metrology

MDPI

Conference Report

Report of the CCU/CCQM Workshop on “The Metrology of Quantities Which Can Be Counted”

Richard J. C. Brown [1,2,*], Bernd Güttler [3,*], Pavel Neyezhmakov [4], Michael Stock [5], Robert I. Wielgosz [5], Stefan Kück [3] and Konstantina Vasilatou [6]

1 National Physical Laboratory, Hampton Road, Teddington TW11 0LW, Middlesex, UK
2 School of Mathematics and Physics, University of Surrey, Guildford GU2 7XH, Surrey, UK
3 Physikalisch-Technische Bundesanstalt, 38116 Braunschweig, Germany
4 National Scientific Centre “Institute of Metrology”, UA-61002 Kharkiv, Ukraine
5 Bureau International des Poids et Mesures, Pavillon de Breteuil, 92312 Sèvres, CEDEX, France
6 Federal Institute of Metrology METAS, Lindenweg 50, 3003 Bern-Wabern, Switzerland
* Correspondence: richard.brown@npl.co.uk (R.J.C.B.); bernd.guettler@ptb.de (B.G.)

Abstract: This article provides a report of the recent workshop on “The metrology of quantities which can be counted” organised jointly by the International Committee for Weights and Measures’ Consultative Committees for Amount of Substance (CCQM) and for Units (CCU). The workshop aimed to trigger a discussion on counting and number quantities across the metrological community so that a common understanding of counting and a common nomenclature could be achieved and there was clarity on the differences between these increasingly important concepts. This article details the background to the workshop, provides a summary of the presentations given and the discussions on the topics raised. It also reports the conclusions, agreed actions and next steps resulting from the workshop.

Keywords: metrology; units; dimensionless quantities; one; counting

check for updates

Citation: Brown, R.J.C.; Güttler, B.; Neyezhmakov, P.; Stock, M.; Wielgosz, R.I.; Kück, S.; Vasilatou, K. Report of the CCU/CCQM Workshop on “The Metrology of Quantities Which Can Be Counted”. *Metrology* **2023**, 3, 309–324. https://doi.org/10.3390/metrology3030019

Academic Editor: Han Haitjema

Received: 8 August 2023
Revised: 22 August 2023
Accepted: 25 August 2023
Published: 4 September 2023

1. Introduction

Ever since dimensional analysis was first considered, quantities with the unit one, symbol 1, have been a perennially difficult subject for metrology, and this continued even after the International System of Units (SI) was formalised [1]. These are either derived quantities where the dimensions of the SI base units comprising the quantity cancel, e.g., m/m, or quantities that cannot be expressed in base units of the SI at all, but instead that express a number of entities, e.g., the number of cells.

The debate about how to treat quantities with the unit one within the SI was reignited in the lead up to the major revision of the SI in 2019. Primarily, this was because there was renewed discussion concerning the status of angle within the SI [2] being a derived or base unit. Whilst no consensus was reached on this issue, and the status quo was maintained in the 9th Edition of the SI Brochure, some progress was made with understanding how some of the possible confusion surrounding the expression of angle and other ratio quantities could be mitigated. However, it was clear that quantities relating to quantities which can be counted (number of entities, processes, or other phenomena) had not received sufficient attention during this process and yet these quantities were becoming increasingly important in metrology for two reasons. First, the redefinition of the mole had brought into sharper focus the concept of number of entities since the new definition explicitly relied on this concept (rather than implicitly as it did prior to 2019) [3]. Second, the use of these number quantities is expanding rapidly as metrology becomes more embedded in biological measurements and also emerging regulatory measurements such as particle number concentrations from engine emissions. The SI brochure already acknowledges the relevance of “quantities that cannot be described in terms of the seven base quantities of the SI but have the nature of a count" with the associated unit one. This guidance may be interpreted in a number of ways and raises the question of how the unit one is recognized and realized within the SI system. This issue is further complicated by the infinitely many quantities that can be expressed as a number of entities (for example 'molecules', 'cycles' or 'copies'). The lack of specific guidance in this area is potentially far reaching. It is not yet clear if the result of such quantities can and should be expressed in terms of the SI and whether the notion of traceability to the SI extends to these quantities. This also applies to the question of which communities and International Committee for Weights and Measures' Consultative Committees should take responsibility for standardizing measurements for such quantities [4].

As a result, a workshop was conceived and organised jointly by the Consultative Committee for Units (CCU) and the Consultative Committee for Amount of Substance (CCQM) to discuss these issues and suggest possible ways to improve the understanding of these quantities. It was organised by a Workshop Steering Committee comprising P. Neyezhmakov (NSC-IM), R. J. C. Brown (NPL), B. Güttler (PTB), M. Stock (BIPM) and R. I. Wielgosz (BIPM). The objectives of the workshop were:

- To trigger a discussion on counting and number quantities across the metrological community so that a common understanding of counting is achieved.
- To prepare proposals for a clear delineation between the:
    - Kinds of quantity that can only be expressed as a count of a number of entities;
    - Kinds of quantity where counting is involved in the measurement process, but results are not expressed as a count of a number of entities;
    - Kinds of quantity not involving counting in the measurement process but where the results are expressed as a count of a number of entities.
- To give guidance for:
    - A clearer nomenclature for counted quantities;
    - A better metrological understanding of counts and their traceability;
    - Where the responsibility for providing traceability for such quantities lies.

The workshop took place online from 12.00–14.00 UTC on 28–30 March 2023. Each day's session concentrated on a different theme. The first session involved an introduction to the event and presentations of theoretical aspects of counting and the unit one. The second session concentrated on case studies of counting entities from electrical, mass, chemical and biological metrology. The final session considered other processes in metrology and how they related to counting and then finished with some concluding remarks and proposed workshop outcomes. The three sessions were attended by a total of 314 participants. See [5] for the workshop background, agenda, and presentation slides, which should be studied in advance for the reader to obtain the maximum benefit from this report.

2. Summaries of the Presentations

2.1. Day 1

2.1.1. "Welcome and Background to the Workshop" Pavel Neyezhmakov (NSC-IM, Ukraine)

The introduction to the workshop highlighted that whilst counting and number quantities are a common consideration within the metrology community, particularly within chemistry and biology, no consensus on how these quantities should be defined, described, and discussed had been achieved to date. Some propose that number quantities can be represented by a positive integer and a specification of the entities being counted. Others contend that, because a full characterisation of what is being counted is required, these quantities are not traceable to the SI. Hence, the aim of holding the workshop would be to achieve a common understanding of number quantities, counting and related processes. This should lead to clearer delineation between counting as a measurement process and number quantities as a measurement output, and also a clearer standardised vocabulary for these topics. As a further output, the metrology community should have a clear strategy and understanding of how traceability to number quantities can be achieved.

2.1.2. "What Questions Is the Workshop Addressing?" Bernd Güttler (PTB, Germany)

This presentation introduced the topics that the workshop would be considering.

Which quantities can be counted? Quantities such as amount of substance, electrical current, mass, or luminous intensity are based on quantized entities, processes or other phenomena that can, at least in principle, be measured by counting. This contrasts with quantities which are, to the best of our current knowledge, of a continuous (or analogue) nature such as length and time.

Why do we count? Quantification based on counting either of single entities (e.g., elementary entities such as atoms, molecules, and electrons) or quantized processes and other phenomena are of increasing importance in metrology because of the ever-increasing need for more precise measurements. This may be related to technological disciplines such as quantum technologies or to environmental and health-related measurements at a level of uncertainty or analyte detection sensitivity that makes the consideration of the quantized nature of the quantity unavoidable.

What is counting? Definitions of counting are not that common. A description is given in the Encyclopedia Britannica [6]: "In a collection (or set) of objects (or elements), the act of determining the number of objects present is called counting". This explanation leaves the 'act' of counting undefined and does not cover countable processes and phenomena such as the oscillation of a pendulum or radioactive decay. A comprehensive assessment of counting is missing. Hence, counting is used in different ways in the scientific literature. Even in the literature explaining the SI system [7], there is no consistent explanation of counting. It is not clear what can be counted or if the results of "the determination of the number of objects present" should be expressed with an SI base unit or as a count with the unit one, symbol 1.

How do we count? The term "counting" was used, for example, in conjunction with the redetermination of the Avogadro constant. A prerequisite for the redefinition of the SI units in 2019 was the redetermination of the Avogadro constant with an accuracy that was sufficient for the intended use. This was achieved by determination of the number of ^{28}Si atoms in a ^{28}Si single crystal sphere using, among other methods, X-ray diffraction and interferometry for quantities on a microscopic and macroscopic scale. "The determination of the number of objects present" (i.e., ^{28}Si atoms in the sphere in this case) was achieved with an uncertainty "that allows a redefinition of the mole in terms of the explicit number of elementary entities" (the Avogadro number) [8]. This "act of determining the number of objects present" (the so-called Avogadro or XRCD experiment [9]) has often been considered as counting by those that were active in the field (see, e.g., [9–11]). Similarly, biochemical (e.g., ddPCR, and flow cytometry), electrical current and luminous intensity measurements aim at "determining the number of objects present". Currently, it remains controversial which of these should or should not be considered as counting.

When the mole is used, the elementary entities must be specified (e.g., ^{28}Si atoms in case of the Avogadro experiment) [8]. Similarly, there is an unequivocal understanding in metrology that, when counting is used, it must be specified what is being counted. But it is also clear that this specification does not lead to identical objects/processes/phenomena being counted (e.g., electrons) in all cases or to a group of objects/processes/phenomena with specified, identical properties (e.g., all isotopes of the same element) or a property that has a range of results within given limits (e.g., a size range in the case of particles). It is also not clear when this leads to a result that can be expressed using an SI base unit, or the unit one, or is not within the SI system at all.

Based on these considerations, speakers at the workshop were given the following questions to address when preparing their presentations:

- What are the quantities within your technical discipline that relate to counting?

- What are the technical challenges for measuring these quantities, for instance, identifying or defining what is being counted, dealing with very small numbers, and calculating uncertainty?
- Do you express your measurement results using the unit one or one of the seven SI base units? What are the reasons for this? What improvements could be made to clarify the status and traceability of the unit one within the SI?

2.1.3. "Concepts of Continuous Quantities & Countable Aggregates and Nomenclature" Charles Ehrlich (NIST, USA)

This presentation explored the conceptual distinctions and overlap between the concepts of 'measuring continuous quantities' and of 'counting aggregates of different kinds of entities', focusing on terminological aspects, predominantly in the International Vocabulary of Metrology (VIM) and SI Brochure. The related concepts of 'dimensionless quantities' and 'quantities of unit one' were also covered, and the idea of distinguishing between 'dimensionless quantities' and 'unitless quantities' was introduced, as was the idea of introducing a symbol "C" for count in 'dimension'. Recognizing the distinction between 'dimension', 'unit' and 'number' was also emphasized.

The SI Brochure says significantly more than the VIM about counting, but neither says a lot. The VIM implies that counting and measurement have something in common, and perhaps that counting is even a form of measurement. Neither the VIM nor the SI Brochure define 'counting'. The suggestion was made to develop a rigorous concept system for 'counting', in parallel to the VIM concept system for 'measurement', and to possibly add "Counting" as a new chapter in the VIM in the future, just as a new chapter on "Nominal properties" is being added now.

It was discussed whether the concept of 'countable aggregates of entities' should be considered 'quantities' in the same, more modern sense of the concept and term, as espoused by Maxwell in 1873 and elaborated by DeBoer in the mid-1990s. The several ways that people have interpreted what Maxwell said and intended were explored, including what is meant by the symbols in the famous equation $Q = \{Q\}\cdot[Q]$. The first symbol, "Q", denotes what Maxwell first calls a "quantity", and which the VIM defines as an attribute or property. Depending on native language, some people consider 'quantity' to be 'amount of a property', distinguishing the concept 'property' from the 'amount' of it. The symbol "$[Q]$" denotes a measurement unit, such as 'metre' for the general quantity corresponding to the property "(amount of) separation between two points in space". The symbol "$\{Q\}$" denotes what Maxwell terms the "numerical value", which for length provides an amount of physical separation with respect to the measurement unit 'meter'.

Defining the measurement unit 'meter' is of course a matter of convention. If we wanted to communicate with galactic neighbours about lengths of things, we would need to describe to them how we construct a 'meter' based on properties of nature, such as light and caesium atoms. The same is not true for counting. It is not necessary to arbitrarily ascribe a (measurement) unit for counting, such as is necessary for length with the 'meter', since numbers alone suffice for communicating number of aggregates of entities. Of course, what the entities are also needs to be communicated.

From a metrological perspective, this is the key conceptual difference between 'continuous quantities' and 'countable aggregates'. The question is then whether the concept of 'counting unit' is necessary and, if so, should it be the number 1, or the entity being counted or even something else?

2.1.4. "Quantities with the Unit One" Peter Blattner (METAS, Switzerland)

This presentation considered the concept of the unit one, giving an historical background. The treatment of quantities with the unit one in successive editions of the SI Brochure was considered, with various terms being used such as ratio of two comparable quantities and dimensionless quantities. This treatment had changed over the years, highlighting perhaps that there had been a change in thinking over time and that there was no one single truth, but instead an agreed convention. It was not until the seventh edition of the SI brochure that the concept of 'counts' was presented.

The definition of unit and quantity in the present version of the VIM [12] was considered. If a quantity could be expressed as a number and a reference, then what was the reference for a number of entities—the unit one, or a reference relating to that which is being enumerated? For ratio quantities, there was the possibility of highlighting the units explicitly to give some indication of the quantity being described. This was not possible for quantities related to number of entities. It was noted that on many of these topics the SI Brochure and the VIM were not fully harmonised, but that there were already several joint efforts within the CCU and within the Joint Committee for Guides in Metrology (JCGM) working group on the VIM (JCGM-WG2:VIM) aimed at improving this situation. Finally, metrological concepts were tested with respect to quantities with the unit one. First, calibration was considered. The VIM refers to calibration requiring a measurement standard—a concept that is not clear for a number of entities. Similarly, for metrological traceability, the VIM refers to a reference and again this concept is not clear for a number of entities, and most probably method-defined. This is not different to the calibration of ratio quantities such as a power ratio where it is only 'characterisation' that is required (for instance, ensuing the linearity of the instrument). This raised some questions about what the role of NMIs was for quantities with the unit one if only this 'characterisation' was required. The presentation concluded by addressing the dimensional analysis issue related to quantities with the unit one, proposing that, currently, this concept adds little value, and is potentially troublesome as it may seem to allow the addition of two dissimilar quantities (e.g., photon flux and electron flux). It was proposed that the solutions to these issues were via the harmonisation of definitions and their consistent usage in the authoritative literature, NMIs offering the lowest possible uncertainties enabling the validation of realisations by customers, and for all stakeholders to avoid misunderstanding by providing a full description of the quantities under consideration. It was remarked that many of these requirements were no different to what we would expect from any traditional SI quantity, but they need reiterating in the context of quantities with the unit one.

2.1.5. "Counting and Why It Is Different from Amount of Substance" Richard Brown (NPL, UK)

This presentation considered number quantities and why these are different from amount of substance, the quantity for which the base unit is the mole. Whilst counting as a measurement process has been known for many thousands of years, it is only recently that it has been considered seriously by the metrology community as a measurement process. It was proposed that one of the main reasons for this was the confusion in this area between measurement processes (of which counting is an example) and measurement results (of which the expression of a number quantity is an example). As an analogy, whilst interferometry is a measurement process by which a length is determined, we do not call length an interferometric quantity. This distinction had been highlighted recently by the 2019 redefinition of the mole that now related amount of substance explicitly to a number of entities, with the Avogadro constant as the constant of proportionality between these two quantities. It was shown that the added complication with number quantities was the requirement to state sufficiently what was being enumerated—the problem of identity. In this way, the generic 'number' or 'amount of substance' mean less than 'length'. We must state 'amount of nickel' or 'number of fish'. Even then, 'number of fish' requires elaboration in a way that 'amount of nickel' does not. Are these fish sad, old, red, or possessing any other properties? Before we count 'some things' we must decide what counts as 'something'. The standardisation of such characteristics is likely to make these quantities method-defined. This highlights the distinction between amount of substance and other number quantities.

The 14th General Conference on Weights and Measures adopted the mole as the seventh base unit of the SI in 1971. This not only brought chemistry formally within the

SI, but also gave amount of substance its own dimension, and provided a clear basis for distinguishing between amount of substance and number. The definition of the mole was clear: the key parts being that it relates to 'elementary entities' that are 'of a system'. The consideration of only 'elementary entities' (atoms, molecules, ions, electrons, etc.) means that there is no identity consideration: nickel atoms are all identical, give or take isotopes of nickel which can in any case be uniquely specified. The term 'of a system' requires the elementary entities considered to be located in close enough vicinity that they could in theory react together stoichiometrically. Hence, the mole is used when it is useful and meaningful to consider elementary entities together in a system. Useful means that expression in amount of substance terms provides information or context to another property that is of interest. There is nothing to stop expression using 'number of atoms/molecules/etc.' but this loses the benefits of the dimensionality of amount of substance. The reverse is, however, not true: numbers of non-elementary entities (such as apples, planets, cells, etc.) cannot be expressed in amount of substance terms.

2.1.6. Summary of the discussions on Day 1

The main discussion points from the first session considered the relationship between the mole and a number of entities, and importantly, the distinction between counting as a measurement process and quantities that represent a number of entities. The mole was identified as a special case where the number of entities that could be considered was a special sub-set of things that could be counted, i.e., just elementary entities. In many cases, the user community wants to know about the 'number of entities' rather than the amount. There was also some support for 'pseudo units' (using the name of the entity being counted as a unit) that were placeholders for the unit one, and how these could be useful to standardise and use downstream of the SI. Conversely, the use of '1' in these cases could look strange and be confusing. The identity aspect of counting was also raised, and how specific we need to be when identifying what is being counted. This naturally led to requirements for documentary standardisation and often meant that number quantities were considered to be method-defined measurands. Nonetheless, these number quantities were considered as neutral quantities within the SI, and so being method-defined did not affect the coherence of the measurement result. There was some discussion about the mole becoming less useful than other units as chemistry moved into biology, perhaps when the molecules become so large that they are not identical, or when other properties such as mass or biological activity become more relevant. Consideration of the dimensions of quantities with the unit one was problematic since there was a danger of incomparable quantities being mistakenly compared, unless there was a clear description of the quantity being described. There was a reminder that whilst common sense and the context of the quantity being described may solve many of these measurement problems when there is interaction between humans, these issues still cause problems for computers. For that, there is no easy solution and there will not be one until computers are able to interpret fully quantity descriptions.

2.2. Day 2

2.2.1. "Counting Electrons for Metrology of Electrical Currents" Hans Werner Schumacher (PTB, Germany)

This presentation was about counting electrons for electrical current measurements. The ampere was defined by fixing the numerical value of the elementary charge, e. Since all electrons carry the same charge, 'counting these' can lead to the measurement of a current, and indeed the *mise en pratique* mentions that the ampere can be realised by using a single-electron-transport device. Single-electron pumps do not count electrons directly but instead pump electrons repetitively with a pumping frequency. This frequency must be high enough to give a measurable current and with a low enough uncertainty such that it is better than the old realisation of the ampere (around 3 parts in 10^7). Semiconductor single-electron pumps provide the best candidate technology using GaAs/AlGaAs quantum dots [13]. Electron tunnelling is stochastic and errors can arise from backwards tunnelling, or two or more electrons tunnelling at once [14]. These errors need to be controlled as far as possible and, in any event, 'counted' as well. A smaller quantum dot leads to a larger energy level separation, a larger ratio of tunnelling probabilities and hence a better pump because of a lower error rate. Experimentally, these devices have now been demonstrated with an uncertainty of 1.6 parts in 10^7. This demonstrates the verification of single-electron pumps with better accuracy than ampere realisation in the old SI [15].

Counting single electrons on these devices allows the quantification of errors and pumping statistics (i.e., pumping no electrons, one electron, two electrons, etc.) as a function of voltage [16]. When operated under optimum conditions, it has been demonstrated that no error was observed after 10^6 pumping cycles. As the frequency of pumping increases (perhaps up to 10^8 electrons per second), it becomes easier to count the errors (perhaps 10^3–10^4 per second) than it is to count the electrons. A method for in situ error detection was presented using in-series electron pumps based on signal correlation across these pumps. This provides the counting statistics for errors and therefore an uncertainty in the current. These are the first steps in producing a self-referencing quantum current source [17]. The current generated is only a few attoamperes. Ultimately, this technology will result in much lower uncertainties for primary realisations of low currents below 100 pA, demonstrating that counting electrons is a key tool in low-current metrology.

2.2.2. "Counting Si Atoms in a Silicon Sphere" Olaf Rienitz and Axel Pramann (PTB, Germany)

This presentation reviewed the process of 'counting' atoms in a silicon sphere as a route to the redefinition of the mole and the kilogram in 2019—the 'Avogadro project'—using the X-ray crystal density method [18]. There were four key measurements in this experiment: first, the volume of the macroscopic silicon sphere (V); second, the volume of the unit cell, containing 8 Si atoms, (a^3); third, the mass of the silicon sphere (m); and fourth, the molar mass of silicon in the sphere (M(Si)). The value of the Avogadro constant is then given by $N_\mathrm{A} = \left(8V/a^3\right)(M(\mathrm{Si})/m)$. The determination of the molar mass of silicon was a limiting factor in the uncertainty of the overall determination. Silicon has three natural isotopes and whilst the silicon sphere was very highly enriched in ^{28}Si, the other isotopes still had to be measured accurately, requiring measurements over at least six orders of magnitude, from ^{28}Si ($\geq$99.99% abundant) to ^{30}Si ($\leq$0.00004% abundant) [19]. This needed a new analytical approach, called 'virtual element isotope dilution mass spectrometry' [20]. In effect, this meant measuring just the ^{29}Si and ^{30}Si isotopes and assuming the remaining silicon was ^{28}Si in order to determine the overall molar mass. This was performed with a relative uncertainty of $\leq 5 \times 10^{-9}$ [21], unparalleled in analytical chemistry, with this uncertainty being validated via the CCQM-P160 comparison [22]. Ultimately, this work was key in the revision of the mole and the kilogram. The talk concluded by considering the quantities being considered during these experimental processes. Many of these were ratio quantities with the unit one (mass fraction, isotope ratio, amount fraction, and intensity ratio), and it was observed that elemental analysis and isotope ratio determination did not require special quantities or units related to counting, although subsequently, it was discussed that in fact 'number of atoms' and other atomic properties such as 'volume per atom' were key input quantities in the measurement equation.

2.2.3. "Quantification of Nucleic Acids by Counting" Inchul Yang (KRISS, Republic of Korea)

This presentation discussed the quantification of nucleic acids by counting. There were many biological entities that could be counted, some with the naked eye, some needing amplification by size to count them with microscopy, and some needing amplification by number in order to count them at all, for instance, bacteria or DNA. It was remarked that in biology, quantification by number was more meaningful than using mass or amount descriptions. Quantification requires, first, separation and partition of individual entities and, second, making these detectable. These processes are hampered by sampling issues, volume issues, detection failure, and the presence of impurities. (This also introduced the discussion about what counted as 'the measurand' with the example given of whether a whole apple, half an apple or a mouldy apple counts as an apple). The process of digital Polymerase Chain Reaction (PCR) was outlined. This required several steps: introducing the PCR mixture, partitioning, amplification, fluorescence detection, data analysis and final interpretation to give a result as 'copies' per volume. The advantages of digital PCR were its high sensitivity and resolution (potentially down to single-molecule levels), high tolerance to inhibitors and background nucleic acids, absolute routes to quantification with no calibration needed, high throughput, and the ability to quantify multiple DNA targets in one analysis. This meant that digital PCR had many applications including the quantification of DNA, the certification of nucleic acid reference materials, the detection of pathogens, gene ratios (for instance, in genetically modified organisms), and characterisation of DNA linkage and breakage. So far, there have been seven international comparisons undertaken by CCQM on digital PCR in several application areas, the measurand in most cases being 'copy/μL'. There had also been several reference materials certified using digital PCR. The presentation concluded that counting was a simple and direct approach for the realisation of a traceable measurement of biological entities (specifically cells, viruses, nucleic acids, and proteins). Digital PCR was a key tool in making these measurements because of the number of advantages it has in sensitivity, reproducibility, throughput and broad applicability. It also has the potential to be a primary method for the quantification of nucleic acids. Finally, open questions were posed about how non-SI terms such a 'copies' should be treated in the literature (both in CCQM comparison reports, associated publications and the literature in stakeholder communities) and what recommendations there should be for the expression of these measurands, some options being 'copies/mL', '1/mL', '/mL', 'mL^{-1}', etc.

2.2.4. "Counting Cells" Jonathan Campbell (LGC, UK)

The presentation began by discussing biological complexity. There were some classical counting methods for prokaryotes to enumerate viable cells involving amplifying their number using nutrient broth and then counting them as 'colony-forming units'. The situation with eukaryotes is more complex since there is a requirement then to determine what constitutes a cell (perhaps having a continuous cell membrane and a nucleus as two defining factors). The complexity of characterisation was highlighted, made more difficult because many of these properties were continuous rather than discrete, making classification more difficult. First, it was necessary to delineate the cells from the medium and any impurities present. Second, there was the challenge of identification (both in terms of biomarkers and morphology). Finally, there was the requirement to determine cell viability. All these characteristics had to be assessed before any counting began. There were many methods that were usable for cell counting. Those that counted directly (those interacting with individual cells, for instance, manual counting or automated microscopy) and those that counted indirectly (those looking at populations of cells).

The measurands of interest were then discussed. These were the total cell count and the differential cell count (a subset of the cells of interest), expressed as the 'number of cells' with the unit 1. There were also several derived quantities such as cell concentration and cell area density expressed as 'number of cells per volume' and 'number of cells per area', respectively. Cell fractions could be expressed as 'number of cells of interest' divided by 'total number of cells'. There were also other quantities, especially for prokaryotes such as the 'number of colony-forming units per volume' and 'number of plaque-forming units per volume'. The importance of the practical handling of the material to enable cell counting was also presented. This included considerations about diluting the material to present an optimal concentration to the measurement device and aspects such as avoiding cell damage and ensuring homogeneity were also important. Manual cell counting techniques required high operator skill since there were subjective decisions involved. Automated image microscopy uses automated algorithms allowing cell differentiation by factors which would not be possible simply by observation. The development of robust machine learning algorithms was an important aspect of this technology, but this still required operator intervention to decide what 'counted' as a cell. This emphasized the importance of defining the measurand in cell counting and the role of examination in nominal property determination (such as cell viability). The talk concluded by posing several outstanding questions in cell analysis about how to count entities that are fundamentally different, understanding the measurand for cell counting, the assignment of values to cells and cell properties, and the role of examination (both human and digital) in cell analysis.

2.2.5. "Counting Particles in Air" Konstantina Vasilatou (METAS, Switzerland)

This presentation demonstrated that the number of airborne particles is important in a growing number of applications, e.g., vehicle/aircraft emission control, bioaerosol monitoring, aerosol health or climate-related studies and product manufacturing in clean rooms. Measurements are reported in terms of particle number concentration (PNC), typically expressed as cm^{-3} or m^{-3} (and sometimes also $1/cm^3$, $1/m^3$, etc.). The notation $\#/cm^3$ is also used, though not so commonly and is not accepted by certain journals in the field. Limit values for PNCs emitted by vehicles have been legislated in the EU and there are currently discussions on whether to include target values for the PN concentration of ultrafine particles in the European Ambient Air Quality Directive as well. Instruments measuring PNC are roughly split into two categories: those that measure total particle number concentration, such as optical and condensation particle counters, and those which can additionally identify different particle types, such as bioaerosol monitors coupled to machine learning. Generally, these instruments work by 'counting' individual particles.

Traceability for total PNC has been established in the range 0.1 cm^{-3} to about 60,000 cm^{-3} through various international intercomparisons, with expanded (95% confidence interval) measurement uncertainties down to about 2% [23]. At PNCs more than 1000 cm^{-3}, condensation particle counters are calibrated against a Faraday-Cup Aerosol Electrometer, while at lower concentrations, custom-made optical particle counters or the Inkjet Aerosol Generator [24,25] are used for calibration. Optical and aerodynamic particle counters and size spectrometers can be traceably calibrated against custom-made optical particle counters or the Inkjet Aerosol Generator in the particle size range 100 nm–15 µm [26,27]. National Metrology Institutes are currently working on extending the traceability range to higher PNCs, relevant for vehicle and aircraft emission control. The new generation of particle counters, which are coupled to analytical methods such as laser-induced fluorescence, holography/microscopy and machine learning, open new possibilities in the field of aerosol sciences but are currently lacking traceability. These instruments are primarily developed for pollen monitoring, i.e., for particle sizes in the range above 15 µm for which there exist no primary standards as yet. Another challenge is the training of machine learning algorithms which is currently limited to a small, but growing, number of pollen taxa. To achieve full SI traceability, i.e., to measure number concentration of different particle classes, international collaborations between aerosol and data scientists would be highly desirable.

2.2.6. Summary of the Discussions on Day 2

The discussion began by considering that the silicon sphere experiment was using atomic quantities, for instance, 'volume per atom' and because 'atom' was omitted from the unit expression, there was a danger of this aspect being forgotten. In order to be consistent with the SI Brochure, there is a necessity to only use SI units, and this restricts the use of the unit one for these number quantities. However, it is necessary to have a description of what is being counted (for example, for a number concentration). The option preferred by the SI is to describe this fully in the quantity rather than introduce new 'units'. Another option is to use 'pseudo-units' to indicate what is being expressed, for instance, 'cp/mL' for copies per millilitre. It was highlighted that the stakeholder community uses many of these 'pseudo-units' in a varied and unstandardised manner. It was clear in these situations that there must be clarity and agreement on what 'pseudo-units' are being used and why. It was observed that this might be easier in some areas of chemistry and biology where there was, conceivably, a much smaller, established set of entity names to be standardised. There was further discussion about quantifying the uncertainty of quantities expressing a number of entities, some of this coming from uncertainty in the definition of the measurand itself and some from the measurement procedure.

There was also questioning on what needed to happen to make the realisation of small currents with single-electron pumps as good as that using the Josephson effect and the Quantum Hall effect. This depended on the current range. At the very low current range below 100 pA, the pumps were probably already better, but at higher frequencies, the pumps degrade very quickly which makes operation at high current difficult. Operation of pumps in parallel together with cryogenic current comparators might be a route to achieve higher currents reliably.

The realisation of the kilogram using the silicon sphere was discussed in terms of surface structure and surface contamination. It was important to clean the surface as much as possible, fully characterise it, and make the measurement in vacuum. Because these issues were better understood, and cleaning was more reproducible (and not operator-dependent) for silicon spheres than for platinum–iridium artefacts, these issues were tractable.

The issue of measurand identification was also raised for particle counting in air since particles had different shapes, compositions and fluorescence properties, and currently the measurement instruments were not able to distinguish between these, and simply measured based on average optical or aerodynamic diameter.

2.3. Day 3

2.3.1. "The Metrology of Quantities Which Can Be Counted in Radionuclide Metrology" Ryan Fitzgerald (NIST, USA)

This presentation covered the prevalence, philosophy, and practicality of counting in radionuclide metrology. The SI derived unit becquerel (Bq) is equivalent to (1/s). Measurands in radionuclide metrology include the activity of a radionuclide (expressed in Bq), massic activity of a radionuclide in a sample, emission probabilities, and detection efficiency. Measurement methods often involve counting. Probabilities and efficiencies require explicit definitions as to avoid ambiguity.

There is a fundamental connection between activity measurements and amount of substance measurements as, for a given radionuclide in a sample and a point in time, the activity (A) is related to the number of atoms (N) by the equation, $A = N\lambda$. An application of this relation is the measurement of the decay constant (λ) of a radionuclide by measuring the amount and activity in the same sample. Interestingly, the amount measurement may be based on mass spectrometry, itself a counting method.

One concern regards the reporting of results incorrectly using Hz instead of 1/s as a unit for non-periodic count rates. Another issue is conflating the definition of a measurand with the unit of measurement, for example, using "counts per second (cps)" or "decays per minute (dpm)" as units.

When measurands are particle emission probabilities or detection efficiencies, ambiguities may arise particularly about the denominator (normalization) of a quantity. Explicit definition of the measurement equation is helpful. Further ambiguities arise when probability values are reported as percentages with uncertainties also in percentage.

Technical challenges mentioned include detector pileup and dead time due to the stochastic nature of radioactive decay, spectroscopic interferences that cause ambiguity about which radionuclide decay is being counted. The need for coherence between metrology standard and practical usage was emphasized. The Consultative Committee for Ionizing Radiation (CCRI) works to educate the field on proper use of the SI.

2.3.2. "Measuring by Counting in Length Metrology" Alessandro Balsamo (INRIM, Italy)

The presentation began by discussing some examples of length measurement by counting. Interferometry was highlighted as an example where length as measured by 'counting' signal intensity periods. A second example is when counting is inferred, typically via the calibration of gauge blocks with a Fizeau interferometer. In this example, counting is not explicit since the interference order is carried out with no actual counting. A further example was the use of encoders in industrial metrology measuring positions via a periodic signal, very much as in interferometry. Here, 'cycles' are counted and the intra-cycle phase is measured. A final example is the counting of atomic spacing and steps in a Si lattice, used as a secondary realisation of the metre used in nanometrology.

The presentation then addressed an important distinction between 'counting' as a measurement process (of interference fringes, cycles of a signal, or atomic planes) and 'countable' as an attribute of a quantity (the number of people at this workshop, the number of molecules in a box, or the number of atoms in a Si sphere). Counting requires the separation of individual entities. This separation is made based on thresholds (of a signal level to prompt a count, of image contrast to count spots on an image, or between different people boarding an aircraft via visual inspection or turnstile gates). The threshold sensitivity may or may not be unity. When it is, the uncertainty of counting may approach zero. When it is not, counting may only be possible for clusters or aggregates. On the other hand, countable quantities are by nature independent of how they are measured. They have a finite cardinality that can be related one-to-one to natural numbers and are discrete by nature. Non-countable quantities have infinite cardinality (although the size of physical objects might be thought of as discrete because objects are made up of individual atoms and molecules). There is no definitional uncertainty associated with countable quantities as such, but there may be some associated with the definition of the entities that are counted, as mentioned in earlier presentations.

This thesis was neatly summarised by the four possible states arising from these two considerations. First, countable quantities measured by counting (e.g., the number of people who boarded an aircraft, counted at the entrance). Second, a non-countable quantity measured by counting (e.g., length measured interferometrically by counting the fringes of an interferogram). Third, a countable quantity not measured by counting (e.g., the number of nominally identical items in a set measured by weighing an individual item and then the complete set). Finally, a non-countable quantity not measured by counting, considered to be the 'usual case', and not specifically considered at this workshop. The presentation concluded by recommending that the distinction between counting, as a measurement process, and countable, as an attribute of a quantity, is made clearer in the future.

2.3.3. "The SI Second as a Count of Oscillations and Much More" Elizabeth Donley (NIST, USA)

The talk began by giving some background on the SI second. From 1960 and 1967, the second was defined as the fraction of the 1900 tropical year, but from 1967, a revision was adopted relating the second to the duration of a number of periods of the hyperfine splitting frequency of ^{133}Cs. This perhaps makes it sound like the second is realised by counting periods, whereas instead, it relates more to measuring frequencies. The 2019 rewording of the definition made it clearer that frequencies were important. The SI second is needed to realise the definition of all other SI base units apart from the mole. Currently, the second may be realised with an uncertainty of 1 part in 10^{16}, whereas optical frequency ratios may be compared with an uncertainty of 6 parts in 10^{18}. Atomic frequency standards use a resonant electromagnetic field to cause transitions between atomic energy levels as the 'standard's' oscillator frequency. The standard's oscillator frequency is the field frequency that causes transitions between 'clock' states. The optical clock network in Boulder used optical frequencies (which are no longer 'countable' even if in theory microwave frequencies were) for a variety of clocks based on different atoms. The rapid progress in optical clocks has been made possible by advances in the technology of optical frequency combs. Optical frequency standards now have an accuracy far in excess of microwave frequency standards, and this will likely prompt an update to the definition of the second in the near future. Optical frequency ratio measurements can already be made with uncertainties of several parts in 10^{18}. A final example was given where two independent, optical-to-electronic frequency dividers each produced 10 GHz microwaves with a phase exactly tracking its own Yb clock and yielding an absolute frequency instability of 1 part in 10^{18} in the electronic domain. Whilst the microwave frequency was only 10^{10} Hz, the fact that this could be measured to parts in 10^{18} was a clear example the measurement tracked phase and did not count cycles. This supported the conclusion of the presentation that this technical area does not count periods of radiation but instead measured frequency ratios through frequency synthesis.

2.3.4. "Candela—By Counting Photons?" Stefan Kück (PTB, Germany)

The presentation "Candela—by counting photons?" offered an overview of the SI unit candela and its measurement.

The candela, symbol cd, is the SI unit of luminous intensity in a given direction. It is defined by taking the fixed numerical value of the luminous efficacy of monochromatic radiation of frequency 540×10^{12} Hz, K_{cd}, to be 683 when expressed in the unit lm W^{-1}, which is equal to cd sr W^{-1}, or cd sr kg^{-1} m^{-2} s^{3}, where the kilogram, metre and second are defined in terms of h, c and $\Delta\nu_{\mathrm{Cs}}$. This means that the candela corresponds to a radiant intensity of 1/683 watt per steradian for monochromatic radiation of frequency 540×10^{12} hertz. Measured quantities in photometry must be considered as spectrally integrated quantities, where the integration is carried out over the product of the radiometric quantity and a luminous efficiency function. The most important of these functions is the photopic luminous efficiency function for the light-adapted eye, $V(\lambda)$, which is defined by the CIE over the wavelength range 360 nm to 830 nm at 1 nm intervals.

Expressing the candela numerically, a radiant intensity of 1/683 W/sr corresponds to $4.091942356\ldots \times 10^{15}$ photons/(sr s) at a frequency of 540×10^{12} Hz. Single-photon detectors, like Si single-photon avalanche diode (SPAD) detectors, can measure lower radiant intensities, e.g., $4.091942356\ldots \times 10^{6}$ (photons/(sr s), which corresponds to 1 nCd; however, traceability to classical radiometric methods is currently more accurate than to quantum-based approaches. Generating single photons is another promising method in the realm of photon techniques, utilizing sources like semiconductor quantum dots, single molecules, or colour centres in diamond. However, the accuracy of measurement is influenced by internal quantum efficiency and photon-collection efficiencies. It is important to emphasize that the candela is a unit for luminous intensity, so it must include the steradian, which is sometimes omitted in these considerations. Realizing the candela by counting or producing single photons is currently not as accurate as the classical method of using a cryogenic radiometer.

Despite limitations, single-photon sources find uses in quantum metrology, in particular quantum radiometry and sub-shot noise metrology. They offer sub-Poissonian photon statistics and exhibit the anti-bunching effect, which classical light sources or lasers cannot achieve. Single-photon sources are particularly valuable when paired with digital detectors like SPAD detectors.

The presentation also explored the relation between the candela and the mole. In principle, the mole can replace the number of photons, expressing the candela as $6.794830142\ldots \times 10^{-9}$ (mol/sr)/s at 540×10^{12} Hz. Notably, the mole is the unit of amount of substance, and photons sometimes behave like particles. In horticulture, photons and the mole are combined in units like PPFD (photosynthetic photon flux density) with (μmol/m^2)/s. However, merely knowing the number of photons is insufficient: understanding the spectrum of photons and the receiver's action spectrum is also essential.

To summarize, although counting photons is valuable in various applications, realizing the candela through photon counting is currently suboptimal. Emerging fields like horticulture lighting emphasize the significance of combining photons and the mole. The question of whether a number of photons can be described as amount of substance remains open for discussion.

2.3.5. Summary of the Discussions on Day 3

There was discussion about the 'threshold' required for something to be counted and whether this was a form of calibration, or simply a question of definition. To some extent, this depended on the phenomenon being considered. It was elaborated that in the case of the Si lattice, these 'thresholds' could easily be distinguished.

Further questions were asked about the difference between Hz and s^{-1}. This returned some of the conversation to the issues about pseudo-units, especially the use of 'cycle'. It was noted that ongoing work with the CCU Task Group on Angle and Dimensionless Quantities in the SI Brochure is examining some of these issues.

It was also observed that counting photons has a larger uncertainty than current methods of realising photometric quantities, and if these methods could be improved, then it might lead to a new definition of the candela.

2.3.6. "Concluding Remarks: How Should the Metrology Community Respond"

Sang-Ryoul Park (CCQM President, KRISS, Republic of Korea) and Joachim Ullrich (CCU President, PTB, Germany)

Sang-Ryoul Park considered the issues arising for CCQM. He reflected on the responsibility of metrology to be the guardian against mistakes or confusion in measurement. It was stated that advances in technology had opened up a much greater range of measurands that could be measured by counting or expressed as a number quantity. It was suggested that the CCQM ad hoc working group on the mole could lead the discussions of this topic and the implementation of any decisions within CCQM and that this could include all the relevant technical working groups of the CCQM. This effort would liaise closely with the CCU.

Joachim Ullrich presented some preliminary ideas about how to address the topics raised in the workshop. The first was to produce a report of the workshop with its main findings. Second, it was suggested that the existing CCU TG-ADQSIB examining angle and dimensionless quantities in general extends its scope and membership to address improving the language in the SI Brochure concerning counting and number quantities. Further, it was proposed that the CCU WG-CMT, which is concerned with the definition of core metrological terms, could also consider definitions relevant to the topics discussed in the workshop and feed these through to the JCGM-WG2:VIM.

Joachim Ullrich then gave some more general thoughts about the needs of digitalisation and the requirements for clear machine-to-machine communication. These considerations were relevant for the digitalisation of terms related to counting and number quantities. In particular, it was important to recognise that the metrology of quantities which can be counted perfectly fits in with the overall concepts of metrology since these quantities are just a subset of all quantities. Furthermore, the unit one is a necessary part of any unit system. It was reiterated that it was important to differentiate between counting as a measurement process, quantities that can be realised by counting, and quantities that express a number of entities.

This prompted some further discussion about the use of 'pseudo-units' or other terms to help describe the quantity being expressed, not least because this this would assist machines to understand the nature of the entities that were being counted.

3. Conclusions and Next Steps

Pavel Neyezhmakov brought the workshop to a close, stating that great progress had been made on the topics discussed. It was clear that there were several outcomes and proposals for next steps as a result of the discussion.

On communication, the following were proposed:

1. A clear distinction needed to be drawn between 'counting' as a measurement process and 'number of entities' as a quantity expression.

2. As only the unit one is available within the SI for describing quantities relating to a number of entities, it is essential that a clear description in words of the quantity being described was always used.
3. Standardising nomenclature for number quantities and some relevant 'pseudo-units' downstream of the SI would be beneficial in some cases, especially for digitalisation.

On traceability and ownership, the following were proposed:

4. Traceability for number quantities does not require an etalon but is established though appropriate validated measurement procedures (in the same way as for ratio quantities).
5. This lack of an etalon can cause problems with measurement hierarchy and the definition of the highest point of reference (because these quantities will often be method-defined).
6. NMIs/DIs should maintain traceability for important number quantities where the methods, as applied at the NMIs/DIs, are considered the highest points of reference within a calibration hierarchy. Many of these will be in chemistry and biology areas.

It was also remarked that further work remained to be carried out on understanding the calculation of uncertainty for number quantities, and also on distinguishing between the different challenges of defining and measuring these quantities in physical, chemical and biological disciplines.

Following the workshop, an e-mail was sent to workshop participants on 1 May 2023, on behalf of the Workshop Steering Committee, which outlined how these outcomes would be taken forward and the relevant actions for the metrology community.

"The recent CCU/CCQM Workshop on "The metrology of quantities which can be counted" was a great success with speakers discussing aspects of counting and number quantities within many different fields of metrology. The three sessions were followed by 314 participants. Some areas for clarification relating to the treatment of counting as a measurement process and the description of number quantities in the SI Brochure and in the VIM were identified.

As a follow up to the workshop, the existing CCU *Task Group on Angles and Dimensionless Quantities in the SI Brochure* will create a focus group including participants from the Consultative Committees which have identified specific issues to be addressed (e.g., CCQM, CCRI and CCPR). The focus group shall make a proposal to the CCU on how to clarify the text of the SI Brochure related to counting and number quantities. Once the proposed changes to the SI Brochure are agreed, the Consultative Committees can build on this in clarifying aspects related to counting in the *mises en pratique* for their respective fields. The treatment of counting and number quantities in the VIM will be taken forward separately by JCGM-WG2:VIM, many of whose representatives were present at the workshop".

Not mentioned in this e-mail, and a longer-term aim, is the production of an authoritative documentary standard that catalogues and defines certain 'pseudo-units' that may be of use in place of the unit one to indicate the entities being considered when expressing number quantities. This would likely concentrate on the biology and chemistry areas.

Author Contributions: Conceptualization, R.J.C.B., B.G., P.N., M.S. and R.I.W.; methodology, R.J.C.B., B.G., P.N., M.S. and R.I.W.; writing—original draft preparation, R.J.C.B., B.G., S.K. and K.V.; writing—review and editing, R.J.C.B., B.G., P.N., M.S., R.I.W., S.K. and K.V.; funding acquisition, R.J.C.B. All authors have read and agreed to the published version of the manuscript.

Funding: This work was funded by the UK Department for Science, Innovation and Technology under the National Measurement System.

Data Availability Statement: No new data were created or analyzed in this study. Data sharing is not applicable to this article.

Acknowledgments: The Workshop Steering Committee offers grateful thanks to the speakers at the workshop, not already listed as co-authors of this work: C. D. Ehrlich (NIST, USA), P. Blattner (METAS, Switzerland), H. W. Schumacher (PTB, Germany), O. Rienitz (PTB, Germany), I. Yang (KRISS, Republic of Korea), J. Campbell (LGC, UK), R. P. Fitzgerald (NIST, USA), A. Balsamo (INRIM, Italy), E. Donley (NIST, USA), S. R. Park (KRISS, Republic of Korea), J. Ullrich (PTB, Germany), and also to all other contributors at the workshop.

Conflicts of Interest: The authors declare no conflict of interest.

References

1. International Bureau of Weights and Measures. Resolution 12 of the 11th CGPM (1960). Available online: https://www.bipm.org/en/committees/cg/cgpm/11-1960/resolution-12 (accessed on 21 August 2023).
2. Quincey, P.; Brown, R.J.C. Implications of adopting plane angle as a base quantity in the SI. *Metrologia* **2016**, *53*, 998–1002. [CrossRef]
3. Güttler, B.; Bettin, H.; Brown, R.J.C.; Davis, R.S.; Mester, Z.; Milton, M.J.T.; Pramann, A.; Rienitz, O.; Vocke, R.D.; Wielgosz, R.I. Amount of substance and the mole in the SI. *Metrologia* **2019**, *56*, 044002. [CrossRef]
4. Brown, R.J.C. A metrological approach to quantities that are counted and the unit one. *Metrologia* **2021**, *58*, 035014. [CrossRef]
5. BIPM. CCU/CCQM Workshop on "The Metrology of Quantities Which Can Be Counted". Available online: https://www.bipm.org/en/committees/cc/ccu/wg/ccu-ccqm-ws/2023-03-28 (accessed on 21 August 2023).
6. Britannica. Arithmetic. Available online: www.britannica.com/science/arithmetic (accessed on 21 August 2023).
7. International Bureau of Weights and Measures. *The International System of Units (SI)*, 9th ed.; BIPM: Sèvres, France, 2019; Available online: https://www.bipm.org/en/publications/si-brochure (accessed on 21 August 2023).
8. Marquardt, R.; Meija, J.; Mester, Z.; Towns, M.; Weir, R.; Davis, R.; Stohner, J. Definition of the mole (IUPAC Recommendation 2017). *Pure Appl. Chem.* **2017**, *90*, 175–180. [CrossRef]
9. Azuma, Y.; Barat, P.; Bartl, G.; Bettin, H.; Borys, M.; Busch, I.; Cibik, L.; D'Agostino, G.; Fujii, K.; Fujimoto, H.; et al. Improved measurement results for the Avogadro constant using a 28Si-enriched crystal. *Metrologia* **2015**, *52*, 360–375. [CrossRef]
10. Fischer, J.; Ullrich, J. The new system of units. *Nat. Phys.* **2016**, *12*, 4–7. [CrossRef]
11. Massa, E.; Mana, G. Counting atoms. *Nat. Phys.* **2016**, *12*, 522. [CrossRef]
12. International Bureau of Weights and Measures. *JCGM 200:2012 International Vocabulary of Metrology–Basic and General Concepts and Associated Terms (VIM)*, 3rd ed.; BIPM: Sèvres, France, 2012. Available online: https://www.bipm.org/documents/20126/2071204/JCGM_200_2012.pdf (accessed on 21 August 2023).
13. Stein, F.; Drung, D.; Fricke, L.; Scherer, H.; Hohls, F.; Leicht, C.; Götz, M.; Krause, C.; Behr, R.; Pesel, E.; et al. Validation of a quantized-current source with 0.2 ppm uncertainty. *Appl. Phys. Lett.* **2015**, *107*, 103501. [CrossRef]
14. Kashcheyevs, V.; Kaestner, B. Universal decay cascade model for dynamic quantum dot initialization. *Phys. Rev. Lett.* **2010**, *104*, 186805. [CrossRef]
15. Stein, F.; Scherer, H.; Gerster, T.; Behr, R.; Götz, M.; Pesel, E.; Leicht, C.; Ubbelohde, N.; Weimann, T.; Pierz, K.; et al. Robustness of single-electron pumps at sub-ppm current accuracy level. *Metrologia* **2017**, *54*, S1–S8. [CrossRef]
16. Fricke, L.; Wulf, M.; Kaestner, B.; Kashcheyevs, V.; Timoshenko, J.; Nazarov, P.; Hohls, F.; Mirovsky, P.; Mackrodt, B.; Dolata, R.; et al. Counting statistics for electron capture in a dynamic quantum dot. *Phys. Rev. Lett.* **2013**, *110*, 126803. [CrossRef] [PubMed]
17. Fricke, L.; Wulf, M.; Kaestner, B.; Hohls, F.; Mirovsky, P.; Mackrodt, B.; Dolata, R.; Weimann, T.; Pierz, K.; Siegner, U.; et al. Self-referenced single-electron quantized current source. *Phys. Rev. Lett.* **2014**, *112*, 226803. [CrossRef]
18. Fujii, K.; Bettin, H.; Becker, P.; Massa, E.; Rienitz, O.; Pramann, A.; Nicolaus, A.; Kuramoto, N.; Busch, I.; Borys, M. Realization of the kilogram by the XRCD method. *Metrologia* **2016**, *53*, A19–A45. [CrossRef]
19. Pramann, A.; Rienitz, O. The molar mass of a new enriched silicon crystal: Maintaining the realization and dissemination of the kilogram and mole in the new SI. *Eur. Phys. J. Appl. Phys.* **2019**, *88*, 20904. [CrossRef]
20. Rienitz, O.; Pramann, A.; Schiel, D. Novel concept for the mass spectrometric determination of absolute isotopic abundances with improved measurement uncertainty: Part 1—Theoretical derivation and feasibility study. *Int. J. Mass Spectrom.* **2010**, *289*, 47–53. [CrossRef]
21. Pramann, A.; Lee, K.-S.; Noordmann, J.; Rienitz, O. Probing the homogeneity of the isotopic composition and molar mass of the 'Avogadro'-crystal. *Metrologia* **2015**, *52*, 800–810. [CrossRef]
22. Rienitz, O.; Pramann, A.; Vogl, J.; Lee, K.-S.; Yim, Y.-H.; Malinovskiy, D.; Hill, S.; Dunn, P.; Goenaga-Infante, H.; Ren, T.; et al. The comparability of the determination of the molar mass of silicon highly enriched in ^{28}Si: Results of the CCQM-P160 interlaboratory comparison and additional external measurements. *Metrologia* **2020**, *57*, 065028. [CrossRef]
23. Brown, A.S.; Quincey, P.; Ebert, V.; Nowak, A.; Tompkins, J.; Hessey, I.; Ciupek, K.; Schaefer, C.; Werhahn, O.; Vasilatou, K.; et al. International comparison CCQM-P189: Particle number concentration (100 to 20,000 cm^{-3}) and particle charge concentration (0.15 to 3 fC cm^{-3}). *Metrologia* **2023**, *60*, 08015. [CrossRef]
24. Horender, S.; Auderset, K.; Vasilatou, K. Facility for calibration of optical and condensation particle counters based on a turbulent aerosol mixing tube and a reference optical particle counter. *Rev. Sci. Instrum.* **2019**, *90*, 075111. [CrossRef]

25. Iida, K.; Sakurai, H.; Saito, K.; Ehara, K. Inkjet Aerosol Generator as Monodisperse Particle Number Standard. *Aerosol Sci. Technol.* **2014**, *48*, 789–802. [CrossRef]
26. Vasilatou, K.; Wälchli, C.; Koust, S.; Horender, S.; Iida, K.; Sakurai, H.; Schneider, F.; Spielvogel, J.; Wu, T.Y.; Auderset, K. Calibration of optical particle size spectrometers against a primary standard: Counting efficiency profile of the TSI Model 3330 OPS and Grimm 11-D monitor in the particle size range from 300 nm to 10 μm. *J. Aerosol Sci.* **2021**, *157*, 105818. [CrossRef]
27. Vasilatou, K.; Wälchli, C.; Iida, K.; Horender, S.; Tritscher, T.; Hammer, T.; Rissler, J.; Gaie-Levrel, F.; Auderset, K. Extending traceability in airborne particle size distribution measurements beyond 10 μm: Counting efficiency and unit-to-unit variability of four aerodynamic particle size spectrometers. *Aerosol Sci. Technol.* **2023**, *57*, 24–34. [CrossRef]