in Table 1) by a certain percentage. In these cases, the temperature was kept constant in addition to the conditions described so far (however, with incremental changes of the aerosol concentrations, as described), the respective equilibrium points between rH and LWC were sought and $x_v$ determined. A lower aerosol concentration lead to a smaller LWC, higher rH, and larger $x_v$.

RESULTS AND DISCUSSION

In our selected air masses and the described conditions, the LWC grows significantly when rH approaches 100% (upper panel in Fig. 4). The visual range decreases and reaches in all 5 cases below 1000 m at rH < 100%. In other words, stable fog situations can readily evolve with non-activated aerosol particles. It is not directly evident from
Fig. 4 if these aerosol particles can be classified as CCN, meaning that they would eventually be activated and go into a phase of rapid growth. However, the employed chemical and size information (Table 1) tells us in combination with results from Köhler curve computations (Fig. 1) that a large section of the involved particles would act as CCN eventually.

Visual ranges of 1000 m are reached at relative humidities above 99% in all cases (Table 2). Although all these RH’s, at which foggy conditions are established, are very high, close to 100% and therefore certainly outside the range of being precisely measurable in real-world conditions, we consider it to be remarkable that fog can be established in all 5 cases at RH < 100%. Moreover, when the relative humidity reaches the value of exactly 100%, the visual ranges as shown in Fig. 4 (bottom panel) reach values between 98 m and 345 m for the 5 air masses (Table 2). There is only a rather poor correlation (results not shown in detail) between RH and LWC at \( x_v = 1000 \) m for the 5 air masses (columns 2 and 3 in Table 2), and no significant correlation between \( x_v \) and LWC at RH = 100% (not shown in detail). It is thus not LWC that drives the visual range.

To better understand the relationship between droplet growth and the scattering coefficient, we compare Fig. 5 with Fig. 3. While in Fig. 3 (lower panel), the mass distribution of the dry particles is shown, Fig. 5 shows the LWC, which is a mass distribution as well, for the same air masses, but under the conditions when the visual range of \( x_v = 1000 \) m is established for the respective air masses (columns 2 and 3 in Table 2). All modes are significantly shifted towards larger sizes due to deliquescence of particles, in each case by more than 0.6 logarithmic units or a factor of 4 in diameter. However, clear differences are evident as well. For the London air mass (bold short-dashed lines), the larger of the two apparent modes (Fig. 3) gained relative importance during deliquescence and dominates the LWC distribution in Fig. 5 (upper panel). However, these large particles (\( D_{p,wet} \))
Fig. 5. Liquid water content (LWC, upper panel) and extinction coefficient ($b_{ext}$, lower panel) for the 5 air masses and for the condition $x_v = 1000$ m, as functions of the wet diameters $D_{p,wet}$ of droplets (Table 1, see text for air mass identification and more details): SP30 (bold gray broken lines), R2013 (dash-dotted lines), London (bold short-dashed lines), São Paulo (full lines), and China/Taiwan (bold grey full lines).

> 5 µm) have only a relatively small scattering efficiency and thus contribute only little to the extinction coefficient (lower panel in Fig. 5). It is the small droplets that dominate the extinction coefficient. Elias et al. (2009) found for haze conditions that the reduction of the visual range is dominated by sub-micron particles, which is also in line with the finding Roach et al. (1976) that large fog droplets contribute much more to LWC than to light extinction. The maximums of all our 5 extinction coefficient curves in Fig. 5 (lower panel) lie at diameters $D_{p,wet}$ between 1.0 and 1.3 µm.

The China/Taiwan air mass shows the largest LWC during increasing $rH$ (top panel of Fig. 4) and, at the same time the lowest $x_v$ (bottom panel). The cause for low $x_v$ lies, however, not in its high LWC or high number of relative large particles in the dry state (Fig. 3), but rather in the large number of small particles (mode I, Table 1). The unimodal R2013-distribution with a $D_{p,dry}$ mode diameter $D_m$ of 0.0424 µm leads to $x_v = 1000$ m with the lowest LWC of 1.16 mg m$^{-3}$ and the second lowest $x_v$ of 179 m at $rH = 100\%$, again at a very low LWC.

We analyzed the influence of temperature changes and changes of the aerosol concentrations on $x_v$. The China/Taiwan air mass and the conditions of $x_v = 1000$ m were selected as base case (dot in Fig. 6). An increase of the temperature and a decrease of the aerosol concentrations lead to an increase of $x_v$, which means a decrease or dissipation of fog. On the other hand, a decrease of the temperature or an increase of the aerosol concentration will decrease $x_v$ and intensify the fog. It becomes evident from Fig. 6 that the response of $x_v$ to changes of temperature and aerosol concentrations is not linear. Nonlinearity along both axes occur, along the temperature axis due to non-linearity of the water vapor saturation curve (Clausius-Clapeyron equation), and along the aerosol concentration curve due to the complex interaction between diameter, projected area, volume, and Mie scattering efficiencies of single droplets. However, it seems appropriate to state that the response of $x_v$ to temperature changes near the base case and at constant aerosol concentration is $dx_v/dT = 36$ m/(0.1°C) and that the response to the aerosol concentration $c_a$ at constant temperature is $dx_v/dc_a = -35$ m/(10% change of aerosol concentration). In other words, an increase of the temperature by 0.1°C has about the same effect on the visual range as a decrease of the aerosol concentration by 10%.
CONCLUSIONS

Fog as an optical phenomenon with horizontal visibilities below 1000 m has been studied. Fog often occurs when the relative humidity (rH) in an air mass has exceeded 100% and the critical supersaturation (for example, 100.1%, Seinfeld and Pandis, 2006) was reached for a considerable portion of the present CCN’s, so that fast droplet growth was initiated. This study, however, focusses on conditions of rH < 100%. It could be clearly shown that fog can occur under such conditions as well for various types of urban air masses. Although rH did not have to exceed the value of 100% in order to lead to foggy conditions, values very close to 100%, i.e., above 99%, were required to produce fog (Table 2). The visual range was then reduced by Mie scattering of deliquescent particles (droplets) in the size range around 1 µm diameter (D_{p,wet}). Such particles are hygroscopic and have, under dry conditions, a diameter D_{p,dry} in the order of 0.25 µm, which is within the accumulation range. Although these particles are the most effective ones in Mie scattering, they do not carry much LWC after deliquescence. A lot of LWC, on the other hand, is located far towards larger droplets in the size spectrum (Fig. 5, top panel) at D_{p,wet} > 5 µm. Therefore, there is no close correlation between LWC and x_v.

For the conditions studied here, a (computational) increase of the temperature by a few tens of a degree can have a strong effect on the visual range x_v. In the context of the recently observed climate change, an increase of the temperature by a few tenths of a degree has occurred over vast areas of the terrestrial surface (IPCC, 2013). Therefore, it is reasonable to assume that climate change is a driver for the decrease of fog. This general statement, which is based on theoretical considerations and box model assumptions, can not easily be downscaled to a given situation. Real world fog is a phenomenon with highly dynamic processes happening on time scales of minutes to days, while an increase of the average temperature may not be specifically associated with the drivers of fog formation. Further note that a decrease of fog, if caused by the processes discussed in this contribution, will lead to a one-to-one increase in haze, because an increase of the visual range from, for example, 600 m to 1400 m will results in a hazy condition instead of a foggy one.

The influence of urbanization on fog is manifold. An intensification of the UHI is associated with an increase of the air temperature especially during the nights, thus leading to a decrease of the tendency to form radiation fog. The UHI is also often associated with a decrease of the water vapor content in air, leading to the same effect. If urbanization is associated with an increase of air pollution, the fog formation can be fostered. Overall, the literature data as reviewed in the first section indicate that a decrease of fog happens more frequently along with ongoing urbanization than an increase. In rural or agricultural areas, an increase of the temperature may lead to a decrease of fog due to the reduced rH, however, an enhanced evapotranspiration as result of higher temperature may as well lead to increasing fog. In any case, a verification of the processes on the basis of meteorological data will be challenging because the involved temperature changes are rather small and the associated changes of rH within the range 99% < rH < 100% are unmeasurably small.

A computational increase or decrease of the aerosol concentration also had a strong impact on fog. Higher aerosol concentrations lead to a reduction of x_v, i.e., an
intensification of fog. Ten percent change of the aerosol concentration had about the same effect on fog as 0.1°C temperature change. This is well within the range at which changes (improvements) of air pollution levels have been reported for various regions worldwide (Sivertsen, 2006; Guerreiro et al., 2014; http://www.epa.gov/airtrends/pm.html, last access 21 May 2015).

This conclusion is based on concentration changes that affected the entire size spectrum equally and no changes of aerosol composition were involved. We know from the discussion above that it is mostly the small particles that lead to a reduction of $x_v$. Therefore it seems not feasible to correlate the trend of particulate material (PM) mass concentrations as measured in routine stations (typically PM$_{10}$ or PM$_{2.5}$ for particulate material with aerodynamic diameters below 10 µm and 2.5 µm, respectively) with $x_v$. Note that PM$_{10}$ and PM$_{2.5}$ mass concentrations are dominated by larger particles ($D_{dry} > 1$ µm), which are no effective reducers of $x_v$ when RH increases. A reduction of PM concentrations may therefore be associated with an increase of $x_v$, however, such correlations can be interpreted meaningfully only if sub-micron particle concentrations, which do not contribute much to PM$_{10}$ and PM$_{2.5}$, will decrease together with PM. We speculate that the decrease of fog in urban areas is rather correlated with a decrease of the emissions of precursor gases for the formation of small, hygroscopic particles, mainly SO$_2$ and NO$_x$, than with the emissions or concentrations of coarse particulate material.

Overall, both air quality and climate change can cause fog trends as observed. An evaluation of the described physical processes behind an observed fog trend at a given site on the basis of existing meteorological and air quality routine data should include potential precursor gases for new particle formation.

ACKNOWLEDGMENTS

The authors would like to thank the Taiwan Ministry of Science and Technology (MOST) and the NRW Heinrich Hertz Stiftung for supporting this study. We further thank M. Buhrmester for help with literature search and S. Prahl for providing Mie scattering software at http://omlc.org/calc/mie_calc.html. Helpful comments by three anonymous reviewers on an earlier version of the manuscript are gratefully acknowledged.

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Received for review, May 21, 2015
Revised, August 17, 2015
Accepted, October 13, 2015