New Instrument for Measuring Water Vapor Flux Density from Arbitrary Surfaces

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Abstract

We report the development of a new instrument, the AquaFlux, for measuring water vapor flux density from arbitrary surfaces, including *in-vivo* measurements of transepidermal water loss (TEWL), skin surface water loss (SSWL) and perspiration. It uses a closed measurement chamber equipped with an electronically cooled condenser, to maintain a precisely reproducible microclimate adjacent to the test surface under all ambient conditions. The condenser creates a diffusion vapor density gradient, from which the flux density can be measured.

We explore the properties of the AquaFlux by means of a mathematical model, which was also adapted for calculating comparable properties of open-chamber instruments. In this way, we found the intrinsic sensitivity of the AquaFlux to be approximately 40% higher than that of an open-chamber instrument. However, in an experimental comparison of volar forearm TEWL measurements between the AquaFlux and an open-chamber Evaporimeter, we found a tenfold difference in coefficient of variation. We attribute the much lower than predicted sensitivity of the Evaporimeter to extrinsic noise from diffusion zone instability induced by ambient air movements. The mathematical model was also used to calculate the relative humidity immediately above the test surface, where we found values that generally differed from ambient values in both instrument types. The effect of this on TEWL measurements is discussed in detail. It is concluded that, during the relatively short time of measurement, such microclimate changes affect mainly the transient SSWL component rather than the underlying TEWL. Finally, the model was used to estimate the flux density that would cause the relative humidity at the test surface to reach 100%. This sets an upper limit to the flux densities that can be measured and may be a cause of instrument non-linearity at high flux densities, as microclimate saturation is approached.

Introduction

Water vapor flux density measurement, from which transepidermal water loss (TEWL), skin surface water loss (SSWL) and perspiration rate can be determined, is of fundamental importance in skin bioengineering. For this reason, a wide range of measurement methods have been devised, as reviewed by Elsner *et al.*, for example [1]. Today, the diffusion gradient method of Nilsson [2] is the most widely used, with several implementations commercially available. Its most distinctive feature is an open measurement chamber, which is said to maintain the relative humidity (RH) in the air above the test surface close to ambient levels, thus keeping changes in test surface microclimate to a minimum. However, the open chamber is vulnerable to disturbance from ambient air movements that can cause the readings to fluctuate. This limitation is well recognized, and detailed guidelines have been published to help researchers get the best from these instruments [3-4].

Recently, a number of new instruments have come onto the market. They make use of three different measurement principles, but a closed measurement chamber is a key feature in all of them. They include flowing air instruments [5] (*e.g.*, Skinos, Japan, Asahi Biomed, Japan), stagnant chamber instruments [6] (*e.g.*, Delfin Technology, Finland, Nikkiso-YSI, Japan) and a microclimate-controlled instrument [7-8] (*e.g.*, Biox Systems, U.K.).

In this paper, we focus on the microclimate-controlled instrument, the AquaFlux. It uses a closed measurement chamber to protect the diffusion zone within it from ambient air movements. Vapor build-up within the chamber is avoided by trapping water molecules as ice on an electronically cooled condenser opposite the measurement orifice. The effect is to establish a vapor density gradient parallel to the axis of the chamber, from which the flux density can be determined. The condenser provides the added benefit of maintaining the humidity adjacent to the test surface at precisely known values, irrespective of ambient conditions. These features, together with a precise gravimetric calibration, provide new levels of performance and flexibility, ensuring that measurements performed at different times or with different instruments, indoors or outdoors, can be meaningfully compared.

In this paper, we present a theoretical analysis of the AquaFlux measurement principle and compare the main aspects of its performance with those predicted for Nilsson's open-chamber measurement principle.

Theory

Fig. 1 shows a schematic representation of the AquaFlux measurement chamber. It is in

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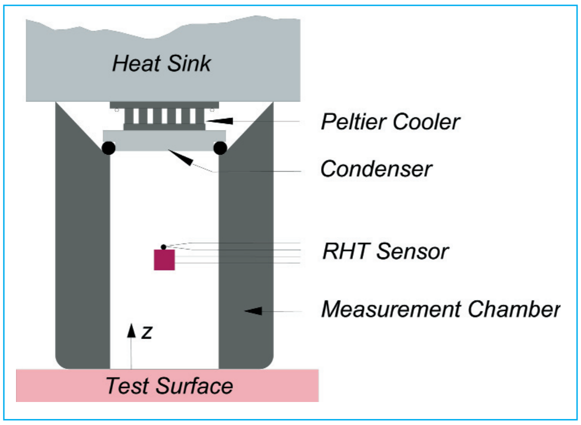

Fig. 1: Schematic diagram of the AquaFlux measurement chamber

the form of a hollow cylinder whose lower end acts as a measurement orifice that is placed into contact with the test surface. Its upper end is sealed with an aluminum condenser plate that is maintained at a precisely controlled temperature by means of a Peltier cooler and associated heat sink. In the center of the chamber is a sensor combination for relative humidity (RH) and temperature (abbreviated to RHT sensor), designed to measure absolute humidity as vapor pressure or vapor density. The main approximations used to model this system are:

(a) The thermal conductivity and water affinity of the cylindrical wall of the measurement chamber are assumed to be vanishingly low. The effect of these approximations is to reduce the model to one spatial dimension.

(b) The internal dimensions of the measurement chamber are assumed to be small enough to suffocate natural convection and other bulk air movements. In Fluid Dynamic terms, this requires the Rayleigh number of the chamber to be below the critical value for its geometry [9].

With these assumptions, the measurement chamber can be described by a *z*-axis parallel the chamber axis, with the test surface at $z = 0$, the condenser at $z = L_C$ and the RHT sensor at $z = L_S$.

AquaFlux Measurement Principle

In the absence of bulk air movements, the migration of water molecules in the chamber can be calculated from Fick's first law of diffusion, the one-dimensional form of which is:

$$J = -D_{VA}\frac{\partial \rho_V}{\partial z} \tag{1}$$

where J is the water vapor flux density from the test surface, D_{VA} is the molecular diffusion coefficient for water vapor in air (D_{VA} = 2.42x10^{-5} m^2 s^{-1} at room temperature) and ρ_V is the water vapor density (or concentration). If J is constant, then a steady-state vapor distribution is eventually established, where the vapor entering the chamber at $z = 0$ is removed by the condenser at $z = L_C$. This steady-state solution is characterized by a constant gradient, $\partial \rho_V / \partial z$, which can be written in terms of quantities accessible to measurement, *i.e.*, the vapor densities ρ_{VS} and ρ_{VC} at positions $z = L_S$ and $z = L_C$ respectively:

$$J = D_{VA}\left(\frac{\rho_{VS} - \rho_{VC}}{L_C - L_S}\right) \tag{2}$$

Vapor density in Eq. (2) is more conveniently expressed in terms of RH using the defining equation

$$\rho_V = \chi \ \rho_{VE}(\theta) \tag{3}$$

where χ is fractional RH, θ is temperature and ρ_{VE} is equilibrium (or saturation) vapor density that can be computed from a parameterization of the saturation vapor pressure curve (*e.g.*, [10]) and the ideal gas law. The vapor density at the RHT sensor can be calculated from Eq. (3) using measured values of RH and temperature, denoted, respectively by χ_S and θ_S. The vapor density immediately adjacent to the condenser, where vapor and ice are assumed to be in thermodynamic equilibrium, is equal to the equilibrium vapor density at the temperature of the condenser, $\theta = \theta_C$. With these quantities, Eq. (2) becomes

$$J = \frac{D_{VA}}{L_C - L_S}\left[\chi_S \ \rho_{VE}(\theta_S) - \rho_{VE}(\theta_C)\right] \tag{4}$$

which allows the flux density to be calculated from measurements of χ_S, θ_S and θ_C.

The microclimate immediately above the test surface can be specified by the temperature and RH of the air at $z = 0$. The temperature of the air, θ_0, is taken as equal to the temperature of the test surface. An expression for RH at $z = 0$ can be derived from Eqs. (2) and (3) by replacing $z = L_S$ with $z = 0$, to give

$$\chi_0 = \frac{\rho_{VE}(\theta_C) + J \ L_C / D_{VA}}{\rho_{VE}(\theta_0)} \tag{5}$$

Nilsson Measurement Principle

Equivalent expressions to Eqs. (4) and (5) can be derived for the Nilsson measurement principle by (a) replacing the single RHT sensor at $z = L_S$ with two RHT sensors at $z = L_{S1}$ and $z = L_{S2}$, and (b) replacing the condenser boundary condition at $z = L_C$ with an ambient atmosphere boundary condition at the open end of the measurement chamber, at $z = L_A$. Although this boundary with the ambient atmosphere is less well defined than the equivalent boundary with the condenser, the model is nevertheless useful for describing the main attributes of the measurement principle. The expression for the flux density works out to

$$J = \frac{D_{VA}}{L_{S2} - L_{S1}}\left[\chi_{S1} \ \rho_{VE}(\theta_{S1}) - \chi_{S2} \ \rho_{VE}(\theta_{S2})\right] \tag{6}$$

and the RH immediately above the test surface becomes

$$\chi_0 = \frac{\chi_A \ \rho_{VE}(\theta_A) + J \ L_A / D_{VA}}{\rho_{VE}(\theta_0)} \tag{7}$$

Results and Discussion

Sensitivity Results

The dependence of the intrinsic sensitivity on the noise contributions from the sensors can be estimated using the error propagation equation of Gaussian statistics [11]. For the AquaFlux, the noise standard deviation of the flux density, σ_J, can be calculated from the noise standard deviations of the RH and temperature sensors, $\sigma_{\chi S}$ and $\sigma_{\theta S}$, respectively, using

$$\sigma_J^2 = \left(\frac{\partial J}{\partial \chi_s}\right)^2 \sigma_{\chi s}^2 + \left(\frac{\partial J}{\partial \theta_s}\right)^2 \sigma_{\theta s}^2 \tag{8}$$

The equivalent expression for a Nilsson instrument has four terms, because both RHT sensors need to be taken into account. Calculations with Eqs. (4) and (6) show that, with identical sensors, the intrinsic sensitivity of the AquaFlux, calculated as σ_J^{-1}, is approximately 40% higher than that of a Nilsson instrument, because the AquaFlux uses a single RHT sensor, whereas the Nilsson instrument uses two.

A more realistic assessment of sensitivity than intrinsic noise is the repeatability of measurements in practice. This was assessed in two independent experiments of repeated volar forearm TEWL measurements using the AquaFlux and an open-chamber instrument (Servo Med Evaporimeter Model EP3, Kinna, Sweden). Ten TEWL measurements, spaced ten minutes apart, were performed with each instrument. The environmental conditions in the two laboratories were similar and within the range recommended in the EEMCO Guidelines [4]. The AquaFlux was powered up approximately 15 minutes before the start of the experiment and zeroed before the first measurement. For each measurement, the probe was placed onto the skin surface for about one minute, until the flux reading had stabilized. The average value of TEWL over the following 10 seconds was then recorded. The measurement orifice of the probe was then closed until the next measurement, in order to reduce the humidity at the RHT sensor and ice build-up on the condenser. The Evaporimeter was powered up 24 hours before the start of the experiment and zeroed before the first measurement. For each measurement, the probe was placed onto the skin surface for one minute to equilibrate. The average value of TEWL over the following 15 seconds was then recorded. The effect of ambient air movements during this time was minimized by placing the open end of a plastic beaker over the measurement chamber. The 2-litre volume of the beaker was thought to be adequate to avoid significant build-up of water vapor within it during this time. After the measurement, the beaker was removed and aired. The probe was also removed and allowed to reach a zero reading before the next measurement. The results, presented in **Fig. 2**, show that the repeatability of AquaFlux measurements is more than an order of magnitude better than that of the Evaporimeter, with coefficients of variation of 1.4% and 16%, respectively.

Sensitivity Discussion

Under the conditions of the experiment, the AquaFlux was found to be more than an order of magnitude more sensitive than the Evaporimeter, although the calculated difference of intrinsic sensitivity is only ~40%. This indicates that extrinsic noise dominates in the Evaporimeter, with diffusion zone instability the most likely cause. Diffusion is a highly inefficient transport mechanism that can only be observed when no other mechanisms operate, as can easily be demonstrated with milk and tea. Therefore, even a slight perturbation of the vapor distribution in the diffusion zone by ambient air movements can cause the readings to fluctuate. Commonly used covering boxes protect against draughts, but do nothing to suppress natural convection, driven by unavoidable temperature differences between skin and ambient air. Chimneys, foam rubber plugs and other devices attached directly to the open end of the measurement chamber may provide some increased protection, but at the expense of intrinsic sensitivity and changed calibration.

Microclimate Results

Typical microclimate RH values at the skin surface, calculated from Eqs. (5) and (7), are presented in **Fig. 3**. The calculations used a skin temperature of $\theta_S = 30°C$ and representative constants for the two instruments (AquaFlux: $L_C = 16$ mm, $\theta_C = -7.4°C$; Nilsson: $L_A = 20$ mm, $\theta_A = 21°C$).

Microclimate Discussion

An interesting observation from **Fig. 3** is that skin surface RH values generally differ from ambient RH values, being lower than ambient at low flux densities. In the case of the AquaFlux, this is due mainly to the action of the condenser, which controls the microclimate RH independently of ambient RH. In the case of Nilsson instruments, this is due to the

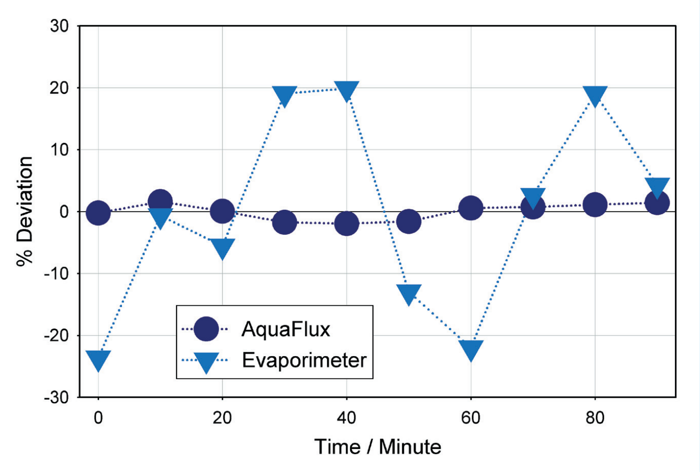

Fig. 2: Repeatability of volar forearm TEWL measurements using the AquaFlux and an Evaporimeter Model EP3. The respective coefficients of variation are 1.4% and 16%.

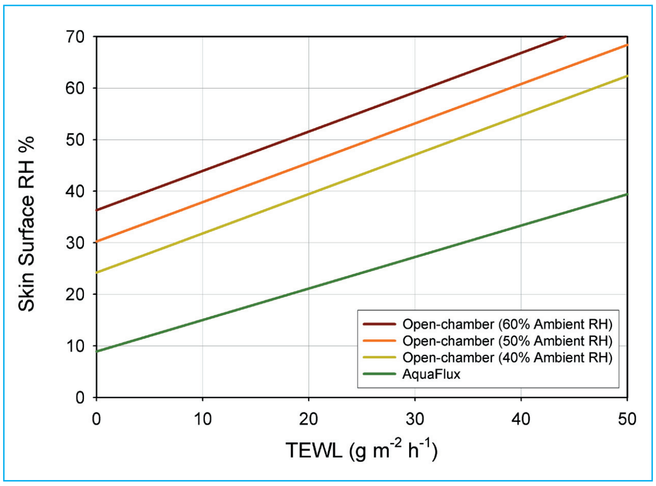

Fig. 3: Typical microclimate RH values at the skin surface for AquaFlux and open measurement chambers

higher than ambient temperature of the air adjacent to the skin.

This raises the question whether such changes of microclimate RH in open-chamber measurements represent a change from pre-measurement conditions or not. If, as is sometimes argued (*e.g.*, [2,3]), there is a stagnant diffusion zone blanketing the skin in its natural condition, then this would have similar properties to the diffusion zone in the open chamber, and the microclimate RH would be little affected. More realistic in our view, is to recognize the intrinsic instability of such diffusion zones (body movements, ambient air movements, natural convection, etc.) and to assume that pre-measurement conditions of exposed skin resemble ambient conditions. The transition from exposed pre-measurement conditions to protected diffusion zone conditions would then represent a significant change of microclimate.

Another aspect that requires further discussion is the effect of microclimate RH on TEWL. We base our understanding on a simple two-step model of TEWL as follows:

Step 1: Diffusion within the Skin

The concentration of free liquid water (*i.e.*, water that is able to migrate) is assumed to be high in the living epidermis, decreasing continuously, without partitioning interfaces towards the surface of the stratum corneum. This concentration profile causes free water to diffuse towards the surface, in accordance with a modified form of Fick's first law, where the diffusion coefficient varies from point to point, depending, among other variables, on temperature and hydration at that point. With these assumptions, the rate of diffusion at any point depends only on the diffusion coefficient and gradient of free water concentration at that point. Simply wetting the stratum corneum surface, for example, would not alter this rate until the surface water has had time to penetrate into the stratum corneum and cause the local concentration gradient and/or diffusion coefficient to change.

Step 2: Evaporation from the Surface

The surface of the stratum corneum is a discontinuous boundary across which liquid water partitions into the vapor phase with an accompanying decrease of concentration of more than three orders of magnitude. Evaporation is a two-way process, whose net rate is the difference between the rate at which water molecules leave the surface and the rate at which they re-condense. At zero RH, there is no re-condensation and the net evaporation rate is a maximum; at 100% RH, the re-condensation rate is equal to the evaporation rate and no liquid is lost from the surface. The amount of free surface water therefore depends partly on the net rate of evaporation and partly on the rate of supply by diffusion from below, described in **Step 1**.

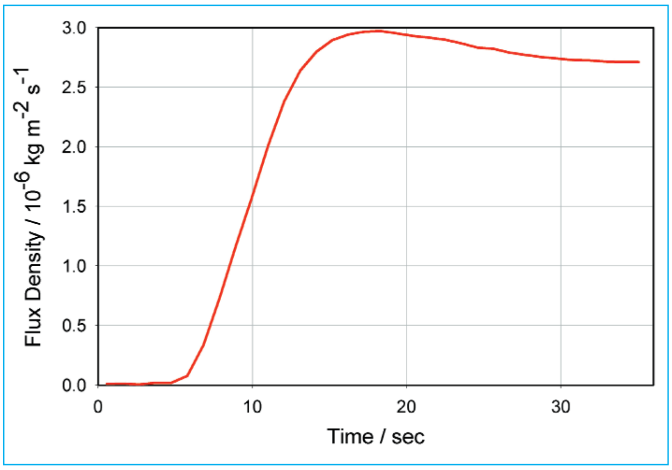

Fig. 4: Typical AquaFlux signal from the volar forearm, showing a small SSWL peak before settling to a steady TEWL reading

According to the above model, the effect of changing the skin surface RH, when a TEWL measurement is initiated, is twofold. Effect 1, associated with **Step 2**, is a change of partitioning between surface water and water vapor. Effect 2, associated with **Step 1** is a change in the rate of diffusion of liquid water through the stratum corneum, as the new level of surface wetness causes the concentration gradients and diffusion coefficients within the stratum corneum to change. The key point is that the timescales of these two effects are different. Effect 1 occurs quickly, in typically one minute or less. The evidence for this comes from Aqua Flux signals such as in **Fig. 4**, where the peak can be ascribed to loss of surface water (SSWL) caused by the lowering of microclimate RH during the measurement. Effect 2 occurs more slowly, typically in some hours. The evidence for this comes from occlusion experiments such as [12] and from our own experiments with the AquaFlux [13], where a mean rate of decrease of TEWL of approximately 0.1% per minute was measured. Therefore, both free surface water and the characteristic timescales of change need to be taken into account in assessing the effect of microclimate RH on measurements.

Another inference that can be drawn from Eqs. (5) and (7) is that the microclimate RH can approach 100% at high flux densities, at which point liquid water would accumulate on the test surface. This sets an upper limit to the flux densities that can be measured. The known underestimation of high TEWL values (*e.g.*, [3]) may also be caused by this effect, given that measurement non-linearities are likely to become apparent before saturation at the test surface is reached. Saturation flux densities for the examples of **Fig. 3** are 4.2×10^{-5} kg m^{-2} s^{-1} (150 g m^{-2} h^{-1}) for the AquaFlux and from 2.5×10^{-5} kg m^{-2} s^{-1} (91 g m^{-2} h^{-1}) for a Nilsson instrument operated in an ambient RH of 50%.

Conclusions

A clear understanding of the interaction of a measurement device with its measured medium is essential, if the information it provides is to be correctly interpreted. The model and supporting experimental evidence presented in this paper provide a basis for understanding three aspects.

Firstly, the model was used to estimate intrinsic measurement sensitivity, finding the AquaFlux to be ~40% more sensitive than an open-chamber instrument. However, in an experimental comparison of volar forearm TEWL measurements using the AquaFlux and an Evaporimeter, we found a tenfold difference in coefficient of variation. We attribute the much lower than predicted sensitivity of the Evaporimeter to extrinsic noise associated with diffusion zone instability in its open measurement chamber.

Secondly, the model was used to calculate microclimate RH, finding it to differ from ambient RH in both AquaFlux and open chambers. We argue however that, during the relatively short time of measurement, this change of microclimate RH would affect mainly the SSWL component of the signal without significantly altering the underlying TEWL.

Thirdly, the model was used to estimate the water vapor flux density that would cause the microclimate RH to reach 100%. We propose this microclimate saturation as the cause of a non-linear instrument response at high flux densities.

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