Resisting Chemical Atomism: Duhem's Argument

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Late nineteenth-century opponents of atomism questioned whether the evidence required any notion of an atom. In this spirit, Duhem developed an account of the import of chemical formulas that is clearly neutral on the atomic question rather than anti-atomistic. The argument is supplemented with specific inadequacies of atomic theories of chemical combination and considerably strengthened by the theory of chemical combination provided by thermodynamics. Despite possible counterevidence available at the time, which should have tempered some of Duhem's concluding remarks, there was no atomic theory of chemical combination, which is wholly a product of the twentieth century.

1. Introduction. Chemical combination is largely understood in terms of the notion of chemical bonding developed in the twentieth century, in which atoms play a central role. Popular presentations trace the introduction of atoms in modern chemistry back to Dalton's interpretation of the law of constant proportions at the beginning of the nineteenth century. Since chemical combination underlies the formation of compounds, and compounds were distinguished from solutions by the criterion provided by the law of constant proportions, it might be thought that Dalton's atomism contributed to the understanding of chemical combination. Nothing could be further from the truth, which was one of the factors underlying the widespread skepticism about atomism during the nineteenth century. Later, "Daltonian atomism" was abstracted from what Dalton actually said about the nature of atoms, and the notion of a molecule was introduced that his conception of the atom had precluded,[1] although without any alternative account of how atoms form molecules. As one historian puts it, "Chemists were almost all atomists" but adds

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1. This prevented him from accepting Guy-Lussac's law of combining volumes of gas; see Chalmers 2005, 12.

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that when pressed, they "recognised atomism as an optional extra" (Knight 1992, 120). A reason for this might well have been that they still had no idea how atoms could form molecules.

At the end of the century, Duhem ([1892] 2000) notoriously persisted in resisting atomism, arguing that the notion of an atom was not needed to understand the progress made during the nineteenth century as captured in chemical formulas. Some 10 years later he elaborated his case in a series of articles that he gathered into a book (Duhem [1902] 2002). Philosophers, who have had difficulty in accepting this as a scientifically respectable view, usually dismiss it as an expression of positivistic instrumentalism. But Duhem wasn't a positivist, and the details of his carefully formulated arguments call for more serious consideration. Unfortunately, he was prone to spoil meticulously developed arguments by rounding off with excessively rhetorical conclusions. The arguments are the main concern here. I begin by outlining his interpretation of chemical formulas and then consider the general strategy of his argument. The position is discussed in the light of some developments in chemistry, including one he overlooked counting against his view, and one lending some justification to his caution.

2. Duhem's Nonatomic Account of Chemical Formulas and Valency. In outline, Duhem's account of chemical formulas runs as follows. A basic notion of a compositional formula expresses the proportions of the elements in a compound in terms of chemical equivalents rather than mass. Thus, H_2O and H_2S are the compositional formulas of water and hydrogen sulfide, respectively, which have combining proportions of elements in the ratios of 1 : 8 and 1 : 16 by weight but in the same ratio of 2 : 1 when expressed in terms of equivalents. Element symbols in Duhem's formulas actually stand for what he calls "proportional numbers," assigned to preserve the analogy between compounds such as these in terms of which the one is said to contain as much oxygen as the other contains sulfur. These whole-number ratios are based on the law of definite proportions, said to have been established when Proust was considered to have won the debate against Berthollet early in the nineteenth century. Agreement on a universal system for compositional formulas was eventually established in 1860 after the work of Cannizzaro, whose method relied on Avogadro's principle, which could be interpreted macroscopically (equal volumes of gases at the same temperature and pressure contain the same number of equivalents).[2] By this time, the phenomenon of isomerism was

2. Although as Chalmers (2005, 14) emphasizes, other work in organic chemistry had done much to determine formulas independently of the considerations Cannizzaro brought to bear.

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well known, requiring compounds with the same compositional formula to be distinguished in terms of different structural formulas. Duhem provides an account of this in terms of substitution by radicals in simple archetypal chemical compounds dividing them into chemical types. Members of the water type, for example, are thought of as derived by substitution of one or more equivalents of hydrogen in water. Ethyl alcohol can be represented by the formula $(C_2H_5)HO$, and alcohols in general by formulas of the kind $(C_nH_{2n+1})HO$, whereas ethers can be regarded as resulting from the replacement of each of the two equivalents of hydrogen in water by alkyl groups of the kind C_nH_{2n+1}. Thus, dimethyl ether, although isomeric with alcohol and sharing the compositional formula C_2H_6O, can be represented by the formula $(CH_3)_2O$. Structure emerges with the identification of functional groups responsible for the compound's chemical reactivity, distinguishing the hydrogen in ethyl and hydroxyl groups and facilitating the prediction of chemical reactions and synthetic pathways to new compounds. Thus, Williamson predicted the possibility of asymmetric ethers, with different alkyl groups substituted for the hydrogens in water.

Further structure was elaborated by developing the notion of substitution to encompass the idea of replacing *several* equivalents of an element by a *single* equivalent of an element or group in order to account, for example, for dibasic acids. Thus, sulfuric acid is regarded as derived from two equivalents of water in which two equivalents of hydrogen are replaced by a single equivalent of the group SO_2, leaving two OH (hydroxyl) groups. Sulfuric acid can then be given the formula $(SO_2)(OH)_2$. Duhem speaks here of the dibasic acid belonging to the water type twice condensed, and the SO_2 group is said to be *bivalent* in sulfuric acid, as distinct from groups such as the ethyl group C_2H_5, which is *univalent* in ethyl alcohol since it substitutes for a single equivalent of hydrogen in water.

Compounds considered to be derived by substitution from one particular archetypal substance, and thus of that type, can equally well be considered to be derived from other archetypal substances and so belong to several other types. A choice of any one particular type would therefore be somewhat arbitrary, and Duhem understands the fully developed structural formula to simultaneously represent a compound as belonging to all the possible types to which it can be assigned. This has the effect of analyzing functional groups into their constituent elements and displaying the relation between the elements in the compound. Structural formulas are drawn by connecting elements that *exchange a valency* by a line. Duhem ([1902] 2002, 114) defines the general notion valency by considering

> any chemical type formed by the union of two elements or two groups of elements *a* and *b*. If it is the hydrochloric acid type, *a* will be Cl

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> and *b* will be H. If it is the water type, *a* will be O and *b* will be H_2; and so on. If the type corresponds to the hydrochloric acid type condensed *n* times, each of the two groups *a* and *b* is said to be *n*-valent in the compound *ab*, or, when united in forming the compound *ab*, *the two groups a and b exchange n valencies,* and the formula of the compound is written by drawing *n* lines between the two symbols *a* and *b*.

The group of *n* lines between *a* and H_n can then be written as *n* lines between *a* and a single H. The structural formula for ammonia, for example, is written with three lines between nitrogen and a distinct hydrogen, where "each line marks the place of one equivalent of chlorine replaced by substitution and indicates the equivalent of hydrogen that was united with it" (Duhem [1902] 2002, 115). Duhem's definition assigns a valency to an element in a particular context, allowing that "the number of valencies of an element can change according to whether the element is part of one compound or another. Iodine, univalent in hydroiodic acid, is trivalent in iodine chloride; nitrogen, trivalent in ammonia, is quintivalent in ammonium hydrochlorate; carbon, quadrivalent in methane and in carbonic acid, is bivalent in carbon monoxide" (Duhem [1892] 2000, 447). The valency of an element is not only relativized to a compound, however, but even to a functional group. Two equivalents of the same element may occur in the same compound "with different valency numbers. In ammonium nitrite, for example, the equivalent of nitrogen which comes from ammonia is quintivalent and that which comes from nitrous acid is trivalent" (Duhem [1892] 2000, 147–148).

Duhem thought of structural formulas as abstract topological structures rather than two-dimensional pictures of the arrangement of atoms in molecules. The phenomenon of optical isomerism called for a modification of this view and is not considered at all in the 1892 article, although the phenomenon was well known by then. Optical isomers display the same pattern of chemical reactivity and physical properties, such as melting and boiling points, but differ in their ability to rotate the plane of plane-polarized light in different directions and in the solid phase to form crystals that are the mirror image of one another. It must therefore be possible to preserve the structure representing chemical function in a suitable variant. To accommodate this kind of isomerism with the right number of isomers, Duhem elaborated his topological structures with a "new element taken from geometry" ([1902] 2002, 128), representing optical isomers as three-dimensional mirror images. But they were still abstract structures representing the properties of particular substances by particular structures of real numbers (the proportional numbers that the element symbols stand for). Viewing them as depicting molecules would require additional

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descriptions of the atoms in these putative molecules that would explain rotatory power—something the atomists couldn't supply.[3]

It should be borne in mind that Duhem held that positive contributions to the explanation of chemical phenomena were forthcoming from thermodynamics, which engaged him throughout the course of his career, much of it directly concerned with physical chemistry. In particular, thermodynamics offered for the first time in the history of chemistry a systematic theory of chemical combination in the sense of a theory governing the stability of compounds and a general theory of mixtures. Duhem's nonatomic stance was anything but a hollow shell.

3. The Structure of Duhem's Argument. The proponent of a negative ontological thesis must show, as Church (1958) required when supporting Quine's notion of ontological commitment, how to do without the entities in question. This is precisely what Duhem does. But further empirical facts might come to bear, and Duhem was clearly aware that only further considerations would decide the issue: "A foundation," he says, had "yet to be discovered" ([1902] 2002, 147). The account itself is neutral on the atomic question. Duhem certainly thought there were alternatives to the atomic conception, relying on ideas of matter as continuous, going back to Aristotle and the Stoics rather than Leucippus and Democritus, meriting further investigation.[4] He also thought there were further considerations counting against atomism, but these were raised independently of the first step in the argument.

What Duhem put into his account was also common to Daltonian atomism, or at least an abstracted form of it in which "the nature and number of atoms juxtaposed to form a molecule are characteristic of the properties of the compound." Referring to the laws of constant and multiple proportions as well as Mitscherlich's law of isomorphism, he goes on to say that "as Dalton showed, it is easy to deduce the fundamental laws of chemistry" (Duhem [1892] 2000, 441). Nevertheless, he clearly thought that Daltonian atomism provided no explanation of the laws that it entailed. Moreover, Duhem contends that the role that Dalton's atomic theory apparently played in his discovery of the law of multiple proportions cannot be considered a decisive argument in favor of the atomic theory. "Were this the case, it would be necessary that the interpretation of the law of multiple proportions provided by the atomic theory was not only a plausible and appealing interpretation, but also the only interpretation possible" (Duhem [1902] 2002, 147). Duhem's account is a more

3. It wasn't until well into the twentieth century that explanatory accounts of rotatory power based on molecular structure began to be articulated (Needham 2004).

4. For a discussion, see Needham 2002.

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abstract, less committed one than Dalton's. Parsimony should speak in favor of it unless something can be said about the nature of atoms that actually gives them some explanatory role and solves more problems than it raises. Abstracting from what Dalton actually said about the nature of atoms removed a source of problems. But was anything more illuminating forthcoming, improving on other interpretations?

The atomists of Duhem's time made play on the notion of valency. What was ostensibly a contribution to the understanding of chemical combination amounted to no more than reading features displayed in chemical formulas into their notion of an atom, however, and was not the source of independent theory that it promised to be (and which the modern reader might well read into it). Duhem's more circumspect development of the notion of valency allowed a little more subtlety, which better accommodated the variable valency of certain elements. To the extent that the atomists actually said anything about the nature of their atoms, they soon became embroiled in paradox. Some physicists were well aware of problems facing the classical conception of the atom, and Duhem was in other contexts particularly scathing of atomists who didn't allow this to worry them. "W. Thomson, for example, never asks himself philosophical questions such as . . . can one of the ultimate elements of matter occupy a variable volume or not? . . . Still less does he ask himself what one must understand by the volume occupied by an atom" (Duhem [1893] 1996, [57] 354). But when discussing chemistry, the additional considerations he brought to bear involved the specific inadequacies of what the atomists said about atoms in addressing chemical issues.

Duhem discusses the projection of valency onto atoms found in Wurtz's theory, where an element's valencies are simply called atomicities and attributed to each atom of the element. An atomic theory that was actually proposed, J. J. Thomson's vortex theory of atoms, Duhem found wanting (Duhem [1892] 2000, 447). Moreover, the fact that the valency of an element varies from one compound to another, which he accommodated on his own account, seemed to him a serious problem for atomism, which required a distinction between different kinds of atomicity for a single atom. Thus, nitrogen is sometimes trivalent, sometimes quintivalent, but "if the five atomicities were absolutely identical, reasons of symmetry would render absurd the existence of compounds such as ammonia where three of these atomicities would be satisfied while the other two would be free" (Duhem [1892] 2000, 448). Call atomicities of nitrogen fixed by a hydrogen atom in ammonia atomicities of the first order, and the further two exhibited in ammonium chloride atomicities of the second order. Now consider that in ethylamine, $N(C_2H_5)H_2$, the ethyl group C_2H_5 is fixed by a first-order atomicity of nitrogen. Reaction with hydroiodic acid generates a new substance, the hydroiodate of ethylamine $N(C_2H_5)H_3I$, in

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which the new elements are fixed by atomicities of the second order. A substance of the same composition can be obtained via an alternative route, however, by reacting ammonia with ethyl iodide. Since the hydrogen atoms of ammonia are fixed by atomicities of the first order, the ethyl group will be fixed by an atomicity of the second order. The reactions must then generate two distinct isomeric substances in which the atomicities fixing the ethyl group are different. But experiment shows they are one and the same substance. Duhem claims that many similar cases are to be found.[5]

He thought these considerations were decisive and concluded that atomic doctrines were "without a future" (Duhem [1902] 2002, 147). In the light of developments in the twentieth century, which have met Duhem's challenge to provide an account of the atom that can actually do explanatory work, this claim seems premature. Such retrospective criticism is not easy to assess, however. Quantum mechanics has substantially changed the notion of an atom that was the object of Duhem's criticism, saving it from the incoherences implied by classical physics, and it is far from clear that the atomism accepted today speaks for the view Duhem actually criticized. Much of the nineteenth-century chemistry that has been preserved is what Duhem captured in a theory formulation whose foundation had "yet to be discovered." Although the microworld of modern chemistry is semiclassical, the foundations of this amalgamation of classical and modern theory have yet to be fully articulated.

4. The Problem of Steric Hindrance. The experimental work on ammonium salts to which Duhem appeals in his argument against Wurtz's notion of atomicities was conducted by Victor Meyer in the 1870s (Thorpe 1911, 444–445). Meyer did other work, less conducive to Duhem's case, shortly after the publication of the 1892 article, which is not mentioned in the 1902 book. Meyer found that although benzoic acid reacted normally as an organic acid with methyl alcohol and hydrochloric acid to produce the corresponding ester, substitution on the benzene ring could radically affect the result. In particular, the symmetrically trisubstituted derivatives of benzoic acid, with the three hydrogens substituted by the methyl radical, halogens, the nitro or carboxylic acid group, yield no ester whatsoever. The same is true for disubstituted derivatives on positions adjacent to benzoic acid's carboxylic acid group. There is no intrinsic thermodynamic instability preventing the existence of the esters themselves, which can be derived from the silver salts of the respective substituted acids by reaction with methyl iodide. Moreover, the esterification

5. Werner subsequently distinguished between primary and secondary valence, the latter now understood as a coordinate bond.

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proceeds normally in compounds where the carboxylic acid group is separated from the substituted benzene ring by one or more carbons (Meyer [1894] 1952; Thorpe 1911, 474).

The remarkable difference in behavior can be explained as being of stereochemical origin, the substituent groups hindering the approach of the alkyl group on the alcohol where no ester is produced. Here the literally interpreted spatial structure of the molecular framework, as depicted by the atomistic interpretation of the corresponding structural formulas, is actually doing explanatory work. There is still no working account of the nature of the atoms in the molecule that would explain the origin of the molecular structure. But appeal to molecular shape offers an explanation whereas Duhem's abstract account offers none. This is the first evidence, as far as I am aware, that calls for a more concrete interpretation of chemical formulas than is ventured in Duhem's abstract account and definitely points in the direction of molecules.

5. Are Compounds Molecular? We shouldn't be too quick to generalize from this to a claim about the molecular constitution of all substances, however. Duhem ([1902] 2002, 134–135) describes the view he resisted as follows:

> All compound bodies are reducible to *molecules*. The molecules of a given compound are all identical with one another; each is formed from a certain number, necessarily whole, of atoms of each simple body which combine to bring about the formation of the compound. The chemical formula of a compound simply expresses which atoms and in what number they are united in a molecule of the compound. Thus, to say that the formula of hydrochloric acid is HCl is to say that the molecule of hydrochloric acid contains one atom of hydrogen and one atom of chlorine. To say that the formula of water is H_2O is to say the molecule of water contains two atoms of hydrogen and one atom of oxygen.

The naive idea that a macroscopic amount of the substance is just a collection of molecules may give a fair account of many organic compounds. But it is not generally true. It is not true of hydrochloric acid and water, whose microstructures must be understood in terms of the dissociation of molecules into ions and the intermolecular forces building complex units larger than single molecules. There are several reasons why the molecular paradigm is not universally applicable.

Molecules might be distinguished from complexes formed in water on the basis that the intermolecular forces are significantly weaker than intramolecular forces. But this criterion doesn't distinguish molecules in sodium chloride and silicon dioxide, which form indefinitely repeating arrays in

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the solid phase in which no particular pairs (NaCl) or triples (SiO_2) of atoms can be distinguished from their neighbors by the nature of the combining forces. A radical advance of another kind, challenging the molecular view, came within Duhem's lifetime. This questions the assumption of definite proportions forming the basis for the stoichiometric compositional formulas on which the structural formulas differentiating isomeric variations are built. This matter was supposedly settled at the turn of the nineteenth century. But Greenwood goes so far as to say that in his extended debate with Proust, Berthollet "brought much cogent evidence to support his contention that, for *solid* compounds, constancy of composition was by no means universal and variation in composition frequently occurred. His data were not refuted but gradually became ignored due to the upsurge of interest in the new organic chemistry" (Greenwood [1968, 5], who goes on to point out that the devices of chains and rings that save carbon's fixed valency of four fail in inorganic compounds). Toward the end of Duhem's life, Kurnakow highlighted the issue in an article on the foundations of the concepts of chemical combination and chemical substance. He emphasized the importance of Gibbs's concept of phase and credited Duhem with making the point in his 1902 book that the phase rule "makes no difference in principle between bodies of constant and variable composition" (Kurnakow 1914, 116).[6]

Kurnakow used what were new physico-chemical methods, unavailable to Berthollet, to investigate the phase diagrams of two-component mixtures, which trace how the effect of changing temperature on the solid and liquid phases of the system varies with composition (proportion of the elements). A solubility, or melting point, curve is traced out by connecting points corresponding to the temperature and composition at which solid appears as the temperature of the homogeneous liquid mixture falls. Of particular interest were cases where the solid that forms at points below the melting point curve was not one of the pure components but a solid solution. The following question arises: what distinguishes a solid solution from a genuine compound of the two component elements? If a definite compound forms at a definite composition, then mixtures of overall composition in the vicinity of this definite composition will in effect comprise a mixture of the compound with one of the elements as an impurity, which would have the effect of reducing the melting (freezing) point of the mixture compared to that of the pure compound. The melting point curve will therefore show a maximum at this point of definite composition. Other physical properties such as electrical conductivity, viscosity, hardness, and refractive index fall off like the melting point when a pure substance is

6. He refers to pp. 190–198 of Duhem ([1902] 2002), which ends on p. 187, however. The mistaken reference is carried over from the Russian original.

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mixed with other substances, and isotherms following these variations with composition at different fixed temperatures display systematically shifted values but always with a maximum at the same composition—that corresponding to the maximum in the melting point curve. These independent methods thus agree for many mixtures on distinguishing a particular composition that Kurnakow called the Daltonian point, defining the composition of a compound whose proportions are constant.

But these methods, applied to systems at equilibrium, brought to light new substances "which do not conform to this definition and call for a broadening of our concept of a chemical individual" (Kurnakow 1914, 123). A case in point is the so-called γ-phase of the binary thallium-bismuth system, whose composition varies from 55% to 64% bismuth (by relative atomic weight). Although the melting point curve displays a maximum at a composition of 62.8% bismuth, which would ordinarily be taken to indicate a definite compound, the isotherms for electrical conductivity display no singular point at the composition 62.8% point, and the same goes for other properties that vary with composition. This is an example of a compound of variable composition, confirming the view Berthollet defended some 100 years earlier in his dispute with Proust that the law of constant proportions is not generally true of compounds.

The composition of Berthollides varies with conditions and must typically be rendered with nonintegral subscripts, such as $Fe_{0.98}O$. There is no way of construing this as describing a molecule and certainly not as a substance comprising particles of a single kind. Microscopically, it is a crystal structure with some ferrous ions, Fe^{2+}, replaced by ferric ions, Fe^{3+}, and others replaced by holes in the crystal lattice. The stability of the structure is understood thermodynamically, in terms of a minimum in the free energy achieved with the additional energy required to convert ferrous to ferric ions by removal of an electron being offset by the increased entropy afforded by the holes in the crystal structure.

6. Conclusion. Duhem succeeded in giving an account of chemical formulas that consistently avoided ontological commitment to atoms and molecules. He was able to pose a problem for atomism about the nature of variable valency but failed to take account of the first indication of stereochemical microstructure. Although Meyer's work on steric hindrance had been published when Duhem (1902) 2002 appeared, Duhem might have considered it an isolated study. It certainly didn't bring any account of the nature of atoms to bear that could shed any light on how they might combine to form molecules and other microscopic constellations or explain how plane-polarized light is rotated in opposite directions by stereoisomers. These were only forthcoming well into the twentieth century. His unwillingness to draw too hasty conclusions about the significance of

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chemical formulas finds some reflection in the warnings inorganic chemists give much later in the twentieth century of generalizing from organic compounds to compounds in general. Chemical substances do not, in general, consist of collections of the same kind of molecule.

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