A simple approximation for size-mobility relationship of singly charged spherically symmetric airborne particles

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Concept of the particle diameter

Molecules and fine nanometer particles do not have solid surface and their geometric size is a conventional parameter. Mason (1984) recommended the mass diameter $d = (6m/(\pi\rho))^{1/3}$, which is calculated for an equivalent sphere according to the mass *m* and density of particle matter ρ. The density of particle matter exceeds the density of condensed bulk matter; e.g. an array of packed spheres has the density of 0.52ρ in case of the simple cubic lattice and 0.74ρ in case of the closest packing.

The attempts to define the diameter of particle according to mobility and an arbitrary size-mobility model are not acceptable, because the result could be voluntarily manipulated choosing the specific model. Opposite, a model of size-mobility relationship should be fitted to the empirical relation between the particle mobility and diameter, where the diameter is defined independent of the mobility. The concept of mobility diameter is to be reserved for usage as a characteristic of nonspherical particles and defined as the mass diameter of a spherical particle of equal mobility. A brief review of different size-mobility models can be found in a paper by Mäkelä et al. (1996).

ISO15900 Millikan model

Official approximation of the aerosol particle size-mobility relation is decided by the international standard for aerosol particle sizing ISO15900. The standard is based on the classic Stokes-Cunningham-Knudsen-Weber-Millikan model (briefly Millikan model) and prescribes step by step calculations beginning from the diameter of a spherical aerosol particle and finishing with the estimate of its electrical mobility. The standard is limited with air as the carrier gas. Initial data are:

T – absolute temperature of air,
p – pressure of air,
n – number of elementary charges on the particle.
d – diameter of the particle.

Calculations should be made as follows:

Air viscosity
$$\eta = \eta_0 \left(\frac{T}{T_0}\right)^{1.5} \left(\frac{T_0 + S}{T + S}\right), \tag{1}$$

free path
$$l = l_0 \left(\frac{T}{T_0}\right)^{2} \left(\frac{p_0}{p}\right) \left(\frac{T_0 + S}{T + S}\right), \tag{2}$$

Knudsen number
$$Kn = \frac{2l}{d}, \tag{3}$$

slip correction
$$S_C = 1 + Kn\left(A + B\exp\left(-\frac{C}{Kn}\right)\right), \tag{4}$$

particle electric mobility:
$$Z = \frac{ne}{3\pi\eta d} S_C . \tag{5}$$

ISO15900 defines the values of required general constants:

$$\left.\begin{array}{l} l_0 = 67.3 \text{ nm}, \\ T_0 = 296.15 \text{ K}, \\ p_0 = 101.3 \text{ kPa}, \\ S = 110.4 \text{ K}, \\ \eta_0 = 18.3245\ \mu\text{Pa sl}, \end{array}\right\} \tag{6}$$

and of Cunningham-Millikan constants

$$A = 1.165,\ B = 0.483,\ C = 0.997. \qquad (7)$$

Additionally is required the value of elementary charge:

$$\mathrm{e} = 1.6022 \times 10^{-19}\ \mathrm{C}. \qquad (8)$$

The equations above are accessible in a publication by Wiedensohler et al. (2010). The standard values of Cunningham-Millikan constants are acquired from a paper by Kim et al. (2005). The same research team continued the measurements and the last results (Jung, 2011, personal communication) are:

$$A = 1.165,\ B = 0.480,\ C = 1.001. \qquad (9)$$

The procedure that delivers electrical mobility of a single charged particle following the above described rules is called the Millikan *Z*-function below and denoted

$$Z = f_{\mathrm{MillikanZ}}\ (p,\ T,\ d). \qquad (10)$$

This function does not include a size supplement considered in the next section and it represents the *plain Millikan model* of the aerosol particle size-mobility correlation.

Updating the Millikan model

The plain Millikan model is good when particles are much bigger than the gas molecules and small enough to neglect the inertia of the surrounding air. The fine nanometer aerosol particles are not much bigger of the gas molecules. This defect can be roughly recovered by replacing the particle diameter in the Millikan Z-function with the sum of the particle mass diameter and a supplement Δd that includes the diameter of gas molecule (Fernandez de la Mora et al, 2003). The resulting algorithm can be called the *updated Millikan model*. The supplement Δd was estimated in different studies between 0.3 and 0.6 nm.

In case of particles of size about 1 nm another simple correction can be made considering the effect of the mass of the gas molecules m_g like in the kinetic theory and introducing the factor of $(1 + m_g / m)^{1/2}$ (Larriba et al., 2011).

In a paper by Tammet (1995) the kinetic approach was systematically used and some small effects were additionally considered including

- dependence of the law of the reflection of gas molecules on the particle size,
- polarization interaction between charged particles and gas molecules,
- dependence of the collision distance on the interaction energy.

As a result, an improved model *Tammet95* was advanced. Main shortcoming of this model is sophisticated iterative structure that complicates application of the algorithm in actual processing of aerosol measurements. Afterwards, several advanced theoretical models were proposed, e.g. (Li and Wang, 2003; Shandakov et al., 2006). These models are computationally less convenient and did not have applications in aerosol measurements till today. This motivates a search for new algorithms that should well approximate the theoretically founded size-mobility models, be technically convenient in actual data processing, and support simple interpretation when compared with the classic Millikan model.

The idea of the present study is to convert the updated Millikan model flexible while the diameter supplement Δd is considered not as a constant but as a variable depending on the particle size. Additionally, the dependence of required diameter supplement from the air temperature and pressure should be tested as potential arguments of Δd. A method for estimating of variable Δd is:

- choose the parameters of air p, T and of particle ρ, d, where ρ is the density of the particle matter and d is the mass diameter,
- estimate corresponding mobility Z according to measurements
  or calculate it using a precise reference model,
- calculate the diameter d_2 corresponding to Z according to the plain Millikan model,
- estimate $\Delta d = d_2 - d$.

This procedure results in different values of Δd depending on the parameters of air and particle. Next the dependence of Δd on the particle diameter and air parameters is to be approximated with a possibly simple mathematical function $\Delta d \approx f_\Delta (p, T, \rho, d)$ and the new flexibly updated model can be written as

$$Z \approx Z_1 (p, T, d) = f_{\text{MillikanZ}} (p, T, d + f_\Delta (p, T, \rho, d)). \quad (11)$$

Diameter is the main argument of the function $f_\Delta (p, T, \rho, d)$. Some or even all other arguments could be neglected in specific approximations depending on the required accuracy.

Numerical experiments

There was no good set of measurements available in the present study and the method of the reference model is used in the experiments below. The mobility function of the reference model is denoted

$$Z = f_{\text{reference}} (p, T, d). \quad (12)$$

The model *Tammet95* is used as a basis of the reference model in the experiments below. The advanced theoretical models are subjects to be tested in the future research.

The approximations of the air viscosity and free path in *Tammet95* differ from the definitions of ISO15900. This could distort the comparison of the models. Thus the original approximations of viscosity and free path are replaced with approximations acquired from ISO15900 in the final version of the reference model called *Tammet95m*. Different of *Tammet95*, the model *Tammet95m* is restricted with an assumption that the carrier gas is air.

The density of particle matter is expected to be a constant of 2 g cm^{-3} in the experiments below and excluded from the list of explicit arguments of the function f_Δ.

Some curves drawn according to the reference model *Tammet95m* and the Millikan model are shown in Figure 1.

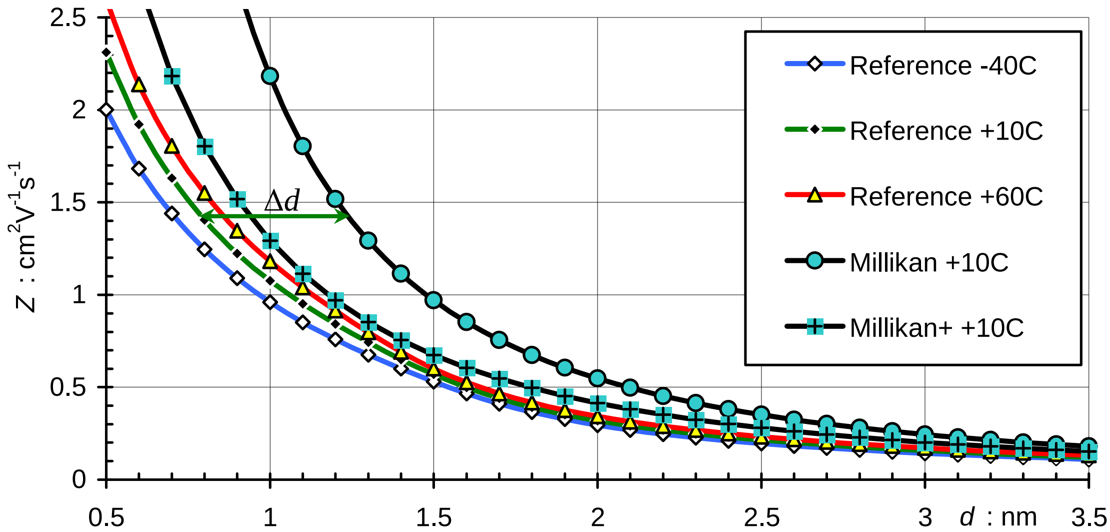

Figure 1. Depencence of electrical mobility on the diameter according to different models at 1013 mb and indicated temperature. “Reference” denotes the model Tammet95m, “Millikan” the plain Millikan model, and “Millikan+” the Millikan model with argument of d + 0.3 nm.

Figure 2 shows the dependence of the estimated Δd on the diameter at three values of the temperature. The asymptotic value of Δd in the model *Tammet95* for air is 0.6 nm that is composed of 0.37 nm collision diameter of gas molecules and of 0.23 nm of extra distance caused by minor effects. The values of Δd are lowered in the range of sub-3 nm particles. Fernandez de la Mora et al. (2003) studied fine nanometer particles down to 2 nm and found that the measurements well fitted with updated Millikan model assuming a constant value of Δd = 0.53 nm. Ku et al. (2009) and Larriba et al. (2011) studied the particles of size down to 1 nm and found that Δd = 0.30 nm is a better approximation. These findings roughly fall in line with calculations illustrated in Figure 2.

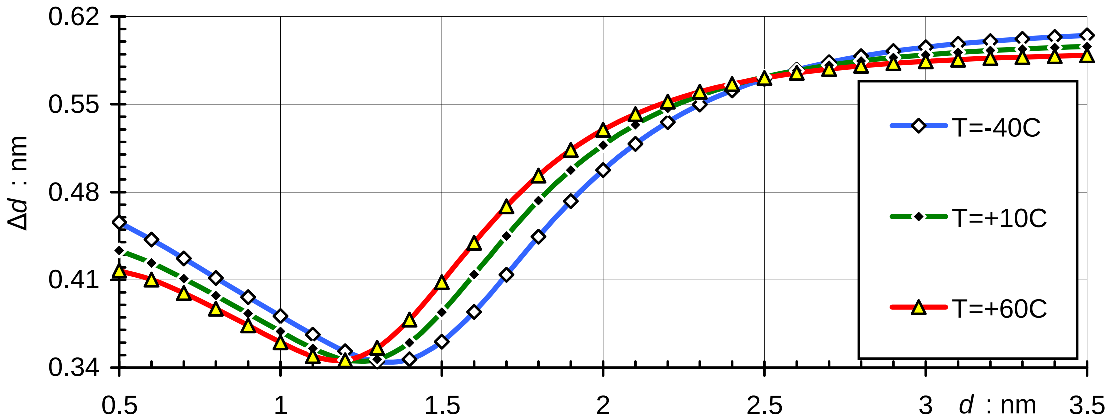

Figure 2. Required supplement of diameter in flexibly updated Millikan model (p = 1013 mb).

The curve T = +10 C° in Figure 2 can be approximated with a simple expression

$$\Delta d \approx f_{\Delta 1}(d) = 0.6 - 0.22 \times \exp(-1.8 \times (d - 1.2)^2) - 0.033 / d, \tag{13}$$

where additional arguments of the function f_Δ are neglected.

If $Z_0 = f_{\mathrm{reference}}(p, T, d)$ and $Z_1 = f_{\mathrm{MillikanZ}}(p, T, d + f_{\Delta 1}(d))$ then the relative error of the model (11, 13) EZ = $(Z_1 - Z_0) / Z_0$. Dependence of the error on particle diameter and air temperature is illustrated in Figure 3.

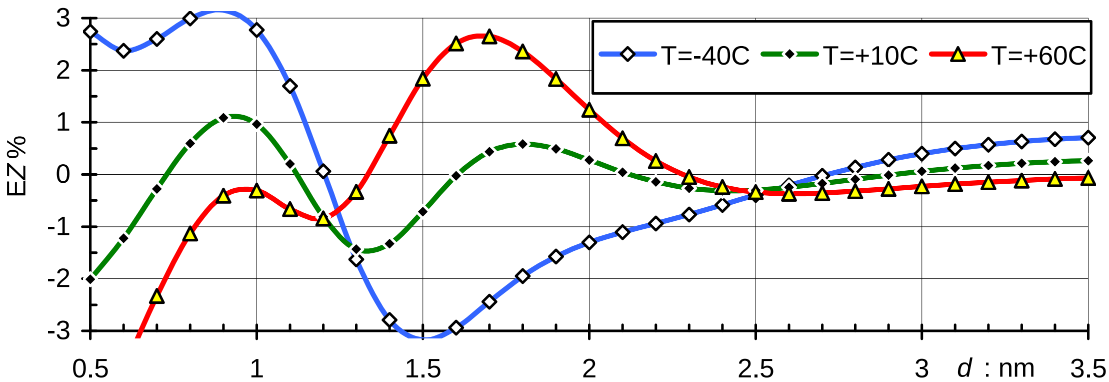

Figure 3. Relative error $(Z_1 - Z_0) / Z_0$ of the approximation (11, 13) at p = 1013 mb.

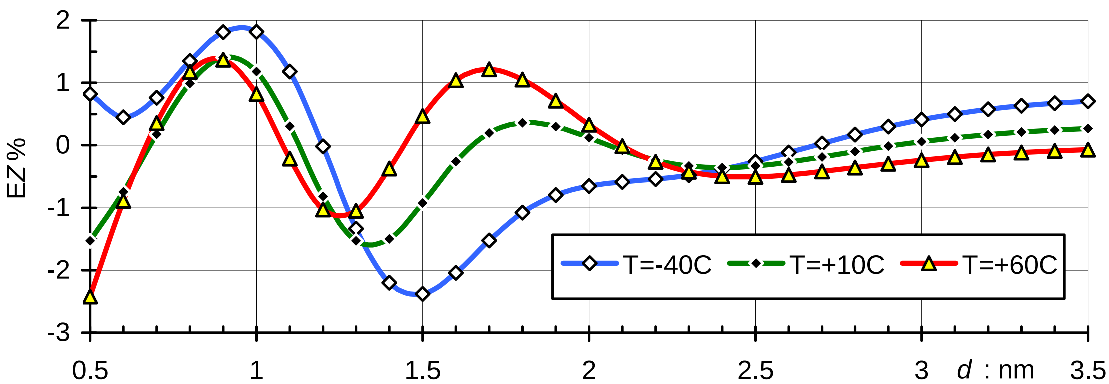

Figure 4. Relative error $(Z_2 - Z_0) / Z_0$ of the approximation (14, 15) at p = 1013 mb.

The temperature effect can be a little suppressed including a small and simple modification into the equation of the size supplement:

$$\Delta d \approx f_{\Delta 2}(d, T) = 0.6 - 0.22 \times \exp(-1.8 \times (d + 0.001\, T_C - 1.2)^2) - 0.033 / d, \qquad (14)$$

where T_C is value of the temperature in Celsius scale. Corresponding model is

$$Z \approx Z_2(p, T, d) = f_{MillikanZ}(p, T, d + f_{\Delta 2}(d, T)). \qquad (15)$$

The relative mobility error of this model is illustrated in Figure 4.

The error of the inverse calculation is estimated in following way. Let $Z_0 = f_{reference}(p, T, d_0)$ and the diameter d_2 is calculated as a function of Z_0 according to the inverse transformation of Equation (15). The absolute diameter error $d_2 - d_0$ is illustrated in Figure 5 and relative error $(d_2 - d_0)/ d_0$ in Figure 6.

Figure 7 illustrates the effect of the air pressure on the error of the model. The pressure effect is presented by deviation of the relative error $(Z_2 - Z_0) / Z_0$ when changing the pressure from 1000 mb to 800 or 1200 mb. The effect appears to be marginal and pressure is to be excluded from the list of explicit arguments of the size supplement.

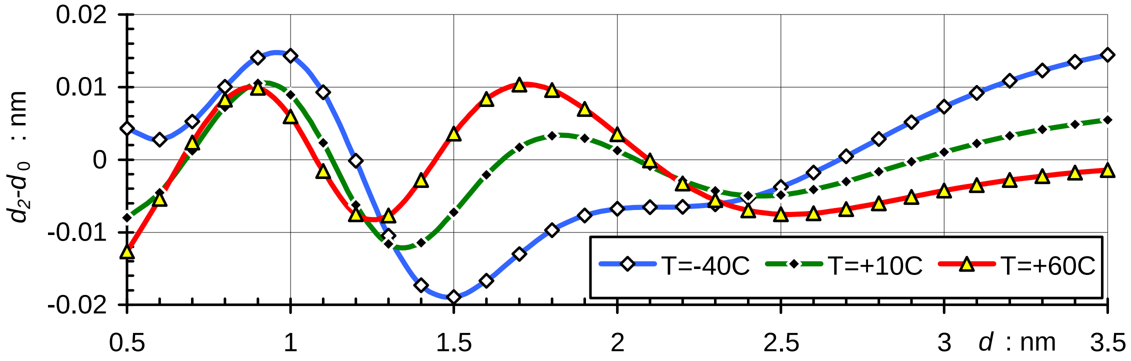

Figure 5. Absolute diameter error of approximation (15) at p = 1013 mb.

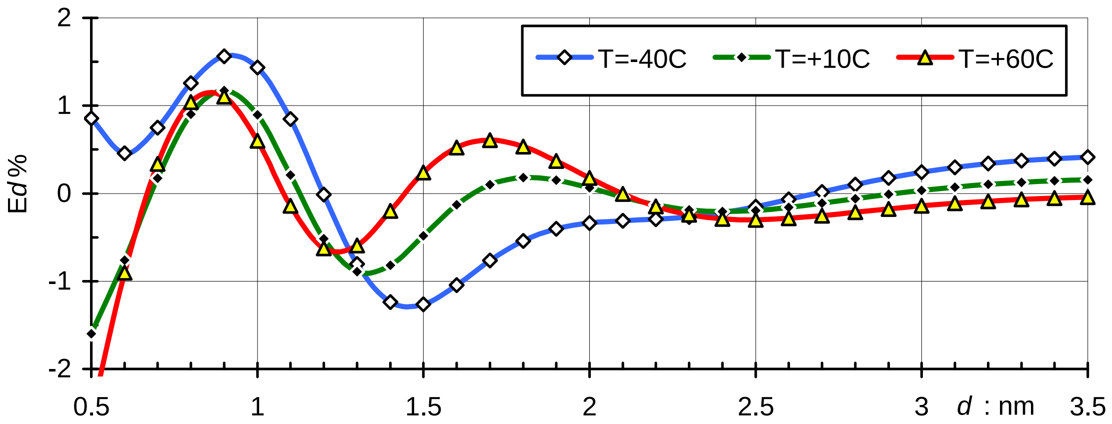

Figure 6. Relative diameter error of approximation (15) at p = 1013 mb..

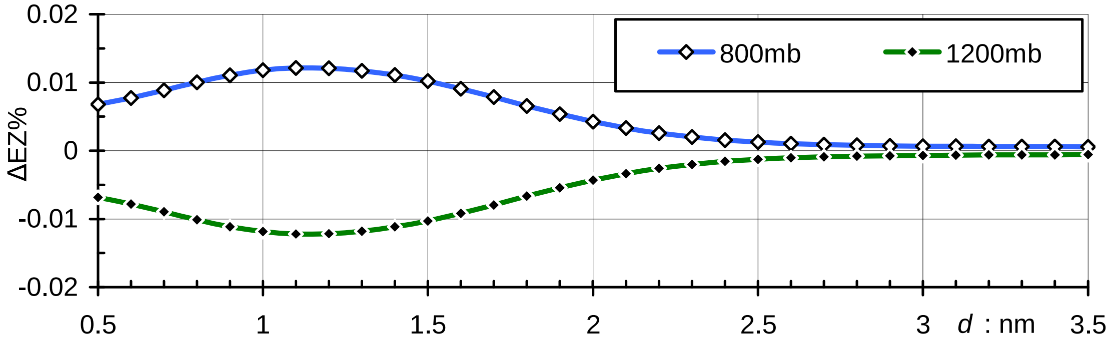

Figure 7. Pressure effect EZ(1000±200 mb) – EZ(1000 mb) at T = 10ºC.

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Recommended algorithms

The Pascal-algorithm of the flexibly updated Millikan model is:

```
function Z_approx_air (millibar, Celsius, d {nm} : real) : real;
  {cm2 V-1 s-1, mathematical approximation of mobility Tammet95m}
  const a = 1.165; b = 0.48; c = 1.001; {Jung et al., 2011}
  var Viscosity, FreePath, Kn, r1, r2, y, dd : real;
  begin
    y := d + 0.001 * Celsius - 1.2;
    dd := d + 0.6 - 0.22 * exp (-1.8 * y * y) - 0.033 / d;
    r1 := (Celsius + 273.15) / 296.15;
    r2 := 406.55 / (Celsius + 383.55);
    Viscosity {microPa s} := 18.3245 * r1 * sqrt (r1) * r2;
    FreePath {nm} := 67.3 * sqr (r1) * (1013 / millibar) * r2;
    Kn := 2 * FreePath / dd;
    if Kn < 0.03 {underflow safe} then y := 0 else y := exp (- c / Kn);
    Z_approx_air :=
            1.6022 * (1 + Kn * (a + b * y)) / (3 * PI * Viscosity * dd);
   end;
```

The inverse conversion can be performed using the algorithm:

```
function d_approx_air (millibar, Celsius, Z {cm2V-1s-1} : real) : real;
 {Uses external function "Z_approx_air". If failed then result = 0}
  var c, d, test : real;
               n : integer;
  begin
    c := 100; n := 0;
    repeat
      n := n + 1;
      d := (0.6 + sqrt (0.36 + 200 * c * Z)) / (c * Z) - 0.4;
      test := Z_approx_air (millibar, Celsius, d);
      c := (1.2 / (d + 0.4) + 200 / sqr (d + 0.4)) / test;
    until (abs (test / Z - 1) < 1e-6) or (n = 99);
    if n < 99 then d_approx_air := d else d_approx_air := 0;
  end;
```

Test of mobility functions using the program *A_tools*

Electrical mobility (cm2V-1s-1) at 1013 mb and 10 C:

d:nm	Millikan+	Tammet95	Reference	Approx	T95/Ref	Mkn+/Ref	Appr/Ref
0.6	2.48522	1.88764	1.92191	1.90759	0.9822	1.2931	0.9925
0.8	1.66449	1.38013	1.40518	1.41913	0.9822	1.1845	1.0099
1.0	1.19233	1.05643	1.07559	1.08827	0.9822	1.1085	1.0118
1.5	0.62271	0.55834	0.56846	0.56319	0.9822	1.0954	0.9907
2.0	0.38187	0.31324	0.31891	0.31930	0.9822	1.1974	1.0012
3.0	0.18597	0.15451	0.15730	0.15739	0.9823	1.1823	1.0006
5.0	0.07246	0.06379	0.06494	0.06511	0.9823	1.1159	1.0026
7.0	0.03839	0.03481	0.03543	0.03549	0.9824	1.0835	1.0017
10.0	0.01943	0.01804	0.01836	0.01838	0.9825	1.0587	1.0010
20.0	0.00514	0.00491	0.00499	0.00499	0.9830	1.0291	1.0003
50.0	0.00091	0.00088	0.00090	0.00090	0.9843	1.0112	1.0001

Millikan+ or Mkn+ is the ISO15900 algorithm updated with a constant diameter supplement of 0.3 nm. Tammet95 or T95 is the original function from paper (Tammet, 1995) where the three Millikan constants are updated but the gas viscosity and free path are calculated according to original approximations that differ from ISO15900. The reference model Tammet95m (presented in the next page) is basically the same, but the air viscosity and free path are calculated according to ISO15900. Approx or Appr is the above described function *Z_approx_air*.

Excel macros

Two macros below are equivalent of Pascal-functions. Different of Pascal function, the VBA function *d_approx_air* is self-contained and does not require any external subroutine.

```
Sub Z_approx_air() ' HT 20120118
  n = ActiveCell.Value
  For i = 0 To n - 1
    mb = ActiveCell.Offset(i, -3).Value
    Celsius = ActiveCell.Offset(i, -2).Value
    d = ActiveCell.Offset(i, -1).Value
    y = d + 0.001 * Celsius - 1.2
    dd = d + 0.6 - 0.22 * Exp(-1.8 * y * y) - 0.033 / d
    r1 = (Celsius + 273.15) / 296.15
    r2 = 406.55 / (Celsius + 383.55)
    vi = 18.3245 * r1 * Sqr(r1) * r2
    fp = 67.3 * r1 * r1 * (1013 / mb) * r2
    Kn = 2 * fp / dd
    y = 0
    If Kn > 0.03 Then y = Exp(-1.001 / Kn)
    Z = 1.6022 * (1 + Kn * (1.165 + 0.48 * y)) / (3 * 3.14159 * vi * dd)
    ActiveCell.Offset(i, 0) = Z
  Next i
End Sub

Sub d_approx_air() ' HT 20120118
  n = ActiveCell.Value
  For i = 0 To n - 1
    mb = ActiveCell.Offset(i, -3).Value
    Celsius = ActiveCell.Offset(i, -2).Value
    Z = ActiveCell.Offset(i, -1).Value
    r1 = (Celsius + 273.15) / 296.15
    r2 = 406.55 / (Celsius + 383.55)
    vi = 18.3245 * r1 * Sqr(r1) * r2
    fp = 67.3 * r1 * r1 * (1013 / mb) * r2
    c = 100
    m = 0
    OK = False
    While Not OK
       m = m + 1
       d = (0.6 + Sqr(0.36 + 200 * c * Z)) / (c * Z) - 0.4
       y = d + 0.001 * Celsius - 1.2
       dd = d + 0.6 - 0.22 * Exp(-1.8 * y * y) - 0.033 / d
       Kn = 2 * fp / dd
       y = 0
       If Kn > 0.03 Then y = Exp(-1.001 / Kn)
       test = 1.6022 * (1 + Kn * (1.165 + 0.48 * y)) / (3 * 3.14159 * vi * dd)
       c = (1.2 / (d + 0.4) + 200 / ((d + 0.4) * (d + 0.4))) / test
       OK = (Abs(test / Z - 1) < 0.000001) Or (m = 99)
    Wend
    If m > 99 Then d = 0
    ActiveCell.Offset(i, 0) = d
  Next i
End Sub
```

Working with imported macros may cause problems with security settings. A simple way in case of a single application is following (conversion of mobility to diameter in the example):

- Select the text of both macros *Sub Z_approx_air* and *Sub d_approx_air* above, press Ctrl+C, the text of macros is stored in the clipboard.
- Open an Excel spreadsheet, select tools ==> macro ==> record new macro, remember the name of the macro, press OK (a tiny recorder toolbar appears), press stop button on lower left corner of the recorder toolbar (recorder toolbar disappears), a new empty macro is recorded.
- Select tools ==> macro ==> macros ==> name of the new macro ==> edit (VBA window appears), select the text of the recorded empty macro, press Ctrl+V (the code of empty macro will be replaced with the code of two mobility functions).

- Go to the Excel window, fill three neighbor columns in the spreadsheet with values of air pressure (mb), temperature (Celsius), and mobility ($cm^2V^{-1}s^{-1}$), write number of rows to be processed into the cell just right of the first value of the mobility (see the example) and click this cell. All data must be presented as values.
- Select tools ==> macro ==> macros ==> *d_approx_air* ==> Run.

Example of results:

p:mb	T:C	Z:cm2V-1s-1	d:nm
1013	0	0.5	1.581632069
1013	10	0.5	1.593991555
990	20	1	1.095856087
990	20	2	0.598609765

Here was number 4

Afterwards, the macros can be saved into the personal workbook and repeatedly used in different spreadsheets (the security may be required to set to a lower level). In this case is enough to fill three columns with data and one cell with number of rows to be processed (this cell must stay active), and run macro.

The reference model

```
function electrical_mobility_air                                  {Tammet95m}
                   (Millibar,
                    Celsius,
                    ParticleDensity {g cm-3, for cluster ions typically 2.08},
                    ParticleCharge  {e, for cluster ions 1},
                    MassDiameter    {nm} : real) : real;
  function Omega11 (x : real) : real; {*(1,1)*(T*) for (*-4) potential}
    var p, q : real;                  {and elastic-specular collisions}
    begin
      if x > 1 then Omega11 := 1 + 0.106 / x + 0.263 / exp ((4/3) * ln (x))
      else begin p := sqrt (x); q := sqrt (p);
        Omega11 := 1.4691 / p - 0.341 / q + 0.181 * x * q + 0.059 end;
    end;

  const GasMass = 28.96; {amu} Polarizability = 0.00171; {nm3}
        a = 1.165; b = 0.48; c = 1.001; {the slip factor coefficients}
        ExtraDistance = 0.115 {nm}; TransitionDiameter = 2.48 {nm};
  var   Temperature, GasDiameter, Viscosity, FreePath, DipolEffect,
        DeltaTemperature, CheckMark, ParticleMass, CollisionDistance,
        Kn, Omega, s, x, y, r1, r2 : real;
  begin
    if MassDiameter < 0.2 then {emergency exit}
    begin electrical_mobility_air := 1e99; exit; end;
    Temperature := Celsius + 273.15;
    r1 := Temperature / 296.15; r2 := 406.55 / (Temperature + 110.4);
    Viscosity {microPa s} := 18.3245 * r1 * sqrt (r1) * r2;
    FreePath {nm} := 67.3 * sqr (r1) * (1013 / millibar) * r2;
    ParticleMass {amu} := 315.3 * ParticleDensity * exp (3 * ln (MassDiameter));
    DeltaTemperature := Temperature;
    repeat
      CheckMark := DeltaTemperature;
      GasDiameter {nm} := 0.3036 * (1 + exp (0.8 * ln (44 / DeltaTemperature)));
      CollisionDistance {nm} := MassDiameter / 2 + ExtraDistance +
                                GasDiameter / 2;
      DipolEffect := 8355 * sqr (ParticleCharge) * Polarizability /
                          sqr (sqr (CollisionDistance));
      DeltaTemperature := Temperature + DipolEffect;
    until abs (CheckMark - DeltaTemperature) < 0.01;
    if ParticleCharge = 0 then Omega := 1
                          else Omega := Omega11 (Temperature / DipolEffect);
    Kn := FreePath / CollisionDistance;
    if Kn < 0.03 {underflow safe} then y := 0 else y := exp (- c / Kn);
    x := (273.15 / DeltaTemperature) *
         exp (3 * ln (TransitionDiameter / MassDiameter));
    if x > 30 {overflow safe} then s := 1
      else if x > 0.001
      then s := 1 + exp (x) * sqr (x / (exp (x) - 1)) * (2.25 / (a + b) - 1)
      else {underflow safe}  s := 1 + (2.25 / (a + b) - 1);
    electrical_mobility_air := 1.6022 * ((2.25 / (a + b)) / (Omega + s - 1)) *
                           sqrt (1 + GasMass / ParticleMass) *
                           (1 + Kn * (a + b * y)) /
                           (6 * PI * Viscosity * CollisionDistance);
  end;
```