**A Review of Hygroscopic Growth Factors of Submicron Aerosols from Different Sources and its Implication for Calculation of Lung Deposition Efficiency of Ambient Aerosols**

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**ABSTRACT**

Hygroscopic properties are an important parameter in determining the atmospheric behaviour of aerosols and their optical properties, influencing the direct and indirect effect of aerosols upon climate. As a result, particle hygroscopicity has received much attention with a rapid increase of publications in recent years. Likewise, hygroscopicity is an important characteristic influencing the deposition efficiency of particles in the human respiratory tract by affecting the particle size. The object of this study is to review the existing knowledge on the hygroscopic growth factor (Gf) of atmospheric submicron particles and its influence on the lung deposition calculation. The study briefly reviews first the Gf values of particles generated from various sources, including nucleation, traffic emissions and biomass burning, discussing the spatial and temporal variations. It then summarizes Gf values of submicron particles and number fraction of each hygroscopic groupmeasured in different ambient environments. These include marine, roadside, urban background and rural environments. The study concludes by estimating the lung deposition efficiency of ambient particles using a modified version of the ICRP model for hygroscopic particles. Furthermore, the effect of hygroscopicity on lung deposition efficiency of ambient particles has been estimated. The ICRP model seems to predict well the deposition efficiency (DE) values for small ambient particles in the extra-thoracic and tracheo-bronchial region, but not the alveolar region, where they are overestimated. However for larger particles (Dp>200 nm) the ICRP model underestimates the DE values, with the extra-thoracic region the most affected of the three. Hygroscopic atmospheric particles with a small diameter (Dp < 200 nm) showed a lower total lung deposition than hydrophobic particles of the same initial size due to their hygroscopic properties. On the other hand, larger hygroscopic particles (Dp > 200 nm) showed much higher lung deposition than hydrophobic particles.

**Keywords:** Aerosol; airborne particles; hygroscopic growth; growth factor

**INTRODUCTION**

Hygroscopic properties are well known to play a vital role in the atmospheric behaviour and health implications of aerosols ([Ferron et al., 2005](#_ENREF_15); [Hiller, 1991](#_ENREF_21); [Tu and Knutson, 1984](#_ENREF_58)). Aerosol particles can shrink or grow in size by exchanging water vapour with the surrounding air as relative humidity (RH) changes, thus having a direct effect on the radiation balance of the atmosphere through the change of scattering and absorption of light ([Tang et al., 1981](#_ENREF_52)). Furthermore, hygroscopicity has a strong influence on the ability of particles to act as cloud condensation nuclei ([Hämeri et al., 2001](#_ENREF_19); [Petters and Kreidenweis, 2007](#_ENREF_41); [Reutter et al., 2009](#_ENREF_42)). Consequently, it also affects global climate indirectly. Particles can also absorb the water vapour in the respiratory tract when they penetrate into the lung, changing in size, and hence affecting their lung deposition efficiency ([Broday and Georgopoulos, 2001](#_ENREF_4); [Chan et al., 2002](#_ENREF_8); [Finlay and Stapleton, 1995](#_ENREF_16); [Hofmann, 2011](#_ENREF_21)).

Hygroscopic growth factor (Gf) is one of most used parameters in determining hygroscopic properties of particles and their influences. The hygroscopic growth factor (Gf) is defined (equation 1) as the ratio between particle diameter measured at a specific RH (Dw), and dry particle diameter (Dp) measured at a low RH (RH<10%) ([Massling et al., 2005](#_ENREF_31); [Swietlicki et al., 2008](#_ENREF_49)):

Gf (RH) = $\frac{D\_{w}(RH)}{D\_{p} (RH<10\%)}$ (1)

Gf of atmospheric aerosols is determined by several methods, including measurement using particle spectrometers (i.e. Hygroscopic Tandem Differential Mobility Analyzer H-TDMA) or estimated using models from the growth factors of each particle component ([Meier et al., 2009](#_ENREF_32)). Gf values depend upon both physical and chemical properties of particles such as particle size and composition. Based on Gf values, ambient aerosols have been classified into four hygroscopic groups: nearly hydrophobic particles (Gf =1.0-1.11 for 100 nm particles at RH=90%), less-hygroscopic particles (Gf =1.11-1.33), more-hygroscopic particles (Gf = 1.33- 1.85) and sea-salt particles (Gf > 1.85) ([Swietlicki et al., 2008](#_ENREF_49)).

Recently, there have been numerous publications which have reported measurements of the hygroscopic growth factors of particles in various environments. [Swietlicki et al. (2008)](#_ENREF_49) did an excellent review on the hygroscopic properties of submicron atmospheric particles measured with H-TDMA instruments. Swietlicki’s work describes the Gf measurement methods, the variation of measured Gf between different atmospheres as well as the influence of chemical and physical properties on Gf. In addition, the implications of Gf in predicting particle critical supersaturation have been considered ([Rissler et al., 2010](#_ENREF_43); [Swietlicki et al., 2008](#_ENREF_49)). However, the temporal variation or influence of sources upon Gf has not yet been fully investigated. Therefore, this work aims to summarize the measured Gf values from different sources and the chemical and physical processes which cause the change of Gf values during ageing processes. Furthermore, we have estimated the influence of hygroscopic properties on the lung dose of ambient particles.

**HYGROSCOPIC PROPERTIES OF AEROSOLS EMITTED FROM DIFFERENT SOURCES**

**Traffic Emissions**

Traffic emissions are recognised as a main source of particles in urban environments (Morawska et al., 2008), and hence hygroscopic properties of traffic particles are of much interest. One of the earliest Gf measurements of traffic particles was conducted by [Weingartner et al. (1993)](#_ENREF_58). Aerosols were generated by a four-stroke spark ignition engine using unleaded gasoline and then analyzed by a Tandem Differential Mobility Analyzer (TDMA) system. Measured growth factors of particles with original diameters of 29, 42, 77 and 111 nm at sub-and-super saturations showed very low values (~0.98- 1.15), and therefore they were considered to be hydrophobic. The study found that particles of different sizes showed different hygroscopic behaviours. For example, primary particles with diameters of 29 and 42 nm were observed to grow faster than aggregates with a diameter of 77 and 111 nm. In addition, [Weingartner et al. (1995)](#_ENREF_57) noted that particles with a diameter of 51.5 nm kept their initial size, whilst larger ones (108 nm), typically non-spherical fractal-like structure aggregates, shrank to around 99% of their initial size at RH ≤ 95%. It could be explained by the capillary forces of water condensed in angle cavities of aggregates, which caused any asymmetric part of the aggregates to collapse. When they exposed to RH ≥95%, both primary particles and aggregates started to grow, but this growth was not appreciable. Similarly, [Weingartner et al. (1997)](#_ENREF_59) conducted an experiment on soot particles from a diesel engine and found that diesel soot particles were also nearly hydrophobic with growth factors roughly 1.01 for 50-110 nm particles. This result was confirmed by [Dua et al. (1999)](#_ENREF_12) who measured the hygroscopicity of particles emitted from two different diesel-powered vehicles. The diesel particles were found to be slightly soluble, and did not show a significant growth at a RH greater than 99%.

Hygroscopicity of traffic particles depends upon the type of fuels, fuel additives and pre-treatment devices. [Weingartner et al. (1997)](#_ENREF_59) found that growth factors of gasoline particles were smaller than those of diesel particles measured under loads representing a lean air/fuel ratio. In addition, a higher sulphur level in fuel was reported to lead to a higher growth factor of particles. Also, soot particles become more hygroscopic once they are subjected to pre-treatment with O3 and UV radiation ([Weingartner et al, 1997)](#_ENREF_59), which can be explained by the oxidation or photolysis of hydrocarbons such as PAHs on the particle surface, which produce more soluble compounds. In a further study on the effect of fuel on hygroscopic properties of diesel particles, [Happonen et al. (2013)](#_ENREF_20) studied the effect of adding oxygen into the fuel using hydro-treated vegetable oils (HVO) and with HVO + fuel-oxygenate (di-n-pentyl ether) blend. They found an increased hygroscopicity of exhaust particles after addition of the oxygen rich blend. Oxygen atoms, which were emitted in larger quantities with oxygenated fuel, may increase the oxygen to carbon ratio of an hypothetical superficial thin layer of organic material on the exhaust particles ([Happonen et al., 2013)](#_ENREF_20), resulting in more polar compounds by oxidation, and thus increasing the hygroscopic growth of the emitted particles. This is consistent with the findings of Jimenez et al. (2009), who reported that an increase of the oxygen to carbon-ratio resulted in an in increased hygroscopicity of the organic aerosol.

During the ageing process, hygroscopic properties of such particles could be changed leading to a higher growth factor. [Weingartner et al. (1995)](#_ENREF_57) measured the hygroscopicity of aged aerosols in a dark bag filled with exhaust gases from an engine. Their investigation found that particles became more hygroscopic when the residence time increased in the bag. After six hours of the aging process, Gf values of traffic particles increased from 1.0 to 1.04. [Weingartner et al. (1997)](#_ENREF_59) suggested four possible mechanisms, to understand the changes of the hygroscopicity of the soot particles during the aging process. These included coagulation, gas-to-particle conversion, cloud processing and photochemical degradation. [Kotzick and Niessner (1999)](#_ENREF_26) studied the aging process of ultrafine carbon aerosols in order to clarify which chemical and physical processes were responsible for these changes. They suggested that coagulation of ultrafine carbonaceous aerosols with soluble particles such as sodium chloride, sulphuric acid aerosols and oxidised hydrophilic species produced by reaction with ozone could transform carbonaceous particles to become more hydrophilic and act as cloud condensation nuclei in the atmospheric condensation process. This is in agreement with [Decesari et al. (2002)](#_ENREF_11) who suggested that water-soluble organic compounds produced from oxidation of soot particles make soot particles become more hydrophilic. In a recent study on the change of hygroscopicity during ageing of diesel soot, the increase of the hygroscopic growth factor of soot particles was linked with the condensation of secondary organic aerosols formed by photo-oxidation of volatile organic compounds emitted with exhaust gases ([Tritscher et al., 2011](#_ENREF_53)).

**Biomass Burning**

Biomass burning is an important source of particulate matter in the atmosphere. Hygroscopicity of particles emitted from biomass burning is known to change the light scattering and cloud nucleation properties, thereby having a strong impact on the earth’s radiative balance as well as the climate. Hence, hygroscopicity of biomass burning related particles is of much concern.

Freshly emitted biomass burning particles are known to be hydrophobic or less-hygroscopic. [Rose et al. (2008)](#_ENREF_46) showed a low growth factor with Gf values varying between 1.04 and 1.10 at RH 85 %. This low hygroscopic growth factor is due to the chemical properties of particles generated from biomass burning, which mainly consist of organic materials and black carbon, both known to be hydrophobic or less-hygroscopic ([Martin et al., 2013](#_ENREF_29)). [Dusek et al. (2011)](#_ENREF_13) suggested that hygroscopicity of wood burning particles was predominantly influenced by organic compounds. The growth factors of freshly generated biomass burning particles are small and equivalent to particles consisting of 5-20 % ammonium sulphate along with insoluble materials. Investigating the hygroscopic properties of five major organic products (levoglucosan, D-glucose, and vanillic, syringic and 4-hydroxybenozoic acids) from pyrolysis of wood, [Mochida and Kawamura (2004)](#_ENREF_33) found that levoglucosan and D-glucose show a significant hygroscopic growth (G­­f­ ~1.37-1.38 at 90% RH) while the remainder do not show any hygroscopic growth even up to 95% RH.

Like traffic related-particles, particles emitted from biomass burning are expected to show an increase of hygroscopic growth factors during ageing in the atmosphere. [Kotchenruther and Hobbs (1998)](#_ENREF_25) reported that the growth factors for light scattering for aged smoke (1.3-1.5) were higher than those for fresh smoke (1.1-1.3). A recent study by [Martin et al. (2013)](#_ENREF_29) who conducted different wood burning experiments in a smog chamber found that the hygroscopicity of such particles increased with time during the aging process, except in the smouldering phase experiment. Secondary formation of soluble components such as sulphate and polar organic compounds was responsible for the increase of hygroscopicity of smoke particles during ageing ([Hallquist et al., 2009](#_ENREF_18); [Kotchenruther and Hobbs, 1998](#_ENREF_25); [Rogers et al., 1991](#_ENREF_45)). Furthermore, [Decesari et al. (2002)](#_ENREF_11) reported that macromolecular humic-like substances can be formed by atmospheric oxidation of soot; thereby increasing the water uptake properties of aerosol particles. To conclude, the oxidation of organic soot is also known as the main ageing process which makes particles become more hydrophilic ([Martin et al., 2013](#_ENREF_29)).

**Nucleation**

New particles formed by nucleation account for a significant fraction of the total number of particles in the atmospheric environment. [Ogulei et al. (2007)](#_ENREF_38) reported that nucleation related with traffic emissions represented between 15 % to 21 % of the total number particles in Rochester, United States. Similarly, another study conducted by [Kasumba et al. (2009)](#_ENREF_24) found that nucleation represented between 15.4 to 20.7 % of total particle number.

Hygroscopic properties of the nucleation mode particles (4-20 nm) have been measured in various environments such as in urban/suburban areas ([Petäjä et al., 2007](#_ENREF_43); [Sakurai et al., 2005](#_ENREF_50); [Väkevä et al., 2002b](#_ENREF_60)), in boreal forests ([Hämeri et al., 2001](#_ENREF_19); [Park et al., 2009](#_ENREF_42)) and in coastal regions ([Buzorius et al., 2004](#_ENREF_6); [Väkevä et al., 2002a](#_ENREF_59)). [Sakurai et al. (2005)](#_ENREF_50) measured G­f in an urban background of Atlanta and reported that the 10 nm nucleation particles were more hygroscopic; showing a growth factor value of ~1.4 that was equivalent to 10 nm ammonium sulphate particles. This finding is in agreement with the hypothesis that new particles in Atlanta consisted of ammonium and sulphate alone ([Jung et al., 2006](#_ENREF_25)). Similarly, [Petäjä et al. (2007)](#_ENREF_43) found higher growth factors (Gf maximum ~ 1.6) of nucleation mode particles during nucleation event days, compared to those (Gf maximum ~1.3) on non-event days in Marseille (France). However, this study also found that the condensational growth of nucleation mode particles does not only depend upon sulphuric acid concentration, but is also controlled by other water insoluble components. By dividing the nucleation event days into groups based on the anthropogenic influence, [Petäjä et al. (2007)](#_ENREF_43) concluded that the Gf values varied between the different properties of condensing vapours. Particularly, during clean events with less anthropogenic influence, the growth of nucleation particles was due to insoluble materials rather than the condensation of water soluble components, whilst during the polluted events, the growth of particles was mainly contributed by water soluble components.

More evidence of the contribution of other insoluble materials to the growth of nucleation particles was provided by other observations of the lower growth factor values of nucleation particles ([Hämeri et al., 2001](#_ENREF_19); [Väkevä et al., 2002a](#_ENREF_59); [Väkevä et al., 2002b](#_ENREF_60)). [Väkevä et al. (2002a)](#_ENREF_59) reported that the nucleation mode particles (8-10 nm) sampled during nucleation events in the coast of Ireland were less hygroscopic (G­f~1.1). This was attributed to new particles formed in coastal areas as a result of nucleation of iodine oxides ([Carpenter et al., 1999](#_ENREF_7); [O'Dowd et al., 2002a](#_ENREF_39); [O'Dowd et al., 2002b](#_ENREF_40)), resulting in a much lower growth factor. In addition, [Hämeri et al. (2001)](#_ENREF_19) investigated the hygroscopic growth of nucleation particles at a boreal forest site in Southern Finland. This study concluded that the sulphuric acid concentrations cannot explain the growth of nucleation particles by itself in the boreal forest, suggesting that the growth process may be related to organic compounds. To conclude, the hygroscopic growth factor of nucleation mode particles varied between environments, depending upon the air masses as well as the nucleation and growth mechanism in different atmospheres.

**HYGROSCOPIC BEHAVIOUR OF AMBIENT AEROSOLS**

The review of Swietlicki et al. (2008) focused on the influence of air masses and local emissions on hygroscopic properties of particles from different environments. That investigation noted that the hygroscopic properties of ambient aerosol particles vary in different atmospheres largely influenced by the origin of the air masses and local emissions. In this section, we will not focus on the spatial variation of hygroscopic particles; but on the spatial variation of the number fraction distribution and original sources of each group of hydrophobic/hygroscopic particles. Both will be influential parameters to consider in the calculation of lung deposition as discussed in the following section. We will also review in this section the temporal variation of hygroscopic particles.

**Spatial Variation**

Urban environments

The growth factors of urban particles in most previous studies were classified in three groups: near-hydrophobic, less-hygroscopic and more-hygroscopic (Swietlicki et al., 2008). The dominant hydrophobic and less-hygroscopic particles in the urban environments could be explained by the sources of particles at these sites, where traffic-related emissions were found as a main source of urban ultrafine particles ([Morawska et al., 2008](#_ENREF_35)). Near-hydrophobic particles can be assumed to originate from fresh traffic emissions that contain black carbon, while less-hygroscopic particles may have originated from transformed oil combustion particles ([Swietlicki et al., 2008](#_ENREF_49)). In addition, other combustion processes such as wood combustion in some cases contribute significantly to the number fraction of near-hydrophobic and less-hygroscopic particles (Baltensperger et al., 2002; Dusek et al., 2011).

Hygroscopic properties were measured at both urban background and kerbside sites in previous studies. [Ferron et al. (2005)](#_ENREF_15) reported that the growth factors of particles within a certain size from different sampling sites do not vary significantly, but the hydrophobic/ hygroscopic particle fraction is strongly influenced by the type of site. At the kerbside site, the number fraction of the nearly-hydrophobic group accounted for more than 80% of total particles with a size of 50 nm, while this number was 30-40% at the background site. When particle size increases (> 100 nm) the fraction of near-hydrophobic particles decreases while the fractions of less hydrophobic and hygroscopic particles increases. For example, nearly-hydrophobic particles measured at an urban background of Neuherberg, Germany accounted for 42% of total particles with size at 50 nm while they only represented 25% of particles with a size of 200 nm ([Tschiersch et al., 1997](#_ENREF_54)). This is consistent with the decrease of traffic particles at larger sizes. Particle number size distributions of particles emitted from vehicles show peaks at below 50 nm (Charron and Harrison, 2003). Hygroscopic particles, which are known to come from regional background transportation, were found to be dominant in the accumulation mode.

Rural environments

Aerosol particles in rural environments can derive from anthropogenic sources such as traffic or industrial emissions by advection and from local emissions such as biomass burning discussed below. They also originate from atmospheric processes such as nucleation. Since rural sites may be exposed to aerosols from multiple sources, the observed Gf show a large variation. Similar to urban environments, the growth factors in rural areas were classified in three groups: near-hydrophobic (Gf ~1.01-1.14), less-hygroscopic (Gf ~1.14-1.32) and more-hygroscopic (1.34~1.63). The number fractions of the three classes show differences between urban and rural areas. The less- and more-hygroscopic particles are major contributors to the total number of particles. [Swietlicki et al. (1999)](#_ENREF_50) reported that the number fraction of more-hygroscopic particles accounted for 64% of total particle number concentrations with a size of 35 nm; while this number increased to 81% of the total particle number with a size of 265 nm. However, during biomass burning events, the contribution of more-hygroscopic groups was limited and the number fraction of less-hygroscopic particles was dominant ([Rissler et al., 2006](#_ENREF_44)). The large fraction of organic compounds and black carbon emitted from biomass burning were the reason for the abundance of less-hygroscopic particles.

Marine environments

More-hygroscopic particles are ubiquitous in marine environments. [Zhou et al. (2001)](#_ENREF_61) reported that the frequency of occurrence of more-hygroscopic particles was above 81% and when these were present it represented a number fraction larger than 0.93. A mixture of ammonium sulphate and non-neutralized sulphate, which originate from oceanic biota, are the main components of this group, leading to very high growth factors. For example, the observed Gf of more-hygroscopic particles with a diameter of 50 nm in coastal environments ranges from 1.40 to 1.9 at 90% RH ([Zhou et al., 2001](#_ENREF_61)). These are much higher than Gf in rural (Gf ~ 1.32-1.61) and urban environments (Gf ~ 1.36-1.49). The growth factors of marine aerosols are found to be even higher during days with high wind speeds, when the marine aerosols include externally mixed sea salt particles. Wind at high speeds brings freshly produced sea-salt particles with high volume fractions of sodium chloride, with large growth factor Gf > 1.85, resulting in higher growth factors for marine aerosol. However, the lifetime of externally mixed sea salt aerosols is short and limited by ageing processes. During ageing periods, sodium chloride reacts with sulphuric acid or with ammonium and sulphate compounds producing sodium sulphate, which has a lower growth factor (Gf ~ 1.6). The number fraction of less-hygroscopic and near-hydrophobic particles was often lower than 0.1 with the occurrence frequency around 20%. These particles could have arrived from continental air masses or local emission from ships.

**Temporal Variation**

It is clear that the temporal pattern of the number fraction of hydrophobic/hygroscopic particles depends strongly upon the pattern of local sources, as well as the meteorological conditions such as temperature variation, precipitation or the origin of the air mass. Masling et al. (2005) reported that the number fraction of nearly-hydrophobic particles measured at an urban background area showed a distinct diurnal pattern with peaks corresponding to daily rush hours. Nearly-hydrophobic particles which are found to be dominant during daytime in urban areas can be attributed to soot and organic compounds emitted directly from vehicles or the particles formed by nucleation in the dilution and cooling of fresh vehicular emissions, leading to a lower averaged growth factor during daytime. In contrast, another study in a boreal forest by [Hämeri et al. (2001)](#_ENREF_19) showed the opposite behaviour. The growth factors of particles in nucleation and Aiken modes were found to be the highest during the early afternoon and lowest during the late evening and early morning. This trend was consistent with other studies ([Boy et al., 2004](#_ENREF_3); [Ehn et al., 2007](#_ENREF_14); [Fors et al., 2011](#_ENREF_17); [Petäjä et al., 2007](#_ENREF_40)). [Fors et al. (2011)](#_ENREF_17) suggested that an increase of the planetary boundary layer depth which entrains more hygroscopic particles with height when the sun rises could be an explanation for the increase of hygroscopic particles in boreal forests. Another reason could be from the different atmospheric conditions between day and night ([Ehn et al., 2007](#_ENREF_14)). For example, the evaporation of semi-volatile organics with low hygroscopicity during daytime alters more hygroscopic particles ([Fors et al., 2011](#_ENREF_17)). It may also be associated with daytime oxidative processes which produce more functionalised organic compounds that have higher hygrocopicity (Wong et al., 2011).

There are a few studies on long-term measurements of particle hygroscopicity. The growth factors of each group seem to be stable, while fraction numbers exhibit a seasonal variation ([Ferron et al., 2005](#_ENREF_15)). A two-year study in southern Sweden reported that the number fraction of less-hygroscopic particles was higher in winter ([Fors et al., 2011](#_ENREF_17)). This trend concurs with a recent study in Paris ([Kamilli et al., 2014](#_ENREF_23)).

**EFFECTS OF AEROSOL HYGROSCOPIC PROPERTIES ON CALCULATION OF LUNG DEPOSITION**

**Lung Deposition Calculation for Ambient Submicron Aerosols**

The International Commission on Radiological Protection (ICRP) model and the Multiple Path Particle Dosimetry model (MPPD) are most widely used to estimate the regional lung deposition of particles ([Hussein et al., 2013](#_ENREF_22)). However, when these current mathematical models are utilized to calculate the regional lung dose of ambient particles, the result may not be accurate ([Asgharian, 2004](#_ENREF_1)). [Montoya et al. (2004)](#_ENREF_34) compared the total deposition fraction of fine and ultrafine ambient aerosols measured in a group of six healthy adults and the estimated total deposition fraction by the ICRP model. They reported that the ICRP model likely predicts the total deposition efficiency well for ambient particles smaller than 400 nm, but underestimates the deposition of particles larger than 676 nm, which is in agreement with other studies (e.g., [Daigle et al. (2003)](#_ENREF_10), [Löndahl et al. (2009)](#_ENREF_28)). The main reason is that the theoretical models do not account for the particle growth due to hygroscopic properties of ambient particles when they penetrate into the respiratory system ([Löndahl et al., 2009](#_ENREF_28); [Montoya et al., 2004](#_ENREF_34)).

The aim of this section is to investigate and compare the effects of hygroscopicity of particles from different ambient environments on their lung deposition efficiency using a modified ICRP model for hygroscopic particles. This study used 13 published data sets for hygroscopic growth factors and their number fraction in rural, urban background and roadside environments from previous studies (Roadside: Bavaria ([Ferron et al., 2005](#_ENREF_15)), Copenhagen ([Löndahl et al., 2009](#_ENREF_28)), Bresso ([Baltensperger et al., 2002](#_ENREF_2)); Urban background: Neuherberg ([Tschiersch et al., 1997](#_ENREF_54)), Leipzig ([Massling et al., 2005](#_ENREF_31)), Taipei ([Chen et al., 2003](#_ENREF_9)), Beijing ([Massling et al., 2009](#_ENREF_30)), Guangzhou ([Tan et al., 2013](#_ENREF_51)), Shanghai ([Ye et al., 2013](#_ENREF_60)); Rural: Bologna ([Svenningsson et al., 1992](#_ENREF_48)), Berlin ([Busch et al., 2002](#_ENREF_5)), Hohenspeissenberg ([Ferron et al., 2005](#_ENREF_15)), Great Dun Fell ([Swietlicki et al., 1999](#_ENREF_50)) as shown in Table 1.

**A modified ICRP model for ambient particles**

A modified ICRP model for hygroscopic particles is based on the previous work from [Löndahl et al. (2009)](#_ENREF_28), [Kristensson et al. (2013)](#_ENREF_27) and [Hussein et al. (2013)](#_ENREF_22). Ambient particles are divided into three groups, including near-hydrophobic, less-hygroscopic and more-hygroscopic. Subsequently, the ICRP model is applied separately for each type of particle with a consideration of their growth when they penetrate into the respiratory tract. The final deposition efficiency which is defined as the fraction of aerosol particles which deposit in the lung for aerosols in each environment was calculated by the mean value of deposition efficiency for these three groups weighted by their number fraction for each group as shown in equation (2) and assuming equilibration with a relative humidity in the airways of 99.5%. It is assumed that the submicron particles which have increased to their equilibrium size by their growth in the respiratory tract have the same behaviour as insoluble particles with a similar size in the respiratory system ([Asgharian, 2004](#_ENREF_1)). For example, hygroscopic particles with an initial size of 50 nm and a Gf value of 2 (at 99.5% RH) are assumed to have an equivalent deposition efficiency to hydrophobic particles (Gf = 1) with a diameter of 100 nm ([Löndahl et al., 2009](#_ENREF_28)). However, when a particle penetrates into the human respiratory tract, its growth also depends upon its residence time. Hence, the above assumption by Londahl et al. (2009) is limited to small particles which can grow and reach their equilibrium size rapidly. Ferron (1977) found that a particle smaller than 0.2 µm can reach its equilibrium size within 0.1 sec in the human respiratory tract. To correct the size growth for a particle larger 0.2 µm when it penetrates into the regional lung due to its residence time, our study used an approach which described the correlation between particle growth and residence time, provided by Ferron (1977). The residence times of a particle in the extra-thoracic, trachea-bronchial and alveolar regions are 0.116, 0.308 and 1.29 s during inspiration and 0.137, 0.365 and 1.56 s during expiration, respectively (Ferron, 1977).

In the distal airways, the lumen air has likely reached equilibrium. However, in the proximal airways and the extrathoracic region of the respiratory tract, the relative humidity in the airways may depart substantially from 99.5% equilibrium due to interactions between the ambient air temperature and humidity and the temperature and water activity of the mucus layer. Morrow (1986) showed that supersaturations as large as 130% could occur in the trachea under certain ambient temperature and humidity conditions. Calculations presented in this paper have been done considering a relative humidity of 99.5% and a temperature of 37 oC consistent thought the respiratory tract.

Deposition efficiency of the ambient aerosol is the sum over the three particle types:

DEambient = . DEnearly-hydrophobic + . DEless-hygroscopic + γ. DEmore-hygroscopic (2)

where, DEambient, DEnear-hydrophobic, DEless-hygroscopic, DEmore-hygroscopic are deposition efficiencies of ambient particles, nearly-hydrophobic particles, less-hygroscopic particles and more-hygroscopic particles, respectively.

γ are the number fractions of nearly-hydrophobic, less-hygroscopic and more-hygroscopic particles (γ= 1).

**A calculation of the hygroscopic growth factor of particles at 99.5% RH.**

Almost all published data for the growth factor of ambient particles were measured at around 90% RH ([Swietlicki et al., 2008](#_ENREF_49)), whilst the RH in the respiratory tract is approximately 99.5% ([Asgharian, 2004](#_ENREF_1); [Hussein et al., 2013](#_ENREF_22)). To estimate the growth factor of particles at the relative humidity found in the lungs (99.5%), we used Rissler’s model ([Kristensson et al., 2013](#_ENREF_27); [Rissler et al., 2006](#_ENREF_44)) which was introduced in previous studies by [Swietlicki et al. (1999)](#_ENREF_50), [Löndahl et al. (2009)](#_ENREF_28) and [Kristensson et al. (2013)](#_ENREF_27).

Gf = $\sqrt[3]{1+K\_{R}\frac{M\_{w} }{ρ\_{W}}(\frac{a\_{w} }{1-a\_{w}})}$ (3)

Where, K­R is a hygroscopic parameter which presents the effective number of moles of soluble molecules or ions per dry particle volume unit; Mw, ρw are the molecular weight and density of water; aw is water activity.

According to the Kohler equation:

aw = $\frac{RH}{100.C\_{k}}$ (4)

where, Ck is ­the Kelvin curvature correction factor:

Ck = exp($\frac{4M\_{W}σ\_{s}}{RT.ρ\_{w}D\_{p}}$) (5)

σs is the surface tension of the solution; R and T are the ideal gas constant and temperature respectively and Dp is the particle diameter.

Combining equations (1), (3), (4) and (5), the Gf at 99.5% RH is estimated from the following equation:

Gf-99.5% = $\sqrt[3]{1+\left(G\_{f-a}^{3}-1\right).\frac{99.5}{a}.\frac{(100e^{\frac{2.09}{Dp.Gf-a}}-a)}{(100e^{\frac{2.09}{Dp.Gf-99.5}}-99.5)}}$ (6)

Where, Gf-99.5% is the growth factor at 99.5% RH, Gf-RH is the growth factor at a% RH. Dp is the particle diameter measured at low RH (<10%). This model assumes that the number of soluble molecules is constant over the relative humidity and the droplet solution is ideal ([Rissler et al., 2010](#_ENREF_43)), therefore KR is assumed as a constant value as relative humidity changes from a% to 99.5%. The value of 2.09 was calculated assuming a surface tension of 0.072 J/m2. (M water=18 g/mol; R: 8.314 J/mol.K; T=298K) according to Petters and Kredenweis (2007). Different surface tension could cause some bias for growth factors calculation at 99.5%. Most particles in the atmosphere contain a substantial amount of organic material, which is functionalized from atmospheric processing and it is likely to act as a surfactant and substantially decrease the particle surface tension. However the error associated with different surface tensions can be acceptable as proved by a comparison between different models by Rissler et al. (2010).

The H-TDMA system has been used to determine hygroscopic growth factors and their number fractions for particles with selected diameters typically from 30 nm to 350 nm. To estimate the hygroscopic growth factors and number fractions for a full size range (10 nm-1000 nm), it is assumed that the hygroscopic parameter Kr and number fraction of particles at a diameter of 30 nm is suitable for those smaller than 30 nm while those of particles with a diameter of 350 nm to be suitable for those larger than 350 nm ([Hussein et al., 2013](#_ENREF_22)). Growth factors of the three hygroscopic groups at 99.5% RH were estimated according to equation (6) as shown in Figure 1 using growth factors reported in the literature.



**Figure 1:** Estimated hygroscopic growth factors of nearly, less and more hygroscopic particles at 99.5% RH from different environments. Calculations according to equation 6. To differentiate the lines, please see on-line colour version.

**Effects of Hygroscopic Properties of particles on Lung Deposition in Different Environments**

ICRP model curves were modified using particle hygroscopic growth factors reported in the literature in each group and then equation (2) was used to estimate the average value for ambient particles. The total average deposition efficiency (DE) of particles measured from different environments is shown in Figure 2. As seen in Figure 2, the hygroscopic properties of particles have a large effect on the total lung deposition efficiency (DE) of particles. Due to particle hygroscopicity, the deposition efficiency curve shifts its minimum from 400 nm for hydrophobic particles to 120-150 nm for rural and urban background particles and 140-200 nm for roadside particles (dry diameter). This is consistent with the experimental and modelled deposition of airborne particles at a kerbside site conducted by [Löndahl et al. (2009)](#_ENREF_28) who report that in kerbside areas, the lung deposition efficiency has a minimum value around 200 nm. The shifts in deposition curves to the left or right are readily explained by the decrease of Brownian diffusion and the increase of sedimentation and impaction (ICRP, 1995).

In roadside environments, the ICRP model seems to predict well the deposition efficiency of particles smaller than 200 nm. It could be explained by the fact that small particles at traffic sites are freshly emitted from traffic exhaust, therefore mostly comprising hydrophobic particles. At rural and urban background sites, the ICRP model seems to overestimate the lung deposition efficiency for particles smaller than 150 nm. For example, the deposition efficiency of hydrophobic particles with a diameter of 100 nm is found 40% higher than those of particles with same diameter size at rural sites. In contrast, the ICRP model seems to underestimate the total deposition efficiency for particles larger than 200 nm. The DE values for ambient particles at a diameter of 1µm are more than 2 times higher than those predicted by the ICRP model (Figure 2). It means that there is a huge error if the ICRP model is used to predict lung deposition efficiency of ambient particles larger than 200 nm without considering their growth factors. The error of not including the hygroscopicity factor of particles in the lung deposition calculation is of similar magnitude to the individual variability in deposition efficiency for particles in the low deposition efficiency range associated with factors such as different lung geometries, breathing patterns, or lung disease.



**Figure 2:** Calculations of deposition efficiency in total lung of ambient particles in various environments. To differentiate the lines, please see on-line colour version.

There is not much difference between the deposition efficiencies calculated for different sampling sites within each type of environment, suggesting that similar deposition efficiency curves are applicable for each type of environment. The maximum variation between deposition efficiencies for particle smaller 200 nm calculated for different sampling sites is 6.3%, 15.9% and 13.3% (Figure 2) within roadside, urban background and rural environments, respectively.

Figure 2 also shows that values of DE for particles in urban background areas seem to be similar to those in rural areas. On the contrary, DE of a particle in the ultrafine size range in roadside areas are found to be 1.2 to 1.6 higher than those in urban background and rural areas.

Hygroscopic properties of particles have also large influences on regional lung deposition efficiency as shown in Figure 3. In the extra-thoracic region, the hygroscopic properties seem to have little effect on the DE of ultrafine particles (Dp<100 nm), whilst DE increases considerably according to their hygroscopic growth properties for particles larger than 200 nm (Figure 3A). The DE in the tracheao-bronchial region is that less affected by the growth factor (Figure 3B). In the alveolar region, the DE for ambient particles smaller than 200 nm is much lower than for hydrophobic particles, whilst the DE increases for particles larger than 200 nm according to their hygroscopicity (Figure 3C).



**Figure 3:** Calculations of deposition efficiency in regional lung of ambient particles in various environments. A: In extra-thoracic region (ET); B: in tracheo-bronchial region (TB) and C: in alveolar region (AL). To differentiate the lines, please see on-line colour version.

**CONCLUSION**

In summary, the typical hygroscopicity of particles varies widely between different environments, depending upon the local sources and air masses, and shows a clear diurnal and seasonal trend, with higher values found in daytime and summer. Hygroscopic particles emitted from different sources were investigated. Freshly emitted traffic particles and biomass burning particles are hydrophobic or less-hygroscopic, whilst growth factors are found to increase during ageing. Finally, hygroscopic properties of particles have large effects not only on total lung deposition but also regional lung deposition of particles, causing a variation of DEs between sampling environments. For ultrafine particles (Dp <100nm), the DE of particles in kerbside areas was 1.2-1.6 higher than those in urban background and rural areas. The ICRP model seems to predict well DE values for small ambient particles in the extra-thoracic and tracheo-bronchial region, but not the alveolar region, where they are overestimated. However for larger particles (Dp>200 nm) the ICRP model underestimates the DE values, with the extra-thoracic region the most affected of the three. As a consequence, DE values of ambient particles larger than 200 nm in the total lung were much higher than hydrophobic particles due to their hygroscopic growth, while total lung DE was lower than predicted by the ICRP model for smaller particles in rural and urban background areas, but not in roadside areas.

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**TABLE LEGEND**

**Table 1:** Hygroscopic growth factors and number fractions (in parenthesis in the table) of three hygroscopic particle groups (Note: NH: Nearly-hygroscopic, LH: Less-hygroscopic, MH: More-hygroscopic) in the atmosphere.

**FIGURE LEGENDS**

**Figure 1:** Estimated hygroscopic growth factors of nearly, less and more hygroscopic particles at 99.5% RH from different environments. Calculations according to equation 6. To differentiate the lines, please see on-line colour version.

**Figure 2:** Calculations of deposition efficiency in total lung of ambient particles in various environments. To differentiate the lines, please see on-line colour version.

**Figure 3:** Calculations of deposition efficiency in regional lung of ambient particles in various environments. A: In extra-thoracic region (ET); B: in tracheo-bronchial region (TB) and C: in alveolar region (AL). To differentiate the lines, please see on-line colour version.

**Table 1:** Hygroscopic growth factors and number fractions (in parenthesis in the table) of three hygroscopic particle groups (Note: NH: Nearly-hygroscopic, LH: Less-hygroscopic, MH: More-hygroscopic) in the atmosphere.

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| Locations | Dp | RH | Gf (Number fraction) | Type of environments | References |
| NH | LH | MH |
| Bavaria, Germany | 50 | 85 | 1.01(0.83) | - | 1.30 (0.17) | Roadside | [Ferron et al. (2005)](#_ENREF_15) |
| 100 | 1.01 (0.84) | - | 1.33 (0.16) |
| 150 | 1.01 (0.78) | - | 1.39 (0.22) |
| 250 | 1.04 (0.68) | - | 1.47 (0.32) |
| Copenhagen, Denmark | 30 | 91 | 1.03 (0.89) | - | 1.47 (0.11) | Roadside | [Löndahl et al. (2009)](#_ENREF_31) |
| 40 | 1.03 (0.82) | - | 1.39 (0.18) |
| 80 | 1.02 (0.77) | - | 1.46 (0.23) |
| 160 | 1.03 (0.72) | - | 1.62 (0.28) |
| Bresso, Italy | 20 | 90 | 1.02 (-) | - | - |  Roadside (100 m highway) | [Baltensperger et al. (2002)](#_ENREF_2) |
| 50 | 1.03 (0.51) | 1.21 (0.49) | - |
| 100 | 1.02 (0.48) | 1.25 (0.62) | - |
| 200 | 1.02 (0.28) | 1.28 (0.72) | - |
| Leipzig, Germany | 50 | 92 | 1.06 (0.31) | 1.25 (0.31) | 1.44 (0.39) | Urban background | [Massling et al. (2005)](#_ENREF_34) |
| 150 | 1.09 (0.22) | 1.33 (0.15) | 1.65 (0.65) |
| Neuherberg, Germany | 50 | 90 | 1.05 (0.42) | - | 1.36 (0.58) | Urban  | [Tschiersch et al. (1997)](#_ENREF_57) |
| 75 | 1.02 (0.40) | - | 1.23 (0.60) |
| 110 | 1.02 (0.40) | - | 1.25 (0.60) |
| 165 | 1.03 (0.35) | - | 1.29 (0.65) |
| 300 | 1.02 (0.25) | - | 1.34 (0.75) |
| Taipei, Taiwan | 53 | 90 | 1.11 (0.78) | - | 1.43 (0.22) | Urban | [Chen et al. (2003)](#_ENREF_9) |
| 82 | 1.11 (0.74) | - | 1.49 (0.26) |
| 95 | 1.11 (0.61) | - | 1.54 (0.39) |
| 202 | 1.11 (0.59) | - | 1.66 (0.41) |
| Beijing, China | 30 | 90 | 0.99 (0.17) | 1.07 (0.41) | 1.26 (0.42) | Urban background | [Massling et al. (2009)](#_ENREF_33) |
| 50 | 1.04 (0.22) | 1.21 (0.34) | 1.40 (0.45) |
| 150 | 1.06 (0.21) | 1.27 (0.25) | 1.53 (0.54) |
| 250 | 1.06 (0.24) | 1.28 (0.25) | 1.58 (0.54) |
| 350 | 1.06 (0.28) | 1.30 (0.27) | 1.60 (0.46) |
| Guangzhou, China | 40 | 90 | - | 1.16 (0.41) | 1.46 (0.59) | Urban background | [Tan et al. (2013)](#_ENREF_54) |
| 80 | - | 1.14 (0.38) | 1.48 (0.62) |
| 110 | - | 1.13 (0.33) | 1.49 (0.67) |
| 150 | - | 1.12 (0.30) | 1.51 (0.70) |
| 200 | - | 1.11 (0.26) | 1.55 (0.73) |
| Shanghai, China | 30 | 91 | 1.09 (0.35) | - | 1.40 (0.65) | Urban (background) | [Ye et al. (2013)](#_ENREF_64) |
| 50 | 1.08 (0.24) | - | 1.49 (0.76) |
| 80 | 1.06 (0.18) | 1.31 (0.06) | 1.58 (0.76) |
| 100 | 1.05 (0.20) | 1.31 (0.04) | 1.60 (0.76) |
| 130 | 1.04 (0.21) | 1.35 (0.04) | 1.63 (0.74) |
| 150 | 1.04 (0.22) | 1.36 (0.03) | 1.65 (0.74) |
| 180 | 1.03 (0.22) | 1.36 (0.02) | 1.67 (0.75) |
| 200 | 1.03 (0.21) | 1.34 (0.02) | 1.68 (0.76) |
| Bologna, Italy | 30 | 90 | - | 1.12 (0.61) | 1.59 (0.39) | Rural | [Svenningsson et al. (1992)](#_ENREF_51) |
| 50 | - | 1.13 (0.63) | 1.61 (0.37) |
| 100 | - | 1.15 (0.59) | 1.58 (0.41) |
| 150 | - | 1.10 (0.53) | 1.55 (0.47) |
| 200 | - | 1.12 (0.57) | 1.57 (0.43) |
| Berlin, Germany | 50 | 90 | - | 1.12 (0.08) | 1.43 (0.92) | Rural (80 km south east of Berlin) | [Busch et al. (2002)](#_ENREF_5) |
| 100 | - | 1.11 (0.12) | 1.49 (0.88) |
| 150 | - | 1.08 (0.05) | 1.56 (0.95) |
| 250 | - | 1.08 (0.02) | 1.63 (0.98) |
| Hohenspeissenberg, Germany | 50 | 90 | 1.07 (0.52) | - | 1.29 (0.48) | Rural | [Ferron et al. (2005)](#_ENREF_15) |
| 100 | 1.03 (0.44) | - | 1.35 (0.56) |
| 150 | 1.03 (0.38) | - | 1.41 (0.62) |
| 250 | 1.04 (0.30) | - | 1.48 (0.70) |
| Great Dun Fell, UK | 35 | 90 | - | 1.10 (0.36) | 1.38 (0.64) | Rural | [Swietlicki et al. (1999)](#_ENREF_53) |
| 50 | - | 1.12 (0.37) | 1.44 (0.63) |
| 75 | - | 1.11 (0.39) | 1.52 (0.61) |
| 110 | - | 1.12 (0.31) | 1.58 (0.69) |
| 165 | - | 1.14 (0.24) | 1.64 (0.76) |
| 265 | - | 1.15 (0.20) | 1.69 (0.81) |

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